Ultrafast optical control of multiple coherent phonons in silicon carbide using a pulse shaping technique

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We have demonstrated ultrafast optical control of multiple coherent phonons in silicon carbide (SiC) using a pulse shaping technique combined with sub-10 fs optical pulses and a two-dimensional spatial light modulator. Since our technique produces a pulse train with desirable number of pulses and the time intervals, precise manipulation of the amplitudes of the zone-folded optical and acoustic phonon modes could be achieved simultaneously, which were well reproduced by a model calculation assuming the two modes are excited independently by each pulse. These results show that our scheme can provide the high controllability of multiple phonon modes in SiC.

Silicon carbide (SiC), one of wide bandgap materials, holds enormous promise for advances in high-power and broadband electronic devices operating under severe environment because of its superior characteristics of saturation velocity of electrons, breakdown field, and thermal conductivity[1–4] Owing to its substrate availability and compatibility for the stacking oxide layers compared with whose in other wide bandgap materials, various attempts for prototypical power electronic devices such as metal-oxide-semiconductor field-effect transistor (MOS-FET) have been examined.[5–9] To further improve the device properties at high temperatures, thermal management via the manipulation of lattice vibrations is desired,[10–12] and therefore study on the controllability of phonons is of great importance.

There are two typical ways to control lattice vibrations. One is the modification of the energy dispersion relation of phonons by fabricating micro-structures in a host material, which enables to control the propagation directions for particular modes of phonons.[13–15] The other is dynamical control of phonons by utilizing ultrashort optical pulses.[16,17] In this method, a number of optical pulses with an appropriate time interval are applied to a sample prior to the relaxation time of particular phonon modes. In the case of two pulses, for example, the first pulse induces phonon oscillations, and subsequently, the second pulse coherently controls the amplitude; constructive or destructive interference of the phonon oscillations takes place depending on the time interval. In general, materials have a lot of phonon modes with different frequencies. Therefore, the number of optical pulses greater than that of phonon modes with desirable time intervals is indispensable for simultaneous coherent manipulation of multiple phonon modes. In this context, an interferometric setup simply placed with multiple optical pulses is not suitable for the complicated pulse shaping of optical pulses. In contrast, a spatial light modulator(SLM) facilitates desirable pulse shaping such as number of pulses, time intervals between optical pulses, and so on,[18] which is applicable to the manipulation of multiple phonon modes.

SiC has different polytypes depending on the stacking along the c-axis.[19] Here, we used 4H-SiC single crystal stacked with a period of 4 layers in hexagonal crystal structure. The dispersion relation of phonons in 4H-SiC can be expressed as zone folding of the 3C-SiC band diagram, and several folded modes, two E-modes (E_2-FTA and E_2-FTO) and three A modes, appear at the frequency range of 5-30 THz observed by Raman spectroscopy and coherent phonon spectroscopy.[19,20,22] In the present work, the E_2-FTA and E_2-FTO modes in 4H-SiC were coherently controlled based on a pulse shaping technique with an anisotropic polarization apparatus.

Figure 1(a) shows a setup for transient transmission measurements based on the pump-probe method. The light source was a mode-locked Ti:Sapphire laser (ML Ti:S) with the average power of 0.2 W, the repetition rate of 80 MHz, and the pulse duration of 6 fs. The spectrum of the laser was ranging from 600 nm to 1050 nm as shown in Fig. 1(b). The fundamental beam negatively chirped by a set of chirped mirrors (CMs) was separated into pump and probe beams by a beam splitter, and the amplitude of pump beam was modulated at 40 kHz by a reflective optical chopper (RC).[21] The spectrum of the pump was dispersed by a grating then incident to a reflective two-dimensional spatial light modulator based on liquid crystal on silicon (2D LCOS-SLM) for pulse shaping. Using the lateral axis of the SLM, the spectral phase of the pump beam was compensated by considering second- and third-order dispersions. On the other hand, the orthogonal axis of the SLM was periodically modulated to tune the spectral intensity,[25] and the resultant zeroth-order diffraction was shined to the sample. As for the probe bean, optical delay was added by a translation stage, followed by a pair of CaF_2 wedge plate to compensate the chirp of the probe beam. The polarization of the probe was set to 45 degree against that of the pump by a half-wave plate, where the E-mode coherent phonons in SiC was effectively observed. The pump and probe beams were focused on the sample non-coaxially by using the same off-axis parabolic mirror. The horizontal and vertical polarization components of the transmitted probe beam were separated by a polarized beam splitter and then detected by a pair of photodiodes (PD1 and PD2). The difference of photocurrent was amplified by an transimpedance amplifier (TZ amp.) and the voltage signal synchronized with the 40 kHz modulation was detected by a lock-in amplifier.

