Separation of interface and substrate carrier dynamics at a heterointerface based on coherent optical phonons

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Transient reflectivity spectroscopy is widely used to study ultrafast carrier- and phonon-dynamics in semiconductors. In their heterostructures, it is often not straightforward to distinguish contributions to the signal from the various layers. In this work, we perform transient reflectivity measurements on lattice-matched GaP/Si(001) using a near infrared pulse, to which GaP is transparent. The pump laser pulse can generate coherent longitudinal optical (LO) phonons both in the GaP overlayer as well as in the Si substrate which have distinct frequencies. This enables us to track the amplitude of the respective signal contributions as a function of GaP layer thickness $d$. The Si phonon amplitude in the signal exhibits an oscillatory behavior with increasing $d$. This can be quantitatively explained by the interference of the probe light reflected at the air/GaP/Si heterointerface. Based on this knowledge, we can then separate the interface- and the substrate-contributions in the carrier-induced non-oscillatory transient reflectivity signal. The obtained interface signal provides evidence for ultrafast carrier injection from the Si substrate into the GaP overlayer. This is also corroborated by examining the deviation of the polarization-dependence of the GaP coherent optical phonon signal from that of the bulk semiconductor.

I. INTRODUCTION

Ultrafast carrier- and phonon-dynamics in semiconductors and their heterostructures can fundamentally influence the performance of electronic devices and have therefore been studied extensively by a variety of theoretical and experimental methods [1, 2]. Transient reflectivity, a pump/probe, light-in, light-out technique based on a linear optical process, is among the most conventional and widely used experimental methods. It is particularly powerful to detect coherent optical and acoustic phonons, which can be induced by ultrashort laser pulses and detected as periodic modulations of the transient reflectivity at THz and GHz frequencies [3, 4]. Because the modulation frequencies are characteristic to the materials, it is relatively straightforward to interpret phonon-induced signals obtained from heterojunctions into contributions from different semiconductor layers. This is not the case for transient reflectivity associated with dynamics of the photoexcited carriers, which typically manifests itself as a superposition of exponential functions. It requires simultaneous measurements of transient reflectivity and a non-linear spectroscopy with surface- and interface-selectivities, such as second harmonic generation (SHG), to precisely specify the carrier contributions from specific layers [5].

Among various semiconductor heterostructures, GaAs/AlAs quantum wells and superlattices have been studied most extensively [6–8]. For other combinations of semiconductors, however, lattice mismatch often leads to a strain at the heterointerface, which can crucially affect the electronic and phononic properties [9–11]. Recently fabrication of abrupt GaP/Si heterointerfaces without extended defects has been made possible [12–19]. They can also serve as a model heterojunction because of the small lattice mismatch and small intermixing at the interface. In previous studies, systematic transient reflectivity measurements revealed the generation of coherent longitudinal optical (LO) and longitudinal acoustic (LA) phonons upon above-bandgap photoexcitation of the GaP layer and the Si substrate [20–22]. The underlying electron-phonon interaction was found to be qualitatively similar to those of the respective bulk semiconductors under the same excitation condition [23, 24], except for the reduced LO phonon-plasmon coupling for the thinnest GaP layer examined (thickness $d = 16$ nm) [22]. By contrast, the GaP/Si interfaces for below-bandgap excitation of GaP remains mostly unexplored, except for a time-resolved SHG study on an extremely thin ($d = 4.5$ nm) GaP layer [25]. There, a fast ($< 400$ fs) rise and decay in the SH signal was detected, with its intensity peaked at pump photon energy of 1.4 eV. In addition, a delayed ($\sim 2$ ps) rise was observed for pump energies above 1.4 eV. These observations were interpreted as an electronic transition involving a short-lived electronic state at the heterointerface, whose energy lies in the bandgaps of the two semiconductors, and the subsequent transport of the photoexcited charge carriers.

In the present study we investigate the GaP/Si(001) heterointerfaces in pump-probe reflectivity scheme with near infrared optical pulses, whose photon energy exceeds
the indirect bandgap of Si but is well below that of GaP. The reflectivity signals are periodically modulated at the well-resolved frequencies of the LO phonons of GaP and Si, which enables us to separately analyze the phonon signals from the two semiconductors. The amplitude of the Si coherent phonon exhibits an oscillatory dependence on the GaP overlayer thickness, which can be explained quantitatively by taking into account the interference of the probe wave reflected at the heterointerface. This finding enables us to unambiguously decompose the carrier-induced reflectivity signal into the contributions from the interface and from the substrate.

II. EXPERIMENTAL METHODS

The samples studied are nominally undoped GaP films grown by metal organic vapor phase epitaxy with thickness between \( d = 8 \) and 48 nm on exact Si(001) substrates. Details of the fabrication procedure are described elsewhere [14, 16]. An 8-nm thick nucleation layer of GaP is first grown in a slow-rate modulated epitaxy at 450°C for all the samples studied. The as-grown GaP nucleation layer consists of crystalline grains with lateral size of \( \lesssim 30 \) nm [14]. This is followed by the overgrowth of GaP in a continuous epitaxy at 675°C for the samples \( d \geq 18 \) nm. The GaP layers after the overgrowth have steps that directly trace those of the Si substrate [14, 16], as shown schematically in Fig. 1a.

