Coherent optical and acoustic phonons generated at lattice-matched GaP/Si(001) heterointerfaces

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Abstract

Thin GaP films can be grown on an exact Si(001) substrate with nearly perfect lattice match. We perform all-optical pump–probe measurements to investigate the ultrafast electron–phonon coupling at the buried interface of GaP/Si. Above-bandgap excitation with a femtosecond laser pulse can induce coherent longitudinal optical (LO) phonons both in the GaP overlayer and in the Si substrate. The coupling of the GaP LO phonons with photoexcited plasma is reduced significantly with decreasing GaP layer thickness from 56 to 16 nm due to the quasi-two-dimensional confinement of the plasma. The same laser pulse can also generate coherent longitudinal acoustic phonons in the form of a strain pulse. The strain pulse induces not only a periodic modulation in the optical reflectivity as it propagates in the semiconductor, but also a sharp spike when it arrives at the GaP layer boundary. The acoustic pulse induced at the GaP/Si interface is remarkably stronger than that at the Si surface, suggesting a possible application of the GaP/Si heterostructure as an opto-acoustic transducer. The amplitude and the phase of the reflectivity modulation varies with the GaP layer thickness, which can be understood in terms of the interference caused by the multiple acoustic pulses generated at the top surface and at the buried interface.

Keywords: coherent phonon, gallium phosphide, silicon, ultrafast spectroscopy

(Some figures may appear in colour only in the online journal)
substrate [7–11]. Atomically-resolved transmission electron microscopy (TEM) studies revealed that the interface consists of a pyramidal structure with intermixing of ~7 atomic layers [12]. The well-defined GaP/Si heterointerface has the potential for application in high efficiency multi-junction solar cells [13, 14] as well as in monolithic integration of Ga(NAsP) lasers on Si substrates [15, 16].

In our previous study [17] we reported on an all-optical approach to evaluate electronic band structure at the buried GaP/Si(001) interfaces by means of coherent phonon spectroscopy. The technique is based on the generation of coherent longitudinal optical (LO) phonons of a polar semiconductor via transient screening of the depletion field mechanism, in which ultrafast drift-diffusion current suddenly screens the built-in electric field in the depletion region at the surface or interface [18–20]. We performed pump–probe reflectivity measurements using 10 fs near ultraviolet (NUV) pulses on thin GaP films grown on Si(001) under different conditions, and successfully reconstructed the band bendings of the heterostructures from the experimentally observed coherent phonon amplitudes. The same pump–probe scheme was also used to investigate coherent longitudinal acoustic (LA) phonons in the GaP/Si heterostructures [21]. Because of the short penetration depth of the NUV probe pulse, however, we could monitor the acoustic phonon dynamics for only a few tens of picoseconds. To obtain a more complete and quantitative understanding we need to follow the propagation of the acoustic phonons in Si over much longer temporal and deeper spatial scales.

In semiconductor heterostructures, coherent optical and acoustic phonons often exhibit different features from those in bulk semiconductors. In GaAs/AlGaAs quantum wells, for example, the Coulomb interaction between the LO phonons and the electron plasma, which would give rise to the LO phonon–plasmon coupled (LOPC) mode for the bulk GaAs, becomes weaker with decreasing GaAs well width due to the reduced spectral overlap between electronic and phononic bands [22]. When the Bloch oscillation energy in GaAs/AlGaAs superlattices [23] or the splitting energy of the heavy- and light-hole excitons in GaAs/AlAs multiple quantum wells (MQWs) [24] is tuned into resonance with the GaAs LO phonon energy, by contrast, the coherent electronic wavefunctions can drive the coherent phonons strongly. In GaAs/AlAs superlattices, the backfolding of the bulk phonon dispersion into the mini Brillouin zones leads to the emergence of zone-folded acoustic phonons that couple to the non-uniformly photoexcited carriers [25–27]. Photoexcitation of an InGaAs/GaN MQW by a single femtosecond NUV pulse can generate more intense coherent acoustic phonons [28–31] than those excited in a GaN film using a transient grating technique [32]. In contrast to these extensive studies on the GaAs- and GaN-based heterostructures, the electron–phonon couplings in heterostructures based on other semiconductors have not been investigated systematically [33–36].

In the present study we investigate the electron–phonon coupling dynamics at the GaP/Si heterointerfaces with different GaP overlayer thicknesses up to ~60 nm. We perform one-color pump–probe reflectivity measurements with NUV pump and probe pulses to monitor the coherent optical and acoustic phonons with 10 fs time resolution over a few tens of ps, and two-color measurements using NUV pump and visible probe to follow the propagation of the acoustic phonons in Si on sub-nanosecond time scale. We find that the GaP LO phonon–plasmon coupling depends critically on the GaP layer thickness due to the two-dimensional (2D) confinement of the plasmons in the thinner GaP layers. The amplitude of the sub-THz modulation in the transient reflectivity induced by the propagating coherent acoustic phonons also strongly depends on the GaP layer thickness, which can be explained in terms of the interference between reflections by multiple acoustic pulses generated at the GaP top surface and at the GaP/Si heterointerface.

