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Hemang Jani, Lingze Duan, "Wavelength-resolved pump-probe transient-reflectivity characterization of optoelectronic devices," Proc. SPIE 10916, Ultrafast Phenomena and Nanophotonics XXIII, 109161V (27 February 2019); doi: 10.1117/12.2508589

SPIE.

Event: SPIE OPTO, 2019, San Francisco, California, United States

Wavelength-resolved pump-probe transient-reflectivity characterization of optoelectronic devices

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ABSTRACT

Significant advance has been made over the last decade in the development of broadband optoelectronic devices based on novel technologies such as 2D materials, metamaterials, plasmonics, negative electron affinity photoemission, etc. Understanding carrier dynamics in such devices, especially carrier relaxation and transportation near device surfaces, requires time-resolved, broadband reflective spectroscopy with femtosecond temporal resolutions. Femtosecond pump-probe reflectivity measurement (PPRM) has long been used to study carrier dynamics in semiconductor devices. However, conventional PPRM lacks the necessary bandwidth and the ability to make spectroscopic measurement. In this presentation, we report the demonstration of wavelength-resolved transient reflectivity measurement using a ultrabroad-band few-cycle pump-probe system. The system allows device transient reflectivity to be mapped onto a two-dimensional space formed by time and wavelength, providing a comprehensive characterization of ultrafast carrier dynamics. Preliminary results based on a GaAs substrate and GaAs/AlGaAs layered structures have offered interesting insights into device dynamics that otherwise would not be clear. These results demonstrate the feasibility of performing wavelength-resolved transient reflectivity measurement and the effectiveness of this technique in characterizing broadband optoelectronic devices.

Keywords: Ultrafast Spectroscopy, Pump-probe reflectivity, Few-Cycle laser, Optoelectronics, GaAs/AlGaAs, Negative Electron Affinity, Carrier Dynamics, transient response

1. INTRODUCTION

There has been significant advances over the last decade in the development of novel technologies such as 2D materials, metamaterials, plasmonics, negative electron affinity (NEA) structures etc. Accompanying this technological revolution has emerged an equally expanding market of broadband ultrafast optoelectronic devices.[1–4] Understanding the ultrafast dynamics, such as carrier relaxation and transportation, in these devices is vital for maximizing their performances. More specifically, transient processes near devices surfaces are crucial for the operation of ultrafast devices such as NEA photocathodes[5] and photodetectors based 2D materials.[6] Probing these processes calls for sophisticated diagnostic techniques capable of making comprehensive characterization on these devices. One broad family of techniques that can meet such demands is optical reflectometry. By measuring changes in reflectivity off sample surfaces, this technique has proved very useful in deducing vital optical constants of the samples[7–9] and gaining insight in various time dependent processes.[10–12]

Here, we focus on one such technique - pump-probe reflectivity measurement (PPRM). PPRM is a versatile and well established characterization technique to study ultrafast dynamics in materials and devices.[13] However, depending on specific research goals, various groups have exploited PPRM systems either to make time-domain measurements[14,15] or time-resolved spectral domain measurements.[13,16] In this paper, we present preliminary results of wavelength-resolved pump-probe transient-reflectivity measurements with GaAs wafer and GaAs/AlGaAs layered structures. The work aims to showcase the feasibility of performing both time domain and spectral domain measurements simultaneously and with expected sensitivities.

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Ultrafast Phenomena and Nanophotonics XXIII, edited by Markus Betz, Abdulhakem Y. Elezzabi, Proc. of SPIE Vol. 10916, 109161V · © 2019 SPIE
CCC code: 0277-786X/19/$18 · doi: 10.1117/12.2508589

2. ULTRABROAD-BAND PUMP-PROBE TRANSIENT REFLECTOMETER

In a typical pump-probe transient reflectivity measurement, variations in surface reflectivity of a device sample is measured at various time delays following the optical excitation by energetic pump pulses.[14–21] For *only* time-domain PPRM, one measures the evolution of sample's *total* reflectivity as a function of the relative time delay between the pump and probe pulses.[19] On the other hand, by using a spectrometer at the system's output, PPRM can also perform spectroscopic measurements by measuring the *spectrum* of transient reflectivity at various time delays.[13] This addition has proved to be very useful for direct characterization of the dielectric function, offering insight into underlying physical processes.[22]

We have previously reported time-domain transient-reflectivity measurement of a GaAs/AlGaAs photocathode using a home-built, ultrabroad-band (300 nm), few-cycle (sub-10 fs) pump-probe system.[11] In the current report, we take one step further to add the capability of making wavelength-resolved measurement to the system. This is done by inserting a tunable bandpass filters in front of the photodetector to select the probe wavelength that is detected. By changing the center wavelength of the filter (typically with a 10-nm bandwidth), the same time-domain measurement can be made at a series of wavelengths. This offers an extra dimension to PPRM for comprehensive characterization of various opto-electronic devices.