Single-color pump-probe reflectivity measurements are performed in ambient conditions in a near back reflection geometry. To investigate the phonon dynamics, an output of a Ti:sapphire oscillator with 12-fs duration, 815-nm center wavelength and 80-MHz repetition rate is used as the light source. GaP is transparent to the 815-nm light, whereas the optical penetration depth in Si is \( \sim 10 \) \( \mu \)m [26]. A spherical mirror brings the linearly polarized pump and probe beams to a \( \sim 30-\mu \)m diameter spot on the sample with incident angles of \( \lesssim 15^\circ \) and \( \lesssim 5^\circ \) from the surface normal. Anisotropy in the pump-induced reflectivity change, \( \Delta R_{eo} = \Delta R_H - \Delta R_V \), is measured by detecting the horizontal (\( H \)) and vertical (\( V \)) polarization components of the probe light with a pair of matched photodiode detectors, as shown in Fig. 1b. This scheme is suitable to detect the LO phonons of GaP and Si, which have only off-diagonal Raman tensor components as described by eq. (A2) in Appendix A, but not ideal to monitor the mostly isotropic carrier dynamics. The signal from the detector pair is pre-amplified and is averaged in a digital oscilloscope typically over 10,000 times while the delay \( t \) between the pump and probe pulses is scanned continuously with a fast scan delay.

To examine the carrier dynamics, an output of a regenerative amplifier with 150 fs duration, 810 nm wavelength, and 100 kHz repetition rate is used as the light source. The pump and probe spot size on the sample is \( \sim 100 \) \( \mu \)m. Pump light is chopped at a frequency of \( \sim 2 \) kHz for a lock-in detection. Pump-induced change in the reflectivity \( \Delta R \) is measured by detecting the probe lights before and after the reflection with a pair of matched photodiode detectors. The signal from the detector pair is amplified with a current pre-amplifier and a lock-in amplifier. The time delay \( t \) between the pump and probe pulses is scanned with a linear motor stage (slow scan).

III. RESULTS

We first examine the phonon dynamics by measuring \( \Delta R_{eo}/R_0 \). Figure 2a shows a typical anisotropic reflectivity change for the \( d = 48 \) nm GaP/Si sample. We extract its oscillatory part by subtracting the non-oscillatory baseline that can be fitted to a multiple exponential function. The obtained oscillations are summarized in Fig. 2b for all the GaP/Si samples at two representative pump polarizations. The oscillations consist mainly of two frequencies, 12 and 15.6 THz, which are seen as sharp peaks in the Fast Fourier-transformed (FFT) spectra in Fig. 2c. These peaks arise from coherent LO phonons of GaP and Si, respectively, as is evident from the comparison with the signals of bulk GaP and Si shown in the same figure [27].

For the bulk GaP and Si crystals the [-110] and [110]
FIG. 2. (a) Anisotropic reflectivity change for \(d = 48\) nm. (b) Oscillatory parts of the anisotropic reflectivity changes of GaP/Si(001) samples with different \(d\). (c) Fast Fourier transform spectra of (b) in the optical phonon regime. Pump polarization is either parallel to the [-110] or [110] directions of the Si substrate (labeled respectively with I and II). Probe light is polarized nearly along the [100] direction for the anisotropic detection. Reflectivity signals from (001)-oriented Si and GaP wafers with pump polarization parallel to the [-110] and [110] axes (labeled with I' and II') are also shown for comparison. Incident pump density is 0.18 mJ/cm\(^2\). Traces are offset for clarity.

crystallographic directions are equivalent. Correspondingly, the LO phonon amplitudes are comparable between the pump polarizations along these two directions, as shown in Fig. 2bc, while the phases of the oscillations are opposite to each other because of the Raman generation, as explained in Appendix A. For the GaP/Si samples, by contrast, the two directions can be distinguished based on the miscut of the Si substrate surface, as schematically shown in Fig. 1a. The GaP peak height in Fig. 2c is apparently larger for pump polarization along the [110] axis (labeled “II”) than along the [-110] axis ("I"). For a fixed pump polarization, the GaP peak height increases monotonically with increasing \(d\). By contrast, the Si peak height is comparable between the two polarizations for all the GaP/Si samples examined. It depends on \(d\) in an apparently complicated manner, however, i.e., first decreases and then increases with increasing \(d\).

For quantitative analyses we fit the oscillatory signals to a multiple damped harmonic function:

\[
f(t) = \sum_i A_i \exp(-\Gamma_i t) \sin(2\pi \nu_i t + \phi_i),
\]

with \(i\) denoting different phonon modes. Figure 3 compares the experimentally obtained oscillations with the fits and their GaP and Si phonon components at a fixed pump and probe polarization combination. Whereas the GaP oscillation component simply increases in the amplitude with increasing \(d\), the Si component flips its phase between \(d = 38\) and 48 nm. We note that the frequencies \(\nu_i\) and the dephasing rates \(\Gamma_i\) show no systematic dependence on the GaP thickness \(d\) and agree with those of the bulk GaP and Si within experimental errors.
FIG. 3. Comparison of oscillatory parts of \( \Delta R_{eo}/R_0 \) obtained at pump polarization I (black dots) with fits to eq. (1) (grey curves). Blue solid and orange dotted curves represent the fit components of Si and GaP LO phonons, respectively. Signals and fits for the bulk Si and GaP are also shown for comparison. Traces are offset for clarity.

Figure 4 plots the GaP and Si phonon amplitudes, \( A_{GaP} \) and \( A_{Si} \), as a function of pump polarization angle \( \theta \) from the [110] axis of the Si substrate. Here we restrict the initial phase \( \phi_i \) around zero and allow \( A_i \) to take a positive or negative value to represent the phase flip. The Si phonon amplitude (top panel of Fig. 4) always follows a cosine function of \( \theta \):

\[
A_{Si}(d, \theta) = -B_{Si}(d) \cos 2\theta, \tag{2}
\]

for all the GaP/Si samples as well as for the bulk Si. This is the manifest of the generation of the coherent LO phonons via impulsive stimulated Raman scattering (ISRS), as described by eq. (A3) in Appendix A. The GaP phonon amplitude for the bulk GaP, plotted with an orange broken curve in the bottom panel of Fig. 4, shows a similar \( \theta \)-dependence, indicating an ISRS-generation in the present below-bandgap excitation condition [24].