In previous studies, a four-focusing configuration combined with a pair of gratings,[18,25] an ultrashort optical pulse yields a pulse train of multiple optical pulses with equally spaced intervals, and the resulting pulse train could achieve the coherent control of the specific phonon mode. Furthermore, the SLM is capable to compensate three or higher order dispersions, and

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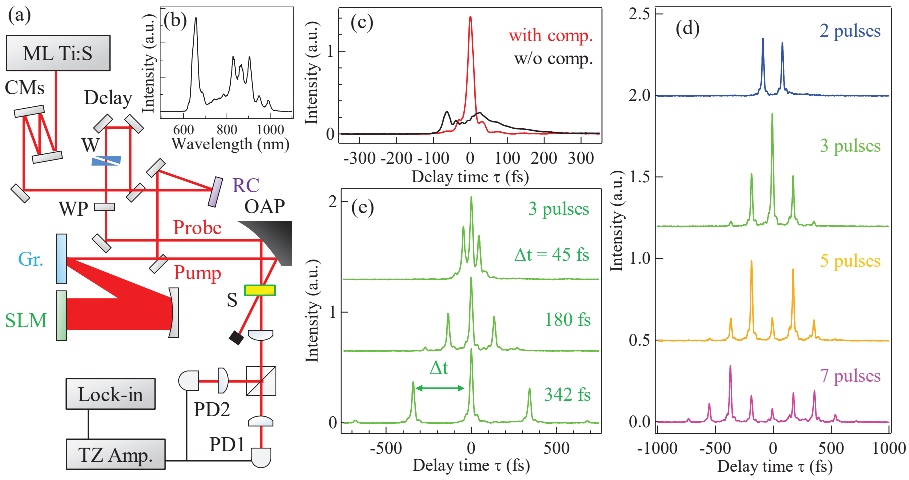

Fig. 1. (a) Setup for transient transmission measurements. Reflective two-dimensional spatial light modulator based on liquid crystal on silicon (LCOS-SLM) was used for pulse shaping. CMs: chirped mirrors, W: wedge plate, WP: half-wave plate, BS: beam splitter, RC: reflective optical chopper, G: grating, OAP: off-axis parabolic mirror, S: sample, PD: photodiode, PBS: polarized beam splitter. TZ amp.: transimpedance amplifier. (b) The spectrum of the laser output. (c) Cross-correlation signals with and without dispersion compensation of single laser pulses by CMs, W, and SLM. (d), (e) Results on the pulse shaping for the number of pump pulses and the interval(Δt) of the three pulses.

compression of sub-10 fs or few cycle pulses, generation of an ultrashort pulse train, precise timing control of optical pulses have been extensively demonstrated.[18,26–29]

To evaluate the temporal waveform of shaped optical pulses, a β-BaB_2O_4(BBO) crystal with the thickness of 40 μm was placed at the sample position. The cross-correlation signal between the pump and probe beams was recorded as the second harmonic (SH) intensity, as shown in Fig. 1(c). The red(black) curve shows a typical temporal waveform with(without) the dispersion compensation. The full width at half maximum (FWHM) of the compensated pulse was ~ 17 fs, which was slightly longer than the Fourier transform limit partly because of the group velocity mismatch in the 40 μm-thick BBO.

Next, we demonstrate how we can manipulate the waveform of ultrashort optical pulses using our pulse shaping technique. Figure 1(d) shows the controllability of the number of pulses. The single sub-10 fs optical pulse yields a pulse train of 2 pulses to 7 pulses with ~ 22 fs (FWHM) by adjusting the spectral phase of the incident optical pulse via the SLM, while the interval Δt between adjacent pulses was fixed at 180 fs. Note that intensities of pulses in the pulse train are not the same in case of more than three pulses because a sinusoidal phase modulation on the frequency domain were applied to the asymmetric spectrum of the laser.[30] The controllability of the time interval Δt is shown in Fig. 1(e), where the number of pulses in a pulse train was fixed at three. The value Δt could be systematically varied up to 342 fs, while keeping the intensity and the FWHM of each pulse. The results shown in Figs. 1(d) and 1(e) clearly demonstrate that we can produce a desirable phase-locked pulse train with different number of optical pulses having arbitrary time intervals, which is indispensable for controlling multiple coherent phonons of materials.