### 2. Experimental methods

The samples studied are GaP thin films grown epitaxially on Si(001) substrates by metal organic vapor phase epitaxy (MOVPE). Details of the MOVPE growth and the structural characterization were described elsewhere [9–11]. (001)-oriented Si wafers with a 0.1° intentional miscut in the [110] direction are used to support the formation of double-steps. After a wet chemical cleaning procedure and a high-temperature heat treatment of the substrate, a Si buffer layer is grown in a chemical vapor deposition process using silane. The GaP-layer is then grown on the buffer layer. The first nucleation step is a pulsed growth scheme, where the precursors for Ga and P are offered intermittently at 450 °C. This is followed by continuous GaP overgrowth, in which both precursors are injected simultaneously at 675 °C, for different temporal durations to achieve different overlayer thicknesses \( t \) from 16 to 56 nm. Structural characterization and polarity determination of the GaP/Si samples are performed by TEM. Cross sectional images of the interfaces confirm the absence of major dislocations, stacking faults or twins in the GaP overlayers and the formation of self-annihilating anti-phase domains. For comparison, (001)-oriented \( n \)-doped Si and GaP single crystal wafers are also investigated.

Pump–probe reflectivity measurements are performed in a near back-reflection configuration under ambient conditions. For the one-color measurements, the second harmonic of a Ti:sapphire oscillator with 3.1 eV photon energy (400 nm wavelength), 10 fs duration and 80 MHz repetition rate is used for both pump and probe pulses. The NUV photons can excite carriers across the direct band gap at the \( \Gamma \) point of GaP and along the \( L \) valleys of Si [37–40]. The optical penetration depths for the 3.1 eV light are \( \alpha^{-1} = 116 \, \text{nm} \) and 82 nm in GaP and Si [41]. The pump and probe laser spots on the sample are ~30 \( \mu \text{m} \) in diameter. The pump-induced change in the reflectivity \( \Delta R \) is measured as a function of time delay \( t \) between pump and probe pulses using a fast scan technique. This scheme allows us to monitor the optical and acoustic phonons as well as the electronic response for the first few tens of ps with 10 fs time resolution.
For the two-color measurements, the second harmonic of a Ti:sapphire regenerative amplifier output, with 3.1 eV photon energy, 150 fs duration and 100 kHz repetition rate, is used as the pump pulse, whereas the output of an optical parametric amplifier with tunable photon energy between 2.56 and 2.00 eV (wavelength between 485 and 620 nm) serves as the probe. The pump and probe laser spots on the sample are \( \sim 250 \) and \( \sim 180 \) \( \mu \text{m} \) in diameter. \( \Delta R \) is measured as a function of \( t \) using a slow scan technique. This scheme allows us to monitor the acoustic pulses on the spatial and temporal scales up to \( \mu \text{m} \) and ns, since the penetration depth of the visible probe light, summarized in figure 1(a), is much larger than that of the NUV pump. In both one- and two-color schemes, the pump density is kept well below the threshold for the irreversible damage, which is \( \sim 200 \mu \text{J cm}^{-2} \) for bulk GaP and GaP/Si samples under irradiation of the 3.1 eV pump pulse.

3. Results

3.1. One-color pump–probe measurements

Figure 2 compares the pump-induced reflectivity changes \( \Delta R/R \) of the GaP films on Si, together with those of the bulk Si and GaP, obtained in the one-color pump–probe scheme. The traces for the bulk Si and GaP show a sharp spike at \( t = 0 \), followed by an increase and then by a decay within 1 ps, as shown in figure 2(a). The non-oscillatory responses provide information on the photoexcited carrier dynamics. The time constants of the increase and decay, 150 and 200 fs for Si, fall within the time scale of the carrier-phonon energy relaxation in the \( L \) valley reported in the previous studies [42–47]. The time constants for GaP, 400 and 440 fs, correspond to the \( \Gamma \rightarrow X_1 \) and \( \Gamma \rightarrow L \) scatterings [48, 49]. The non-oscillatory reflectivity changes of the GaP/Si heterointerfaces cannot be described by simple linear combinations of the two bulk signals. For \( d = 16 \) and 56 nm, especially, \( \Delta R/R \) drops instead of rises after photoexcitation without exhibiting an initial spike. The results suggest the importance of the interfacial carrier dynamics, such as the charge transfer across the heterointerface, in the reflectivity responses of the GaP/Si samples.