The GaP phonon amplitude for the GaP/Si samples, by contrast, is described more appropriately with an additional \( \theta \)-independent term by:

\[
A_{GaP}(d, \theta) = C_{GaP}(d) - B_{GaP}(d) \cos 2\theta. \tag{3}
\]

A similar \( \theta \)-independent contribution was also observed for bulk GaP upon above-bandgap photoexcitation, and was attributed to the ultrafast screening of the dc field in the surface depletion region by photoexcited carriers [24].

In a cubic crystal such as GaP, the driving force for this transient depletion field screening (TDFS) mechanism is independent of the pump polarization, as described by eq. (A4) in Appendix A. Accordingly, we attribute the \( \theta \)-independent term \( C_{GaP} \) of the GaP/Si samples to the TDFS-driven LO phonons. Because the phonon amplitude \( A_{GaP} \) increases linearly with increasing pump fluence for all the GaP/Si samples (not shown in Figure), it is unlikely that the photocarriers are created via a two-photon transition within the GaP layer. The more likely source of the charge carrier is via the injection at the heterointerface, either from the Si substrate or from the interface electronic states reported in the previous SHG study [25].

Figure 5 summarizes the \( d \)-dependence of the amplitude components \( B_i \) and \( C_i \) obtained from fitting \( A_i(\theta) \) to eq. (3). The components for the GaP LO mode, \( B_{GaP} \) and \( C_{GaP} \), increase in parallel with increasing \( d \) up to 38 nm. At \( d=48 \) nm, \( B_{GaP} \) becomes several times larger than \( C_{GaP} \), indicating that the bulk-like ISRS overwhelms the interface-specific TDFS for the thickest GaP layer. By contrast, the component for the Si LO mode, \( B_{Si} \), decreases almost monotonically with increasing \( d \) up to 38 nm. At \( d=48 \) nm it takes a negative value, denoting a phase flip of the coherent oscillation. We will discuss the origin of this peculiar behavior in Sect. IV.

In order to examine the carrier dynamics that can possibly contribute to the TDFS, we also measure the transient reflectivity \( \Delta R/R_0 \). Black curves in Fig. 6a compare the as-measured reflectivity changes of the GaP/Si samples with that of bulk Si. The signal from the bulk Si
FIG. 5. $\theta$-dependent ($B$, filled triangles) and $\theta$-independent ($C$, empty circles) amplitude components of GaP (a) and Si (b) LO phonons, as a function of GaP layer thickness $d$. The components are normalized by $B_{Si}$ of bulk Si ($d = 0$ nm). Solid curves in (a) and (b) represent $P_1$ and $P_2$ calculated with eqs. (9) and (10). Red broken and black chain curves in (b) represent $T_0$ given by eq. (5) and $T_0P_2$, respectively. $P_1$ and $P_2$ are normalized by $P_2(d = 0)$, whereas $T_0$ is normalized by $T_0(d = 0)$.

shows a step-function-like drop at $t = 0$, followed by very little recovery in the present time window. The response is in good agreement with previous reports [28, 29] and can be attributed to the free carrier excitation across the indirect bandgap, followed by their diffusion and recombination at the surface. Bulk GaP, by contrast, exhibits no detectable change under the present photoexcitation condition and its signal is therefore not shown.

Transient reflectivity traces of the GaP/Si samples appear in qualitative contrast to those of the bulk Si and GaP, indicating the carrier dynamics that is characteristic to the GaP/Si heterointerface. For $d = 8$ nm, the reflectivity signal shows an abrupt drop at $t \approx 0$ that is somewhat similar to the bulk Si, though the subsequent recovery is more distinct. The signals from the thicker ($d \geq 18$ nm) GaP/Si, by contrast, show an initial abrupt drop or rise depending on $d$, followed by a sub-picosecond increase and then a slower decrease. The height of the initial abrupt drop or rise, which we represent with the transient reflectivity at $t = 0.5$ ps, exhibits a peculiar $d$-dependence, as shown in Fig. 6b. It starts from a negative value and increases monotonically with increasing $d$ until it reaches a positive value. This trend is very similar to that of the Si LO phonon, $B_{Si}$, if we normalize the initial step height by that of the bulk Si.

FIG. 6. (a) As-measured transient reflectivity signals $\Delta R/R_0$ (grey curves) of GaP/Si(001) and bulk Si. Pump and probe lights are polarized along the [110] axes of the Si substrate. Incident pump density is 0.25 mJ/cm$^2$. Vertical line indicates $t = 0.5$ ps, and arrows indicate the initial step height $\Delta R(t = 0.5\text{ps})/R_0$ for selected traces. Blue broken and red solid curves indicate the contributions from the substrate and the interface given by eqs. (11) and (12). (b) Initial step height $\Delta R(t = 0.5\text{ps})/R_0$ as a function of GaP layer thickness $d$. Solid and chain curves represent the $d$-dependence of $P_2$ and $T_0P_2$. Curves are scaled to $\Delta R(t = 0.5\text{ps})/R_0$ of bulk Si.
FIG. 7. Schematic illustration of the transmission and reflection of probe electric field that is incident on a GaP/Si interface. $E_i$: incident wave, $E_{jr}$ and $E_{jt}$: reflected and transmitted wave at $j$-th interface. Incidence angle is exaggerated for clarity.