3. EXPERIMENTAL RESULTS

As a proof-of-concept study, we have investigated wavelength-dependent transient reflectivity of GaAs substrates. We have also made a comparative study of GaAs/AlGaAs NEA photocahtodes with different doping structures. Similar time-domain studies on these samples have led to deeper understanding about carrier dynamics in such devices and have been used to distinguish the performance of different structures.[5,10,11]

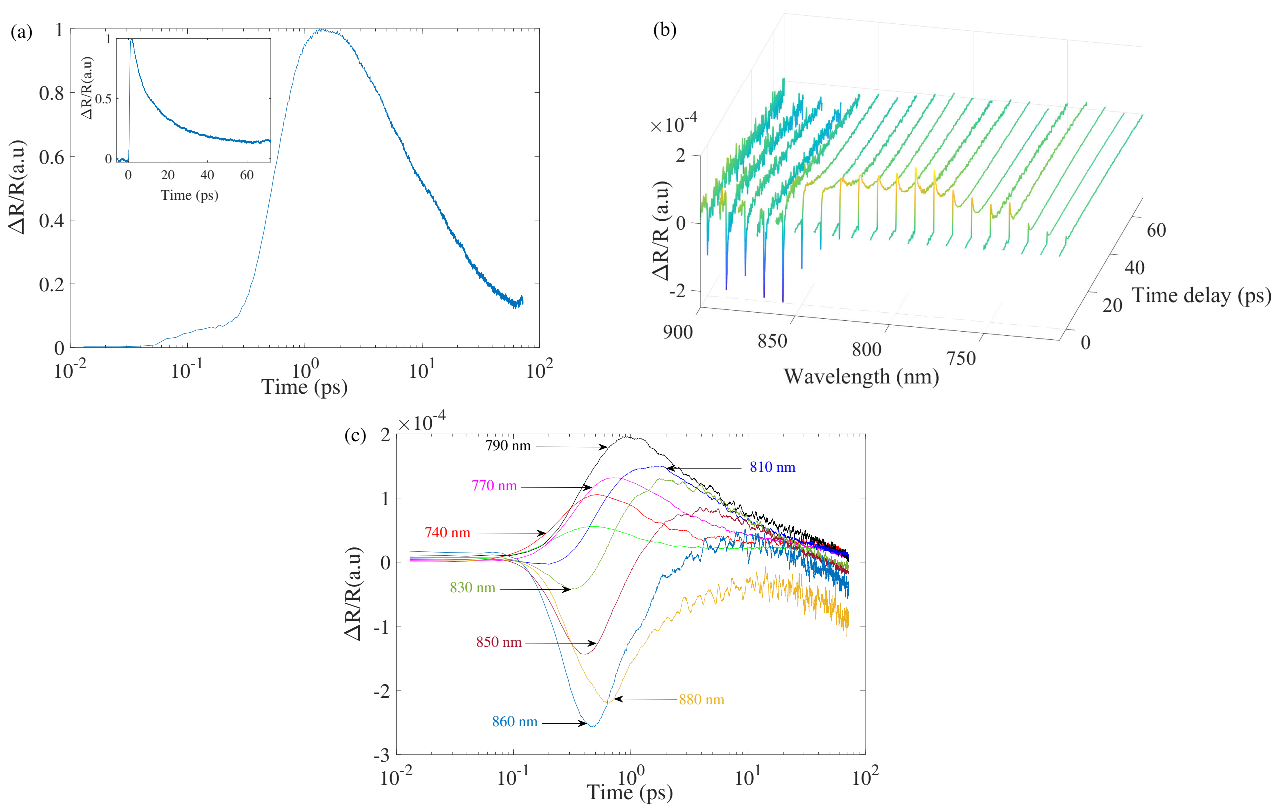


Figure 1. (a) Pump-probe reflectivity measurement of GaAs on a time scale of 0 to 72 ps. The inset figure is plotted on linear time scale. (b) 3D representation of transient reflectivity signal as a function of time delay and probe wavelength. (c) Wavelength-dependent transient reflectivity on logarithm time scale.