The result on the transient transmittance changes of 4H-SiC excited by a pulse train that consists of three pulses with different time intervals (Δt = 45 fs, 65 fs, 85 fs, 152 fs) is shown in Fig. 2(a). As a reference, the result excited by single pump pulses is also shown by a red curve at the bottom of Fig. 2(a). The origin of delay $\tau = 0$ is defined as the time when the third pulse of the train was overlapped with the probe. The positive delay time indicates that the pump pulse reaches the sample earlier than the probe one. The sharp peak was observed at $\tau = 0$ due to the overlap of the pump and probe pulses, followed by a fast decay of ~ 0.1 ps coming from excited carriers.[22–24] Subsequently, the oscillations due to high frequency phonons in 4H-SiC were observed. The Fourier transformed (FT) power spectra obtained from the time domain data from 1 ps to 5 ps are shown in Fig. 2(b). The result on the single pulse excitation shows two sharp peaks located at 6.1 THz and 23.3 THz, which are assigned to the E_2-FTA phonon and the E_2-FTO phonon, respectively.[19]

According to the phonon oscillations excited by a pulse train of three pulses, one can see that the FT intensity as well as the amplitude of the oscillations varies with Δt. In the case of the FTO phonon mode located at 23.3 THz, it intensively appears at Δt = 45 fs, while disappears at Δt = 65 fs. The same behavior is repeated after appropriate time intervals (see, Δt = 85 fs and Δt = 152 fs). In the case of the FTA phonon mode located at 6.1 THz, on the other hand, it solely emerges at Δt = 152 fs. Both the FTO and FTA modes almost disappear at Δt = 65 fs. These results clearly indicate that we could coherently control the two types of phonon modes in 4H-SiC by using our pulse shaping technique.

In order to evaluate the performance of the coherent control quantitatively, we measured transient transmission changes by precisely tuning the Δt from 25 fs to 270 fs with a step of 5 fs. The FT intensity of each phonon mode is shown in Figs. 3(a) and (b) as a function of Δt. As shown in Fig. 3(a), a periodic modulation with 60 fs is clearly observed, indicating the interference of the FTO phonons excited by a pulse train of three pulses. Similarly, the FTA mode shows a periodic modulation with 162 fs as shown in Fig. 3(b), whose period is longer than that of the FTO mode because of its lower phonon frequency. Assuming that a coherent phonon with a given frequency ω_{ph} is instantaneously excited by a single optical pulse, the temporal waveform $f(t)$ of the transient transmission change can be written as $f(t) = A{*}\exp(-\gamma t)\sin(\omega_{ph}t+\phi)\Theta(t)$,

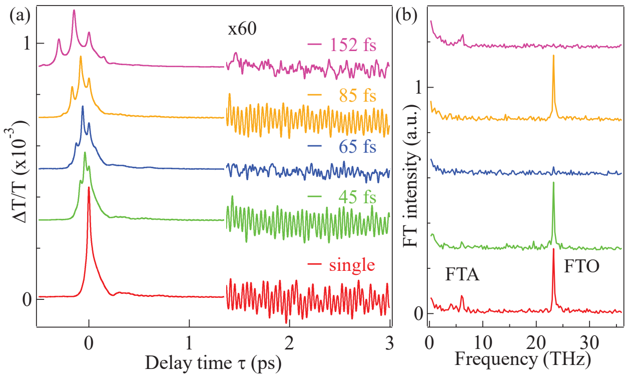

Fig. 2. (a) Transient transmission change of 4H-SiC. The red curve shows the result excited by single pulses, while others are the results excited by pulse trains consists of three pulses with different pulse intervals Δt = 45 fs, 65 fs, 85 fs, and 152 fs. After τ = 1 ps, all data were magnified by 60 for clarity. (b) Fourier transformed power spectra of (a) carried out 1 ps to 5 ps.