IV. DISCUSSION

In the previous Section we have seen that the phonon- and the carrier-induced signals from the Si substrate exhibit similar oscillatory behaviors as a function of overlayer thickness. The observations hint at the involvement of the oscillating electric field of the laser pulse, which has a wavelength of $\sim 200$ nm in the semiconductors. In this Section we theoretically model the effect of the GaP overlayer on the pump-induced reflectivity signal from the buried Si substrate by explicitly taking into account the light carrier wave.

We first examine the effect of the GaP overlayer on the pump light incoming to the Si substrate, whose details are described in Appendix B. We assume a light pulse, whose electric field in air is described by:

$$E_i(z,t) = \tilde{E}_i(z,t) e^{i(k_0 z - \omega_0 t)},$$

being incident on a GaP/Si heterointerface from the normal direction. At the air/GaP ($z = 0$) and GaP/Si ($z = d$) interfaces, the light pulse is partially reflected and partially transmitted, as schematically shown in Fig. 7. Taking into the multiple reflections, the transmittance of the heterointerface, or the intensity ratio of the light penetrating into Si to the incident light, can be expressed by:

$$T_0(d) = \frac{(1 - r_{01}^2)(1 - r_{12}^2)}{1 + r_{01}^2 r_{12}^2 + 2r_{01} r_{12} \cos 2n_1 k_0 d},$$

representing the reflection coefficients for the light wave coming from air into GaP and from GaP into Si, respectively. $n_1$ and $n_2$ are the refractive indices of GaP and Si.

Equation (5) implies that the pump intensity in the Si substrate can depend on the overlayer thickness $d$ due to the interference among different optical paths ($E_{2t}, E_{4t}, E_{6t}, \cdots$ in Fig. 7). This is the same principle as used for anti-reflection coatings on optics surfaces. In the present case of GaP/Si, however, the contribution from the $d$-dependent term is insignificant, because $r_{12}$ is small as a consequence of comparable refractive indices of GaP ($n_1=3.18$) and Si ($n_2=3.68$). The pump intensity in Si is modified no more than by $15\%$ by varying $d$, as shown with a red broken curve in Fig. 5b, which by itself cannot explain the drastic thickness-dependence of the Si phonon and carrier signals observed in our experiments.

Next we examine the effect of the GaP overlayer on the probe light, whose detail is described in Appendix C. The overlayer similarly modifies the reflectance of the interface:

$$R_0(d) = \frac{r_{01}^2 + r_{12}^2 + 2r_{01} r_{12} \cos 2n_1 k_0 d}{1 + r_{01}^2 r_{12}^2 + 2r_{01} r_{12} \cos 2n_1 k_0 d}.$$ (7)

We assume a separate pump pulse induces small disturbances $\delta n_1(z, t)$ and $\delta n_2(z, t)$ in the refractive indices of the GaP layer and the Si substrate. Coherent phonons in the respective semiconductors modulate the refractive indices periodically as a function of $t$, whereas photoexcited carriers in Si induce a step-function-like change in $n_2$. In either case, the transient reflectivity can be expressed by taking its derivatives with respect to the refractive indices:

$$\frac{\Delta R}{R_0} = \frac{1}{R_0} \left( \frac{\partial R_0}{\partial n_1} \delta n_1 + \frac{\partial R_0}{\partial n_2} \delta n_2 \right) \equiv P_1 \delta n_1 + P_2 \delta n_2.$$ (8)

The first and second terms of eq. (8) represent the pump-induced reflectivity changes contributed by the GaP overlayer and by the Si substrate. They can be given respectively by:
Solid curves in Fig. 5 show the $d$-dependences of the pump-induced reflectivity changes given by eqs. (9) and (10). The calculations reasonably reproduce the experimentally obtained Si and GaP phonon signals, $B_{\text{Si}}$ and $B_{\text{GaP}}$, of the GaP/Si samples. Equation (10) also reproduces the $d$-dependence of the initial step height, $\Delta R(t = 0.5 \text{ps})/R_0$, as shown in Fig. 6b, indicating that the initial drop/rise arises mostly (but not entirely) from the photoexcitation in the Si substrate. Taking into account the $d$-dependence of the pump intensity (eq. (5)) introduces only a minor correction, as shown with chain curves in Figs. 5b and 6b. The agreements confirm that the interference effect on the reflected probe light is the origin of the oscillatory behavior of the transient reflectivity signal from the Si substrate, whether it is associated with phonons or carriers.

We can now use eq. (10) to decompose the carrier-induced signal into the substrate- and interface-contributions. We can express the Si substrate contribution with:

$$
P_1(d)\delta n_1 = \frac{1}{R_0} \left( \frac{\partial R_0}{\partial n_{\text{Si}}} \frac{\partial \delta n_1}{\partial n_{\text{Si}}} + \frac{\partial R_0}{\partial n_{\text{Si}}} \frac{\partial \delta n_1}{\partial n_{\text{Si}}} + \frac{\partial R_0}{\partial n_{\text{Si}}} \frac{\partial (\delta n_1 / d)}{\partial n_{\text{Si}}} \right) \delta n_1
$$

$$
= \frac{2(r_{01}^2 + r_{12}^2 - 2r_{01}r_{12}\cos 2\pi k_0 d)(1 + r_{01}^2 r_{12}^2 + 2r_{01}r_{12}\cos 2\pi k_0 d)}{2n_1}
\times \left[ \frac{r_{01}(1 + r_{12}^2) - r_{12}(1 + r_{01}^2)}{2n_1}(1 - \cos 2\pi k_0 d) + 2k_0 r_{01} r_{12} \sin 2\pi k_0 d \right] \delta n_1,
$$

and

$$
P_2(d)\delta n_2 = \frac{1}{R_0} \frac{\partial R_0}{\partial r_{12}} \frac{\partial \delta n_2}{\partial r_{12}}
$$

$$
= \frac{2r_{12}(1 - r_{01}^4) + 2r_{01}(1 + r_{12}^2)(1 + r_{01}^2) \cos 2\pi k_0 d}{(r_{01}^2 + r_{12}^2 + 2r_{01}r_{12}\cos 2\pi k_0 d)(1 + r_{01}^2 r_{12}^2 + 2r_{01}r_{12}\cos 2\pi k_0 d)} \times \frac{-2n_1}{(n_1 + n_2)^2} \delta n_2.
$$