The reflectivity responses are also modulated by coherent optical phonons of GaP and Si with periods below 100 fs, as shown in figure 3(a). FT spectra for bulk GaP, shown in figure 3(b), exhibit a sharp peak of the LO mode at 12 THz at a low pump density of 10 \( \mu \text{J cm}^{-2} \). With increasing pump density, a broad band due to the LOPC mode grows at a lower frequency, and eventually overwhelms the LO peak, indicating the almost complete screening of the LO mode at high pump densities of \( \geq 60 \mu \text{J cm}^{-2} \). The frequency and the dephasing rate of the LOPC mode suggest that photoexcited mixed electron-hole plasma is responsible for the screening [20].

![Figure 1. Penetration depth \( \alpha^{-1} \) (a) and refractive index \( n \) (b) of Si and GaP as a function of photon energy \( E \) [41].](image1)

![Figure 2. Transient reflectivity traces of GaP/Si(001) heterointerfaces with different GaP overlayer thicknesses \( d \), together with those of bulk Si and GaP, pumped and probed at 3.1 eV. Pump and probe pulse duration is 10 fs. Pump densities are 70 \( \mu \text{J cm}^{-2} \) for bulk Si and 40 \( \mu \text{J cm}^{-2} \) for other samples. (a), (b) Show the same traces in different horizontal and vertical scales. Arrows in (b) indicate the reflectivity spikes. Traces are offset for clarity.](image2)
The GaP LO and LOPC modes are also observed in the GaP/Si heterointerface samples, together with the Si optical phonon at 15.6 THz. The appearance of the LOPC mode in the FT spectra (figure 3(b)) depends critically on the GaP layer thickness however. At \( d = 56 \text{ nm} \), the broad LOPC mode becomes dominant with increasing pump power, indicating as efficient screening of the LO phonons as in the bulk GaP. At \( d = 45 \text{ and } 35 \text{ nm} \), by contrast, the growth of the LOPC mode with pump power is less efficient, leaving the LO mode partially unscreened even at the highest pump density. At \( d = 16 \text{ nm} \), the sharp LO mode dominates the FT spectra at all pump densities without exhibiting clear indication of the LOPC mode. Our observation is consistent with the previous report on the well width-dependent LO-plasmon coupling in GaAs MQWs [22] and can essentially be explained in terms of the dimensionality-dependent dispersion relations of the plasmons. For three-dimensional (3D) and 2D plasmons, the dispersion relations in the long wavelength limit are given by [50–52]:

\[
\omega_{\text{3D}}(q) = \sqrt{\omega_{\text{pl}}^2 + \frac{q^2}{c^2}},
\]

\[
\omega_{\text{2D}}(q) = \sqrt{\frac{\omega_{\text{pl}} q^2}{2}},
\]

where \( q \) is the wavevector and \( \omega_{\text{pl}} = \sqrt{4\pi Ne/m^*c^2} \) is the plasma frequency as a function of carrier density \( N \), effective mass \( m^* \) and the high-frequency dielectric constant \( \epsilon_{\infty} \). The 3D plasmons have nonzero frequency at \( q \to 0 \), and can strongly couple with the LO phonons at \( q \sim \omega_{\text{pl}} \) comparable to the LO phonon frequency [20]. By contrast, purely 2D plasmons have a vanishing frequency at \( q \to 0 \) and therefore do not strongly interact with the LO phonons. In the present study, the efficient (inefficient) screening of the LO phonons at \( d = 56 \text{ nm} \) (16 nm) indicates that the plasmons in the GaP film acquire a quasi-3D (quasi-2D) dispersion relation; at the intermediate thicknesses the plasmon dispersion is in between.

The reflectivity traces for bulk GaP and Si are modulated periodically also on a much longer time scale, as shown in figure 2(b). These slow modulations are caused by the interference between the probe beams reflected from the propagating acoustic pulse and from the surface (Brillouin oscillation), as schematically shown in figure 4(a), and their frequencies are given by \( f_{\text{B}} = 2nv/\lambda = 123 \text{ and } 235 \text{ GHz for GaP and Si for normal incidence probe} [40, 53] \). Here \( n \) is the refractive index at the probe photon energy, as plotted in figure 1(b), and \( v \) is the LA phonon velocity in the [001] direction \( \nu_{\text{GaP}} = 5.847 \text{ nm ps}^{-1} \text{ and } \nu_{\text{Si}} = 8.4332 \text{ nm ps}^{-1} [54, 55] \).