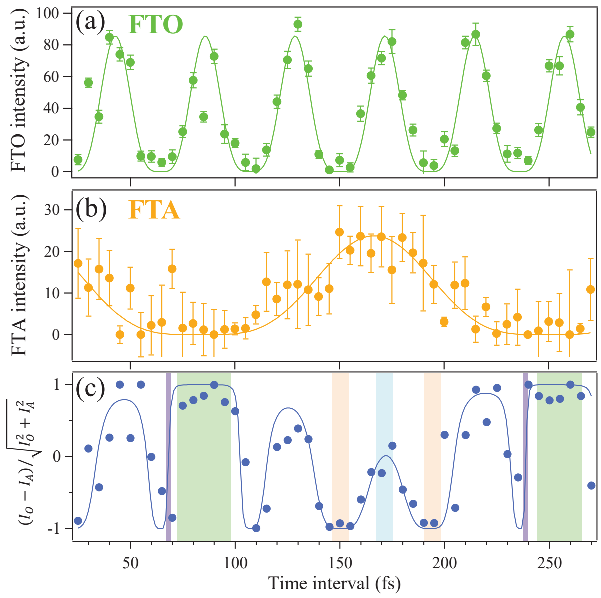

Fig. 3. FT intensity of (a) the FTO mode and (b) the FTA mode as a function of the time interval Δt of the three pulses. Solid curves are fitting curves $I(\omega_{\rm ph}, \Delta t)$. (c) Excitation ratio between the FTO and the FTA modes calculated by the normalized intensity of each modes.

where A is an amplitude, γ is a decay constant, ϕ is an initial phase, and $\Theta(t)$ is a step function at $t = 0$. By considering the three pulse excitation with an intensity ratio of 1 : 2 : 1 (Fig. 1(e)), the intensity of the transient transmission change is written as $I_{\rm ph}(\omega, \Delta t) = |\int_{\Delta_t}^{\infty}[f(t+\Delta t)+2f(t)+f(t-\Delta t)]dt|^2 \propto 6 + 8\cos(\omega_{ph}\Delta t) + 2\cos(2\omega_{ph}\Delta t)$, where the condition of $\gamma\Delta t \ll 1$ and $(\omega + \omega_{ph})^{-2} \ll 1$ were taken into account. Note that the initial phase ϕ does not contribute to the result of the above calculation. The best fits of this calculation are shown as the solid curves in Figs. 3(a) and 3(b). The calculated results are fairly in good agreement with the experimental data using parameter values of $\omega_{\rm ph}/2\pi$ = 23.3 ±0.1 THz for the FTO mode and 6.0 ±0.1 THz for the FTA mode, respectively.[19]

Finally, we would like to mention the selectivity of the FTO and FTA phonon modes induced via our pulse shaping technique. Here, we define the ratio of the two phonon modes coherently driven by a pulse train of three optical pulses as $C = (I_{\rm FTO} - I_{\rm FTA})/\sqrt{I_{\rm FTO}^2 + I_{\rm FTA}^2}$, where $I_{\rm FTO}$ and $I_{\rm FTA}$ are the intensities of the FTO and FTA modes normalized by the maximum amplitude derived from the fitting parameters as shown in Figs. 3(a) and 3(b). In this definition, $C = 1$ shows that the amplitude of the FTA mode is zero, while $C = -1$ represents the opposite scenario. Note that, in the case of $C = 0$, the two modes are coherently driven at the same rate by a pulse train of three optical pulses. Figure 3(c) illustrates the value of C as a function of Δt. The FTO is dominant in the green colored area, while the FTA is dominant in the orange area. As shown in the figure, the value of C is widely ranging from -1 to 1, strongly indicating that the two modes were precisely manipulated with high contrast; the both modes have maximum amplitude at 170 fs in the blue area, while become zero at 65 fs and 240 fs in the purple area. These results clearly show that our pulse shaping technique provides the high controllability of manipulating multiple phonon modes in 4H-SiC.

In summary, we have implemented ultrafast optical control of multiple coherent phonons in silicon carbide using a pulse shaping technique combined with sub-10 fs optical pulses and 2D LCOS-SLM. Our technique could conduct desirable pulse shaping such as number of pulses and time intervals between optical pulses, leading precise manipulation of multiple phonon modes; both the optical FTO and acoustic FTA modes of 4H-SiC were coherently controlled. By precisely tuning the injection timing of a pulse train of three optical pulses via our pulse shaping technique, the amplitude of the two modes were simultaneously manipulated with high contrast. Based on a model calculation assuming that the two phonons are excited independently by each pulse, respectively, the intensity contrast between the two modes was well reproduced. These results show that our scheme proposed in this study provides a simple pulse shaping for the coherent control of the zone-folded optical and acoustic phonon modes, which might be available for thermal managements in future high-power and broadband electronic devices.

Acknowledgments

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