Blue broken and red solid curves in Fig. 6a show the substrate- and interface-contributions given by eqs. (11) and (12), respectively. We see that $\Delta R_{\text{int}}(t, d)/R_0$ for $d = 8 \text{ nm}$ exhibits an abrupt drop at $t \sim 0$, followed by a bi-exponential increase with time constants $\tau_{\text{fast}}=1.6 \text{ ps}$ and $\tau_{\text{slow}}=43 \text{ ps}$ toward a positive value. The slow time constant is comparable to that of the bulk Si, 38 ps, and can be interpreted in the similar relaxation processes involving the carrier diffusion within the Si substrate and their recombination at the GaP/Si interface. The larger amplitude of this slow relaxation indicates higher density of the recombination centers at the GaP/Si heterointerface than at the naturally oxidized surface of the bulk Si. The fast time constant is close to the delayed rise time observed in the previous SHG study [25] and may be associated with the carrier injection from the occupied interface electronic state.

For the overgrown GaP layers ($d \geq 18 \text{ nm}$), $\Delta R_{\text{Si}}(t, d)/R_0$ consists of a sub-picosecond rise followed by a monotonic decrease on $\sim 30 \text{ ps}$ time scale. The initial rise can be attributed to the carrier injection into the GaP overlayer, which would lead to the sudden screening of the dc electric field and thereby TDFS generation of the coherent phonons. The injection should occur sufficiently fast ($< 50 \text{ fs}$) in order to give a driving force for the coherent phonons. The time scale of the following slow decrease is independent of $d$ and comparable to those of $d = 8 \text{ nm}$ GaP/Si and bulk the Si, suggesting a similar recombination process at the heterointerface.

The present study alone cannot give a conclusive evidence for the dominant source of the carriers injected into the GaP layer. On one hand, an early theoretical study on (001)-oriented Si/GaP interface predicted a type-I band alignment with the valence band offset $\Delta E_v=0.8 \text{ eV}$ [30]. Photoemission experiments [31, 32] found comparable or larger $\Delta E_v$. This would give sufficient excess energy for the carriers photoexcited in Si with a 1.5-eV light to overcome the conduction band offset and to be injected into GaP. On the other, the previous SHG study [25] indicated the occupied interface electronic state whose energy is close to the valence band maximum of Si. The transition from the interface state exhibited a resonance peak at 1.4 eV, which is not far from the photon energy employed in the present study. To unambiguously determine the injection pathways we are performing further transient reflectivity experiments on the GaP/Si heterointerfaces with a tunable pump light source, which will be reported in a separate publication.

V. CONCLUSION

We investigated the carrier- and phonon-dynamics of the lattice-matched GaP/Si(001) interfaces upon below-bandgap photoexcitation of GaP by means of pump-
probe reflectivity measurements. We demonstrated the contribution of a buried Si substrate to the transient reflectivity signal, whether it is of electronic or phononic origin, to exhibit an oscillatory dependence on the thickness of an optically transparent GaP overlayer. The oscillatory behavior was quantitatively explained in terms of the interference of the probe light reflected at the heterointerface, whereas the interference effect on the pump intensity penetrating into the Si substrate was found to be minor in the present case. Based on this finding we extracted the interface contribution in the carrier-induced transient reflectivity traces. The obtained signals clearly indicated ultrafast injection of charge carriers into the overlayer, which was in agreement with the polarization-dependence of the coherent LO phonon of GaP. The knowledge obtained in the present study can be also applied to the quantitative analyses of transient reflectivity signals for wide varieties of buried semiconductor heterointerfaces.

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of photoexcited electrons and holes in the surface deple-
tion mechanism [3, 24]. In the TDFS mechanism, separation
of charge carriers can contribute to the coherent phonon
in Fig. 2b.

\[ F_{z}^{\text{TDFS}}(t) = - \frac{e^*}{\varepsilon_\infty \varepsilon_0} \int_{-\infty}^{t} dt' J_z(t'). \]  

(A4)

In a cubic crystal whose optical absorption is isotropic
within the \{001\} plane, the driving force is independent of
the pump polarization.

The coherent phonons can be observed as a peri-
odic modulation of reflectivity at the zone-center optical
phonon frequency. A nuclear displacement \( Q \) associated
with the LO phonon oscillation induces a change in re-
 sistivity \( R \) through the refractive index \( n \) and the sus-
ceptibility \( \chi \). In a first-order approximation the change
\( \Delta R \) is given by [3]:

\[ \Delta R = \frac{\partial R}{\partial n} \Delta n \simeq \frac{\partial R}{\partial \chi} \frac{\partial \chi}{\partial Q} \Delta Q. \]  

(A5)

Here \( \frac{\partial \chi}{\partial Q} = R \) is the first-order Raman tensor given
in eq. (A2). Equation (A5) implies that only Raman
active phonons can be detected in transient reflectivity,
and that the phonon signal depends on the probe light
polarization angle \( \theta \) in the same manner as the pump
light described in eq. (A3).