Similar, but less periodic, modulations appear in the reflectivity traces for the GaP/Si interfaces, as shown in figure 2(b). The FT spectrum for \( d = 56 \text{ nm} \), shown in figure 5(a), features a distinct peak at \( f_{\text{B}} \text{GaP} \) that is indicative of an acoustic pulse propagating in the GaP film. The FT spectra for the thinner GaP films, by contrast, are more complicated and exhibit dips and peaks between \( f_{\text{B}} \text{GaP} \) and \( f_{\text{B}} \text{Si} \), suggesting the contribution also from an acoustic pulse propagating in the Si substrate. In principle, acoustic pulses can be generated not only at the GaP top surface but also at the GaP/Si interface, where the optical

Figure 3. Oscillatory parts of the transient reflectivity changes (a) and their Fourier-transformed (FT) spectra (b) for GaP/Si(001) heterointerfaces with different \( d \), together with those of bulk GaP, pumped and probed at 3.1 eV.

Figure 4. Schematics of the generation and optical detection of acoustic pulses at a semiconductor surface (a) and at a buried interface (b). At \( t = 0 \), pump light generates an acoustic pulse at the surface or interface (upper panels). At \( t > 0 \), probe light is reflected by the acoustic pulses propagating in the semiconductors in the depth direction as well as by the fixed surface and interface. The interference between the reflected probe lights leads to the periodic modulation of the reflected probe intensity as a function of \( t \). The incident angle of the probe light is exaggerated for clarity.
absorption coefficient and the electronic band structure have discontinuities; the latter can propagate in both GaP and Si directions, as schematically shown in figure 4(b). From the one-color pump–probe experiments, however, we cannot determine the relative contributions of the acoustic pulses to the reflectivity modulations, mainly because the short penetration depth of the probe light does not allow us to monitor the acoustic pulses long enough until they all propagate into the Si substrate.

In addition to the quasi-periodic modulations, the reflectivity traces from the GaP/Si interfaces exhibit extra features seen as sharp spikes, as indicated by the arrows in figure 2(a) [21]. These spikes are absent for bulk GaP and Si and are therefore characteristic of the heterointerface. The spikes appear at delay times \( t \sim d/\gamma_{\text{GaP}} \), as plotted in figure 5(b), implying that they are induced by the acoustic pulse generated at the GaP/Si interface and detected at the GaP/air surface, and/or vice versa. The temporal width of the spikes, \( \sim 0.5 \) ps, translates to a spatial extent of \( \sim 3 \) nm or \( \sim 10 \) atomic Ga-P layers, which is comparable with the intermixing of seven atomic layers due to the pyramidal structures at the interface [12]. The extremely narrow spatial extent of the acoustic pulse suggests a potential application as an opto-acoustic transducer for high-resolution nano-seismology for Si-based structures.

### 3.2. Two-color pump–probe measurements

Figure 6(a) shows the transient reflectivity changes of GaP/Si interfaces, together with those of bulk Si and GaP, pumped at 3.1 eV and probed at different photon energies \( E \) in the visible range. Like in the one-color measurements, \( \Delta R/R \) comprises the non-oscillatory component associated with the carrier dynamics and the periodic modulation with sub-THz frequency that is indicative of the propagating acoustic pulse. Unlike the one-color measurements, however, the durations of the pump and probe pulses in the two-color measurements are too long to excite and detect the coherent LO phonons of GaP and Si.

The reflectivity traces of bulk Si drop rapidly after photoexcitation and recover bi-exponentially with time constants of 7 and 65 ps that are independent of \( E \). For bulk GaP, the reflectivity traces show an initial rapid rise, followed by a slower increase and then a decay with the time constants of 11 and 290 ps being independent of \( E \). The shorter time constant is roughly in the range of the \( L \rightarrow X \) scattering in Si [46] and the \( X_3 \rightarrow X_1 \) scattering in GaP [49, 56], whereas the slower decay is more likely to be due to the phonon–phonon scattering (lattice heating) [57] and the surface recombination [43]. The reflectivity traces for the GaP/Si heterointerfaces at \( d = 16 \) and 56 nm are somewhat similar to those of bulk Si and GaP, respectively, in particular at relatively low \( E \), as shown in figures 6(a) and (b). By contrast, those for \( d = 35 \) and 45 nm rise almost instantaneously after photoexcitation and decay bi-exponentially, unlike bulk Si or GaP. For all the heterointerfaces, the decay/rise on sub-50 ps time scale becomes faster with increasing \( E \). The observations indicate that the interfacial carrier dynamics dominate the reflectivity response.