**Appendix B: Reflection and transmission of light at
heterointerface**

In this Section we consider the transmission and reflection
of light at a GaP/Si heterointerface, which consists of
a GaP layer of thickness \( d \) on top of a semi-ininitely
thick Si substrate, as schematically illustrated in Fig. 7.
We assume a light pulse, whose electric field in air is
expressed by:

\[ E_i(z, t) = \tilde{E}_i(z, t) e^{i(k_0 z - \omega_0 t)}, \]  

(B1)

is incident from the normal direction. Here \( \tilde{E}_i(z, t) \) and
\( \exp[i(k_0 z - \omega_0 t)] \) represent the slowly varying envelope
function and the fast varying carrier wave. \( z \) and \( t \)
represent the distance from the air/GaP interface and time.
\( \omega_0 \) and \( k_0 = \omega_0/c \) denote the light wave frequency and the
wavevector. Hereafter we approximate the slowly vary-
ing envelope function in eq. (B1) as a time-independent
constant.

At the air/GaP interface at \( z = 0 \), the light pulse is
partially reflected and partially transmitted into the GaP
layer. The reflected and transmitted light waves can be
expressed by:

\[ E_{1r}(z, t) = \tilde{E}_{1r}(z, t) e^{-(k_0 z - \omega_0 t)}, \]  

\[ E_{1t}(z, t) = \tilde{E}_{1t}(z, t) e^{i(\tilde{n}_1, k_0 z - \omega_0 t)}, \]  

(B2)

where \( \tilde{n}_1 = n_1 + i \kappa_1 \) is the refractive index of GaP. We use
\( n_1 = 3.18 \) and \( \kappa_1 = 0 \) at wavelength \( \lambda_0 = 2\pi/k_0 = 815 \text{ nm} \)
in the following calculations. We apply the boundary
condition that the in-plane components of the elec-
tric and magnetic fields are continuous, and obtain the

**Appendix A: Coherent phonon generation and
detection in bulk Si and GaP**

Illumination with a femtosecond laser pulse can induce
cohert optical phonons in semiconductors. The gener-
ation mechanism can depend on the semiconductor and
the excitation photon energy. When the photon energy is
below the fundamental bandgap, or when the semi-
conductor is transparent to the excitation light, impul-
sive stimulated Raman scattering (ISRS) [33] is the only
known generation mechanism. In ISRS, a broadband
femtosecond optical pulse offers multiple pairs of photons
required for the stimulated process. The driving force \( F \)
depends on the polarization of the optical electric field \( E \)
through a third-rank Raman tensor \( \mathcal{R}_{jkl} \) given
in the form of [1]:

\[ F_j^{\text{ISRS}}(t) = \mathcal{R}_{jkl} E_k(t) E_l(t), \]  

(A1)

where \( j, k, l \) denote the Cartesian coordinates. The Ra-
man tensor of diamond- and zinc blende-structured crys-
tals is given in the form of [1]:

\[ \mathcal{R}_{xkl} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \mathcal{R}_{ykl} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ a & 0 & 0 \end{pmatrix}, \quad \mathcal{R}_{zkl} = \begin{pmatrix} 0 & a & 0 \\ a & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \]  

(A2)

In the back-reflection from the (001)-oriented surface, in
which the pump light polarization has no \( z \) component,
the driving force can be reduced to:

\[ F_z^{\text{ISRS}}(t) = \mathcal{R}_{xyz} E_x(t) E_y(t) + \mathcal{R}_{xyz} E_y(t) E_x(t), \]  

\[ = a |E(t)|^2 \sin 2\theta', \quad \text{where } \theta' = \theta - \pi/4 \]  

(A3)

with \( E \equiv |E| \cos \theta' \), \( E_y \equiv |E| \sin \theta' \), and \( \theta' = \theta - \pi/4 \) being the polarization angle from the [100] axis. The
driving force becomes maximum at \( \theta = 0 \) or \( \pi \), i.e.,
when the pump light is polarized along the [110] or [-110]
axis of the crystal. The direction of the driving force reverses
between these two polarizations, which explains the phase
flip of the coherent LO phonons of bulk GaP and Si shown
in Fig. 2b.

When the photon energy exceeds the bandgap, or when
the semiconductor is opaque, the ISRS generation of
cohert phonons can be resonantly enhanced in the simi-
lar manner as in spontaneous Raman scattering [34, 35].
In the case of a polar semiconductor such as GaP, tran-
sient depletion field screening (TDFS) can contribute in
addition [3, 24]. In the TDFS mechanism, separation
of photoexcited electrons and holes in the surface deple-
tion layer induces ultrafast drift-diffusion current \( J_z \) in
the surface normal direction and thereby offers a driving
force for the coherent polar phonons [3]:

\[ F_z^{\text{TDFS}}(t) = - \frac{e^*}{\varepsilon_\infty \varepsilon_0} \int_{-\infty}^{t} dt' J_z(t'). \]  

(A4)

The direction of the driving force reverses between
the two polarizations, which explains the phase
flip of the coherent LO phonons of bulk GaP and Si shown
in Fig. 2b.

The reflected and transmitted light waves can be
expressed by:

\[ E_{1r}(z, t) = \tilde{E}_{1r}(z, t) e^{-(k_0 z - \omega_0 t)}, \]  

\[ E_{1t}(z, t) = \tilde{E}_{1t}(z, t) e^{i(\tilde{n}_1, k_0 z - \omega_0 t)}, \]  

(B2)

where \( \tilde{n}_1 = n_1 + i \kappa_1 \) is the refractive index of GaP. We use
\( n_1 = 3.18 \) and \( \kappa_1 = 0 \) at wavelength \( \lambda_0 = 2\pi/k_0 = 815 \text{ nm} \)
in the following calculations. We apply the boundary
condition that the in-plane components of the elec-
tric and magnetic fields are continuous, and obtain the

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reflection and transmission coefficients for the light wave
incoming from air into GaP:

\[ r_{01} \equiv \frac{\tilde{E}_{1r}}{\tilde{E}_i} = \frac{1 - n_1}{1 + n_1} = -0.52, \]
\[ t_{01} \equiv \frac{\tilde{E}_{1t}}{\tilde{E}_i} = \frac{2}{1 + n_1} = 0.48. \] (B3)

The light wave transmitted into the GaP layer is again
partially reflected at the GaP/Si interface and partially
transmitted into the Si substrate. We describe the re-
lected and transmitted waves by:

\[ E_{2r}(z, t) = \tilde{E}_{2r}(z, t) e^{-i(n_1 k_0 (z - 2d) + \omega_0 t)}, \]
\[ E_{2t}(z, t) = \tilde{E}_{2t}(z, t) e^{i(n_1 k_0 d + \phi - k_0 (z - d) - \omega_0 t)}, \] (B4)

where \( \tilde{n}_2 = n_2 + ik_2 \) is the refractive index of Si,
with \( n_2 = 3.68 \) and \( k_2 = 0.006 \) at \( \lambda_0 = 815 \text{ nm} \) [26]. Equation (B4)
implies that the light wave gains a thickness-
dependent phase shift, \( \phi \equiv n_1 k_0 d \), while it crosses the
GaP layer once. We apply the boundary condition at
the GaP/Si interface and obtain the reflection and trans-
mission coefficients for the light wave incoming from GaP
into Si:

\[ r_{12} = \frac{\tilde{E}_{2r}}{\tilde{E}_{1t}} = \frac{n_1 - n_2}{n_1 + n_2} = -0.073, \]
\[ t_{12} = \frac{\tilde{E}_{2t}}{\tilde{E}_{1t}} = \frac{2n_1}{n_1 + n_2} = 0.93. \] (B5)

Here we neglect the small optical absorption in Si and
approximate \( \tilde{n}_2 \approx n_2 \). After the reflection at the GaP/Si
interface, the light wave is again partially reflected at the
GaP/air interface and partially transmitted into air. We
describe the reflected and transmitted waves by:

\[ E_{3r}(z, t) = \tilde{E}_{3r}(z, t) e^{i(n_1 k_0 z - \omega_0 t + 2\phi)}, \]
\[ E_{3t}(z, t) = \tilde{E}_{3t}(z, t) e^{i(n_1 k_0 z + \omega_0 t + 2\phi)}. \] (B6)

We apply the similar boundary condition and obtain the
reflection and transmission coefficients for outgoing wave
from GaP into air:

\[ r_{10} = \frac{\tilde{E}_{3r}}{\tilde{E}_{3t}} = \frac{n_1 - 1}{n_1 + 1} = 0.52, \]
\[ t_{10} = \frac{\tilde{E}_{3t}}{\tilde{E}_{3r}} = \frac{2n_1}{n_1 + 1} = 1.52. \] (B7)

The amplitude ratio \( r_0 \) of the outgoing wave into air
to the incident wave can be given by the sum of multiple
reflection pathways:

\[ r_0 = \frac{\tilde{E}_{1r} + \tilde{E}_{3r} e^{2i\phi} + \tilde{E}_{5r} e^{4i\phi} + \tilde{E}_{7r} e^{6i\phi} + \cdots}{\tilde{E}_i} \]
\[ = r_{10} + t_{10} r_{12} t_{10} e^{2i\phi} (1 + r_{10} r_{12} e^{2i\phi} + r_{10}^2 r_{12}^2 e^{4i\phi} + \cdots) \]
\[ = r_{10} + \frac{t_{10} r_{12} t_{10} e^{2i\phi}}{1 - r_{10} r_{12} e^{2i\phi}} \frac{r_{01} + r_{12} e^{2i\phi}}{1 + r_{01} r_{12} e^{2i\phi}}. \] (B8)

Here we use the relations \( t_{10} t_{10} = 1 + r_{10} r_{01} \) and \( r_{10} = -r_{01} \) derived from eqs. (B3) and (B7). Likewise, the
amplitude ratio \( t_0 \) of the incoming wave into Si to the
incident wave can be given by:

\[ t_0 = \frac{\tilde{E}_{2t} e^{2i\phi} + \tilde{E}_{4t} e^{3i\phi} + \tilde{E}_{6t} e^{5i\phi} + \cdots}{\tilde{E}_i} \]
\[ = t_{01} t_{12} e^{i\phi} (1 + r_{10} r_{12} e^{2i\phi} + r_{10}^2 r_{12}^2 e^{4i\phi} + \cdots) \]
\[ = \frac{t_{01} t_{12} e^{i\phi}}{1 - r_{10} r_{12} e^{2i\phi}. } \] (B9)

The reflectance, or the light intensity reflected into air,
can then be given by:

\[ R_0 = |t_0|^2 = \frac{|t_{01} t_{12} e^{2i\phi}|^2}{1 + r_{10} r_{12} e^{2i\phi}} \]
\[ = \frac{r_{10}^2 + r_{12}^2 + 2 r_{10} r_{12} \cos 2\phi}{1 + r_{10}^2 r_{12}^2 + 2 r_{10} r_{12} \cos 2\phi}. \] (B10)

The transmittance, or the light intensity transmitted into
the Si substrate, can be given by:

\[ T_0 = 1 - R_0 = \frac{(1 - r_{10}^2)(1 - r_{12}^2)}{1 - r_{10}^2 r_{12}^2 + 2 r_{10} r_{12} \cos 2\phi}. \] (B11)

Red dashed curve in Fig. 5b shows the transmittance
given by eq. B11 as a function of the GaP thickness \( d \).
The calculation indicates that the pump intensity trans-
mitted into Si is modified by no more than 15 % with
varying \( d \), which is too small to explain the experimental
\( d \)-dependences of \( B_{Si} \) plotted in the same figure.