The sub-THz reflectivity oscillations of all the GaP/Si heterointerfaces, shown in figure 6(c), can be fitted to a single damped harmonic function:

\[
g(t) = A_0 \exp(-\Gamma t) \cos(2\pi ft + \phi),
\]

in the time range of \( t \gtrsim 10 \) ps. The frequency \( f \) corresponds to \( f_{0}^{E_R} \) at given \( E \), as shown in figure 7(a). Unlike in the one-color measurements, we see no clear sign of the modulation at \( f_{0}^{E_R} \) except for the small wiggles for the first cycle or two. This can be understood in terms of the larger probing depth of the visible probe light, which makes the two-color scheme detection less sensitive to the acoustic dynamics in the GaP layer. The dephasing rate \( \Gamma \), summarized in figure 7(b), does not system-}

atically depend on \( E \) or \( d \). The insensitivity of \( \Gamma \) to \( E \) implies the damping of the interference patterns due to the dephasing among the oscillations at different wavelengths within the broadband probe light [58] rather than due to the acoustic pulses moving out of the probing depth \( (2\alpha)^{-1} \), which critically depends on \( E \) as shown in figure 1(a). Calculations of the strain-induced reflectivity changes of bulk Si by taking into account the finite probe spectral width (0.03–0.04 eV) reproduced the experimentally observed damping and the frequency quantitatively [40].

Figure 8 plots the initial amplitude \( A_0 \) of the interference pattern as a function of \( d \). We notice that \( A_0 \) for bulk Si \( (d = 0 \) nm) is negative, i.e., the reflectivity modulation is approximately proportional to \(- \cos(2\pi f_{0}^{E_R} t)\), at all \( E \). \( A_0 \) for the GaP/Si heterointerface for \( d = 16 \) nm is also negative but its absolute value is larger than that of bulk Si by a factor of 3–7 depending on \( E \), suggesting that addition of a thin GaP overlay on top can enhance the acoustic pulse propagating in Si. This is reasonable, because the relative deformation potential coupling constant \( a_{cv} \), which dominates the optical generation of the strain in the semiconductors, was estimated to be at least twice as large in GaP as in Si at 3.1 eV [40]. We note that the efficiency of a single
GaP overlayer as an opto-acoustic transducer appears to be moderate compared with metallic transducers such as thin NiFe, Ti and Au films on top of Si, which can induce at least an-order-of-magnitude more intense ($A_0 > 10^{-4}$) reflectivity oscillations [59, 60]. This is basically because the metallic films can be photoexcited at more intense pump fluence by up to two orders of magnitude ($\sim 10 \text{ mJ cm}^{-2}$), and can generate correspondingly intense strain pulse in Si. As we have seen in the one-color measurements, the GaP overlayer on Si has an advantage in generating acoustic pulses with ultrashort duration, because of the microscopically controlled interfacial structure and because of the ultrafast strain generation via the deformation potential coupling mechanism rather than thermoelastic effect that dominates in metals.

With further increasing $d$, the amplitude $A_0$ does not monotonically increase; instead, it exhibits an apparently oscillatory behavior, with its sign flipped from negative for $d = 16$ nm to positive for the larger $d$. For bulk GaP, where the reflectivity is modulated at $f_{\text{GaP}}$, $A_0$ is negative again. This peculiar $d$-dependence is seemingly inconsistent with the absorption of the pump and probe lights by the GaP overlayer, which would lead to a monotonic decrease in $A_0$ with increasing $d$. Nor can it be explained in terms of the $d$-dependent phase-shift of the pump light due to the different refractive index in GaP and Si, since the coherent acoustic phonons are not induced directly by the electric field of the pump light but through the photoexcited carriers in the present experimental scheme. To understand the seemingly counterintuitive thickness-dependence, we consider a theoretical model of optical generation.

Figure 6. (a) and (b) Transient reflectivity traces of GaP/Si(001) heterointerfaces with different $d$, together with those of bulk Si and GaP, pumped at 3.1 eV and probed at photon energies $E$. Pump and probe pulse duration is 150 fs. Pump density is 90 $\mu$J cm$^{-2}$. Panels (a) and (b) show the same traces in different horizontal scales. (c) The oscillatory part of the reflectivity traces in (a).
and detection of the acoustic pulse at the heterointerface in the following section.

4. Discussion

We start to theoretically model the optical detection of the coherent acoustic phonons by considering a probe light wave, which is described by $E_0(\mathbf{r}, t) = E_0 e^{i(k_0 \mathbf{r} - \omega t)}$ in vacuum, incident on a semiconductor surface (without an overlayer) from the normal direction. Once inside the semiconductor, whose complex refractive index is $\tilde{n}_1 \equiv n_1 + i\tilde{k}_1$, the light wave is expressed by

$$E_1(z, t) = t_01 E_0(t) \exp[i(k_1 z - \omega t)], \quad (4)$$

with $k_1 \equiv \tilde{n}_1 k$ being the light wavevector in the semiconductor. The transmission coefficients for the light incoming to and outgoing from the semiconductor are given by

$$t_{01} = \frac{2}{1 + \tilde{n}_1}; \quad t_{10} = \frac{2\tilde{n}_1}{1 + \tilde{n}_1}. \quad (5)$$