Appendix C: Pump-induced changes in reflectivity
from GaP/Si heterointerface

In this Section we consider the pump-induced change
in the probe light wave reflected from the GaP/Si het-
nerointerface. We assume that a pump wave induces small
disturbances \( \delta n_1(z, t) \) and \( \delta n_2(z, t) \) in the refractive
indices \( n_1 \) and \( n_2 \) in the GaP layer (\( 0 < z < d \)) and in the Si
substrate (\( z > d \), respectively. Because the semicon-
ductors have very small or no absorption to the pump light,
we can approximate the disturbances to be independent
of the depth \( z \) in both GaP and Si and to depend only
on the time delay \( t \) between the pump and probe pulses.
Coherent phonons in the respective semiconductors mod-
ulate \( n_1 \) and \( n_2 \) periodically as a function of \( t \), whereas
photoexcited carriers in Si induces a step-function-like
change in \( n_2 \).

To determine the pump-induced change in the re-
fectance, we can take derivatives of the reflectance with
respect to the refractive indices:

\[ \frac{\Delta R}{R_0} = \frac{1}{R_0} \left( \frac{\partial R_0}{\partial n_1} \Delta n_1 + \frac{\partial R_0}{\partial n_2} \Delta n_2 \right). \] (C1)

A change in \( n_1 \) can modify \( r_{01}, \ r_{12} \) and \( \phi = n_1 k_0 d \),
whereas a change in \( n_2 \) can affect only \( r_{12} \). The first and
the second terms of eq. (C1) can therefore be expressed by:

\[ \frac{\Delta R}{R_0} = \frac{1}{R_0} \left( \frac{\partial R_0}{\partial n_1} \Delta n_1 + \frac{\partial R_0}{\partial n_2} \Delta n_2 \right). \] (C1)
\[ \frac{1}{R_0} \frac{\partial n_1}{\partial n_1} \delta n_1 = \frac{1}{R_0} \left( \frac{\partial R_0}{\partial r_{01}} \frac{\partial r_{01}}{\partial n_1} + \frac{\partial R_0}{\partial r_{12}} \frac{\partial r_{12}}{\partial n_1} + \frac{\partial R_0}{\partial \phi} \frac{\partial \phi}{\partial n_1} \right) \delta n_1. \]

\[ = \frac{2(r^2_{01} - 1)(1 - r^2_{12})}{(r^2_{01} + r^2_{12} + 2r_{01}r_{12}\cos 2\phi)(1 + r^2_{01}r^2_{12} + 2r_{01}r_{12}\cos 2\phi)} \times \frac{\left[ r_{01}(1 + r^2_{12}) - r_{12}(1 + r^2_{01}) \right]}{2n_1} (1 - \cos 2\phi) + 2k_0 d r_{01}r_{12}\sin 2\phi \delta n_1 \equiv P_1(d)\delta n_1. \]  

(C2)

\[ \frac{1}{R_0} \frac{\partial R_0}{\partial n_2} \delta n_2 = \frac{1}{R_0} \frac{\partial R_0}{\partial r_{12}} \frac{\partial r_{12}}{\partial n_2} \delta n_2 \]

\[ = \frac{2r_{12}(1 - r^4_{01}) + 2r_{01}(1 + r^2_{12})(1 - r^2_{01}) \cos 2\phi}{(r^2_{01} + r^2_{12} + 2r_{01}r_{12}\cos 2\phi)(1 + r^2_{01}r^2_{12} + 2r_{01}r_{12}\cos 2\phi)} \times \frac{-2n_1}{(n_1 + n_2)^2} \delta n_2 \equiv P_2(d)\delta n_2. \]  

(C3)

Here we use

\[ \frac{\partial R_0}{\partial r_{01}} = \frac{2r_{01}(1 - r^4_{12}) + 2r_{12}(r^2_{01} + 1)(1 - r^2_{12}) \cos 2\phi}{(1 + r^2_{01}r^2_{12} + 2r_{01}r_{12}\cos 2\phi)^2}, \]  

(C4)

\[ \frac{\partial R_0}{\partial r_{12}} = \frac{2r_{12}(1 - r^4_{01}) + 2r_{01}(1 + r^2_{12})(1 - r^2_{01}) \cos 2\phi}{(1 + r^2_{01}r^2_{12} + 2r_{01}r_{12}\cos 2\phi)^2}. \]  

(C5)

\[ \frac{\partial R_0}{\partial \phi} = \frac{-4r_{01}r_{12}(1 - r^2_{01})(1 - r^2_{12}) \sin 2\phi}{(1 + r^2_{01}r^2_{12} + 2r_{01}r_{12}\cos 2\phi)^2}, \]  

(C6)

\[ \frac{\partial r_{01}}{\partial n_1} = \frac{-2}{(1 + n_1)^2} = \frac{r^2_{01} - 1}{2n_1}, \]  

(C7)

\[ \frac{\partial r_{12}}{\partial n_1} = \frac{2n_2}{(n_1 + n_2)^2} = \frac{1 - r^2_{12}}{2n_1}, \]  

(C8)

\[ \frac{\partial \phi}{\partial n_1} = k_0 d, \]  

(C9)

\[ \frac{\partial r_{12}}{\partial n_2} = \frac{-2n_1}{(n_1 + n_2)^2} = \frac{r^2_{12} - 1}{2n_1}. \]  

(C10)

Equations (C2) and (C3) reproduce the experimental \( d \)-dependences of the phonon-induced reflectivity signals from GaP and Si, as shown in Fig. 5.