The complex reflection coefficient (in the absence of the photoexcitation) is expressed by

$$r_{01} = \frac{1 - \tilde{n}_1}{1 + \tilde{n}_1}. \quad (6)$$

We assume that a separate pump light induces a change $\delta \tilde{n}_1(k, z, t)$ in the refractive coefficient. Since the reflected probe light crosses the semiconductor surface at $z = 0$ twice and propagates in the semiconductor both ways, the corresponding change in the complex reflection coefficient is given by [30, 61]:

$$\delta r = t_{01} t_{10} \left[ -\frac{1}{2\tilde{n}_1} \int_0^\infty e^{2i\tilde{k}_1 z} \frac{\partial}{\partial z} \delta \tilde{n}_1(z, t) dz \right]$$

$$= \frac{-2}{(1 + \tilde{n}_1)^2} \int_0^\infty e^{2i\tilde{k}_1 z} \frac{\partial}{\partial z} \delta \tilde{n}_1(z, t) dz;$$

$$\frac{\delta r}{r_{01}} = \frac{2}{n_1^2 - 1} \int_0^\infty e^{2i\tilde{k}_1 z} \frac{\partial}{\partial z} \delta \tilde{n}_1(z, t) dz. \quad (7)$$

The intensity of the reflected probe light in the absence of the pump light is given by $R_0 = r_{01} r_{10}^*$. The pump-induced change in the reflected probe intensity can be expressed by

$$\frac{\Delta R}{R_0} \equiv \frac{R - R_0}{R_0} = \frac{(r_{01} + \delta r)(r_{01}^* + \delta r^*) - r_{01} r_{01}^*}{r_{01} r_{01}^*}$$

$$\simeq \frac{\delta r^*}{r_{01}} + \frac{\delta r}{r_{01}} = 2 \text{Re} \left( \frac{\delta r}{r_{01}} \right), \quad (8)$$

when $\delta r$ is small ($\delta r \ll r_{01}$). By substituting equations (7) into (8) we obtain

$$\frac{\Delta R}{R_0} \simeq \text{Re} \left[ -\frac{4}{n_1^2 - 1} \int_0^\infty e^{2i\tilde{k}_1 z} \frac{\partial}{\partial z} \delta \tilde{n}_1(z, t) dz \right]. \quad (9)$$

Now we consider the change in the refractive index induced by a pump-induced strain $\eta(z, t)$:

$$\delta \tilde{n}_1(z, t) = \frac{\delta \tilde{n}_1}{\delta \eta} \eta(z, t) = \frac{1}{2\tilde{n}_1} \frac{\partial \eta}{\partial \eta} \eta(z, t), \quad (10)$$

where $\epsilon \equiv \epsilon_r + i \epsilon_i = \tilde{n}_1^2$ is the complex dielectric constant. We assume that the strain modulates the semiconductor bandgap and thereby the dependence of $\epsilon$ on the probe photon energy $E$ [40]:

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure7.png}
\caption{Frequency $f$ (a) and dephasing rate $\Gamma$ (b) of the reflectivity modulation as a function of probe photon energy $E = h\epsilon/\lambda$. Symbols represent experimental data. The solid (dotted) curve in (a) represents $f = 2n\epsilon/\lambda$ with $n$ and $\epsilon$ being the refractive index and the LA phonon velocity of Si (GaP).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure8.png}
\caption{Initial amplitude $A_0$ of reflectivity modulation of GaP/ Si interfaces as a function of GaP thickness $d$ measured at different probe photon energies $E$.}
\end{figure}
\( \epsilon(E, \eta) \simeq \epsilon(E - a_{cv}\eta). \)  

(11)

Here \( a_{cv} \equiv -K(\partial E / \partial \rho) \) is the relative deformation potential coupling constant, \( K \), the bulk modulus, \( E_0 \), the band gap, and \( \rho \), the pressure. Then we can approximately express the variation in the strain-induced dielectric constant at a fixed probe energy \( E = \hbar c k \) by

\[
\frac{\partial \epsilon}{\partial \eta} \frac{\partial E}{\partial \epsilon} \approx \frac{a_{cv}}{E_0} \left. \frac{\partial \epsilon}{\partial E} \right|_{E=\hbar c k} \tag{12}
\]

and that in the refractive index by

\[
d \tilde{\eta}(z, \tau) \simeq \frac{a_{cv}}{2n_1} \left. \frac{\partial \epsilon}{\partial E} \right|_{E=\hbar c k} \eta(z, \tau). \tag{13}\]

By substituting this into equation (9) we obtain

\[
\frac{\Delta R}{R_0} \simeq \Re \left[ \frac{2a_{cv}}{n_1(n_1^2 - 1)} \left. \frac{\partial E}{\partial \epsilon} \right|_{E=\hbar c k} \int_0^\infty \frac{e^{2\hbar c k/\epsilon}}{\epsilon \partial \epsilon} d\epsilon \right] . \tag{14}\]

The generation of coherent acoustic phonons in bulk GaP and Si is dominated by the deformation potential electron–phonon coupling with photoexcited carriers [40]. If we neglect the transport and recombination of photoexcited carriers and approximate the depth distribution of the carrier density by

\[
N(t) = \begin{cases} 0 & \text{for } t < 0 \\ \alpha_{pu}(1 - R_{pu}) Fe^{-\alpha_{pu}t}/E_{pu} & \text{for } t \geq 0 \end{cases} \tag{15}
\]

the strain can be expressed as a linear function of the pump light fluence \( F \) and \( a_{cv} \), by [40]:

\[
\tilde{\eta}(z, \tau) = \frac{\alpha_{pu}a_{cv}(1 - R_{pu}) F}{2E_{pu}n_1^2} \left[ e^{-\alpha_{pu}(z + \tau)} \left( e^{\alpha_{pu}vt} - 1 \right)^2 \right. \\
- \left. (v \tau - z) \left( e^{-\alpha_{pu}(z - \tau)} + e^{\alpha_{pu}(z - \tau)} \right) \right]. \tag{16}\]

Here \( \alpha_{pu}, R_{pu} \) and \( E_{pu} \) are the absorption coefficient, the reflectivity and the photon energy for the pump light. This yields a photo-induced strain consisting of a step-like wave front that is generated at the surface \( z = 0 \) at \( t = 0 \) and propagates into the depth direction with velocity \( v \), and an elastic component that is only moderately dependent on \( z \). We therefore approximate the differential strain with a delta function:

\[
\frac{\partial \epsilon}{\partial \eta} \simeq B \delta(z = \nu t) \tag{17}\]

with a real amplitude \( B \), and obtain

\[
\frac{\Delta R}{R_0} \simeq \Re \left[ \frac{2a_{cv}B}{n_1(n_1^2 - 1)} \left. \frac{\partial E}{\partial \epsilon} \right|_{E=\hbar c k} \int_0^\infty \frac{e^{2\hbar c k/\epsilon}}{\epsilon \partial \epsilon} d\epsilon \right] \\
= \Re \left[ \frac{2a_{cv}B}{n_1(n_1^2 - 1)} \left. \frac{\partial E}{\partial \epsilon} \right|_{E=\hbar c k} e^{2\hbar c k/\epsilon} \right]. \tag{18}\]

Since \( n_1 \gg k_1 \) for the visible light in Si and GaP [41], we can safely neglect the imaginary part of \( n_1 \) and approximate the variation in the reflected probe intensity by

\[
\frac{\Delta R}{R_0} \approx \left| \frac{2a_{cv}B}{n_1(n_1^2 - 1)} \left. \frac{\partial E}{\partial \epsilon} \right|_{E=\hbar c k} \right| e^{2\hbar c k/\epsilon} \left. \frac{\partial \epsilon}{\partial \nu} \right|_{E=\hbar c k}. \tag{19}\]

Equation (19) describes the periodic modulation in the reflected probe intensity (Brillouin oscillation) at a frequency \( f_B = 2k_1v/2\pi = 2n_1v/\lambda \) for bulk semiconductors.

The strain-induced reflectivity change for the GaP/Si interface has a much more complex expression, because the acoustic pulses can be generated also at the heterointerface, and because they can be reflected when they reach the boundaries of the GaP layer. At long time delays of \( t \geq 2d/\sqrt{B_2} \), however, the strain pulses are all located in the Si substrate and propagating in the depth direction, as schematically shown in figure 9(a), if we neglect the small \((\sim 10\%)\) reflection at the

Figure 9. (a) Schematic illustration showing the positions of acoustic pulses at time delays \( t \geq 2d/\sqrt{B_2} \). (b) Calculated \( d \)-dependence of \( A_0 \) at probe photon energy \( E = 2.46 \) eV with different ratios between amplitudes \( B_1, B_2 \) and \( B_3 \) in equation (25). Filled and open circles represent the experimental \( A_0 \) of Si with and without GaP the overlayer measured at \( E = 2.46 \) eV.
heterointerface. In this case the differential strain can be simplified into an expression:

$$\frac{\partial \eta}{\partial z} \approx B_1 \delta (z = z_1) + B_2 \delta (z = z_2) + B_3 \delta (z = z_3)$$

(20)

with

$$z_1(t) = v_{Si} t;$$
$$z_2(t) = v_{Si} (t - d/\sqrt{GaP});$$
$$z_3(t) = v_{Si} (t - 2d/\sqrt{GaP})$$

being the positions of pulse 1, 2 and 3 with respect to the GaP/Si interface. We then obtain the approximate expression for the reflectivity modulation from the heterointerface:

$$\frac{\Delta R}{R_0} \approx \frac{2a_{cv}}{n_1(n_1^2 - 1)} \left| \frac{\partial E}{\partial \epsilon} \right| \text{Re} \left[ B_1 \exp i(2k_1 v_{Si} t + \phi) \right.$$  
$$+ B_2 \exp i \left[ 2k_1 v_{Si}(t - \frac{d}{\sqrt{GaP}}) + \phi \right] \right.$$  
$$+ B_3 \exp i \left[ 2k_1 v_{Si}(t - \frac{2d}{\sqrt{GaP}}) + \phi \right].$$

(22)

By using a polar form:

$$B_1 + B_2 \exp \left( -\frac{2ik_1 v_{Si} d}{\sqrt{GaP}} \right) + B_3 \exp \left( -\frac{4ik_1 v_{Si} d}{\sqrt{GaP}} \right)$$

$$\equiv |A_0| e^{i\psi}$$

(23)

the change in the reflected probe intensity can be expressed by

$$\frac{\Delta R}{R_0} \approx \frac{2a_{cv}|A_0|}{n_1(n_1^2 - 1)} \left| \frac{\partial E}{\partial \epsilon} \right| \text{Re} \left[ \exp \left( 2ik_1 v_{Si} t + \phi + \psi \right) \right]$$  
$$\approx \frac{2a_{cv}|A_0|}{n_1(n_1^2 - 1)} \left| \frac{\partial E}{\partial \epsilon} \right| \cos \left( 2k_1 v_{Si} t + \phi + \psi \right).$$

(24)

Here the amplitude and the phase are defined by

$$|A_0|^2 \equiv \left[ B_1 + B_2 \cos \left( \frac{2k_1 v_{Si} d}{\sqrt{GaP}} \right) + B_3 \cos \left( \frac{4k_1 v_{Si} d}{\sqrt{GaP}} \right) \right]^2 +$$
$$+ \left[ B_2 \sin \left( \frac{2k_1 v_{Si} d}{\sqrt{GaP}} \right) + B_3 \sin \left( \frac{4k_1 v_{Si} d}{\sqrt{GaP}} \right) \right]^2;$$

$$\tan \psi \equiv \frac{B_1 + B_2 \cos \left( \frac{2k_1 v_{Si} d}{\sqrt{GaP}} \right) + B_3 \cos \left( \frac{4k_1 v_{Si} d}{\sqrt{GaP}} \right)}{B_2 \sin \left( \frac{2k_1 v_{Si} d}{\sqrt{GaP}} \right) + B_3 \sin \left( \frac{4k_1 v_{Si} d}{\sqrt{GaP}} \right)}.$$

(25)

Figure 9(b) compares the calculated $d$-dependence of $A_0$ at $E = 2.46$ eV with the experiment. We see that equation (25) reproduces the experimental oscillatory behavior reasonably well when we consider the interference arising only from the pulses 1 and 2 by putting $B_1 : B_2 : B_3 = 1 : 1 : 0$, but not with $1 : -1 : 0$. In both cases, adding small positive or negative $B_1$ does not change the calculated $d$-dependence drastically. If we consider the interference arising only from the pulses 1 and 3 by putting $B_1 : B_2 : B_3 = 1 : 0 : \pm 1$, the oscillation with $d$ has twice the frequency and does not match the experiment. The results confirm that the interference between the reflections from multiple acoustic pulses is responsible for the oscillatory $d$-dependence, with the contribution from pulse 2 as important as from pulse 1. The calculations suggest that the pulses 1 and 2 have the amplitudes with the same sign, which gives us a hint in determining which satellite valley is most relevant in the acoustic phonon generation, since $a_{cv}$ for different valleys can have different signs [62, 63]. The contribution from pulse 3 is small in comparison, $|B_3|/|B_1| < 0.3$, which can be attributed to the partial loss when it is reflected by the GaP surface and transmits across the heterointerface.

5. Summary

We have investigated the ultrafast electron–phonon coupling at lattice-matched heterointerfaces of GaP/Si(001) in the form of coherent optical and acoustic phonons. The screening of the polar LO phonons of GaP with photoexcited plasmas depends critically on the GaP layer thickness, which is attributed to the quasi-2D character of the plasmons confined in the thin GaP layers. The presence of the GaP overlayer on top enhances the acoustic pulse that propagates in Si through the larger deformation potential electron–phonon coupling in GaP. Interference between the probe lights reflected by multiple acoustic pulses generated at the GaP/Si interface and at the GaP top surface leads to the reflectivity modulation whose amplitude depends on the GaP overlayer thickness. We have thereby demonstrated the ultrafast electron–phonon couplings that are characteristic of a semiconductor heterostructure with an abrupt, almost defect-free interface. The insight obtained in the present study will stimulate the development of theoretical simulations that better describe the optical generation and detection of acoustic phonons at the GaP/Si and other semiconductor heterointerfaces.

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