3.1 Observation of Non-Instantaneous Rise Time in Transient Reflectivity of GaAs

By measuring time-resolved reflectivity changes on sample surfaces, one can extract information about various ultrafast processes such as band filling (BF), band-gap renormalization (BGR), free-carrier absorption (FCA), Coulomb enhancement (CE), etc.[23] The vast majority of transient-reflectivity studies so far focus exclusively on the recovery dynamics of reflectivity upon an impulse-like excitation induced by the pump pulse.[21,23,24] The abrupt ascent (or descent) of reflectivity during or immediately after the pump pulse is normally treated as an instantaneous process due primarily to its extremely short rise time (e.g., <100 fs) and the limitation of system temporal resolution.[21] Recent theoretical work, however, has predicted that the rise time of transient reflectivity carries important information about the relative scales of various ultrafast effects.[23] In particular, it is shown that non-instantaneous rise time occurs when carrier cooling rate becomes comparable with carrier decay rate.[23] That is, BF begins to contribute to the change of refractive index, which otherwise is dominated by BGR.[21] Such predictions can only be verified with temporally-resolved measurement of the initial edge of transient reflectivity. Here, we report experimental observation of non-instantaneous rise time in transient reflectivity of an Si-doped GaAs (110) wafer with a carrier concentration of about $3 \times 10^{18} cm^{-3}$.

Fig. 1(a) shows the overall transient-reflectivity trace, which is obtained without using any filter. The inset is plotted on a typical linear time scale, whereas the main panel is plotted on a logarithm time scale. The logarithm scale allows the steep rising edge to be clearly revealed. A mild increase in reflectivity is seen in the first 200 fs, followed by a more rapid rise. The reflectivity reaches peak at slightly more than 1 ps and then begins to decrease. Since the overall reflectivity is a lumped sum of the responses over a broad range of wavelengths, it is instructive to analyze the dvice response at different wavelengths (or photon energies) in order to gain deeper insight into the underlying physics. Fig. 1(b) shows a series of transient-reflectivity measurements obtained over a wavelength span from 710 nm to 890 nm. At each wavelength, a 10-nm bandpass filter is used right in front of the photodetector. In addition, the probe beam spectrum is simultaneously measured and the pump-probe trace at each wavelength is normalized to the probe spectral density at the same wavelength to obtain reflectivity. The results show different reflectivity behaviors at different wavelengths. In particular, the reflectivity shows a positive change at wavelengths above the band gap and a negative change at wavelengths below the band gap. At wavelengths right around the band gap, a bipolar activity is observed. These features qualitatively agree with theoretical predictions.[23] The rise times of reflectivity at individual wavelengths are better revealed by plotting some of the traces in Fig. 1(b) on logarithm time scales, which is shown in Fig. 1(c). A striking feature of the plot is the difference in rise time for different wavelengths, ranging from about 300 fs to well over 1 ps. This can be an indication of an evolving dielectric function, which may lend insights into the relative scales of various ultrafast processes. This data is currently being analyzed based on theoretical models.

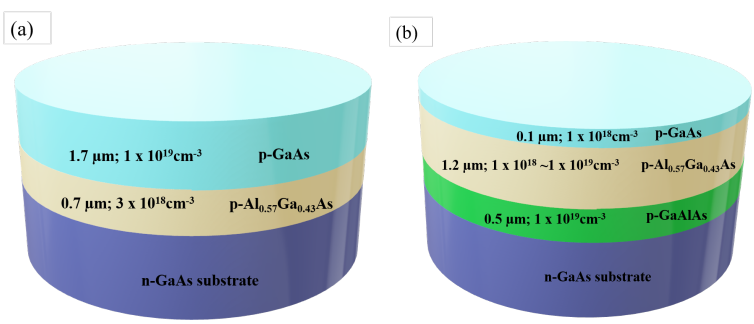


Figure 2. (a)) Device#1: uniform-doped p-GaAs as active layer.(b) Device #2: gradient-doped p-$Al_{0.63}Ga_{0.37}As$ as active layer.

3.2 Time-resolved spectral domain measurements of GaAs/AlGaAs layered structures

In this section, we expand the application of wavelength-resolved PPRM to GaAs/AlGaAs layered structures, whereby confirming the robustness of our PPRM system on a wide variety of applications. The devices under investigation are two NEA photocathodes. Their doping structures are shown in Fig 2. Specifically, Device#1 Fig. 2(a)) has a uniform-doped active (GaAs) layer with a doping concentration of $1 \times 10^{19} cm^{-3}$, whereas

Device#2 (Fig. 2(b)) has a gradient-doped active layer with a doping concentration changing from $1 \times 10^{18} cm^{-3}$ at the surface to $1 \times 10^{19} cm^{-3}$ near the AlGaAs buffer layer.

By changing the center wavelength of the tunable filter, transient reflectivity measurements are taken over a series of wavelengths on both devices. The results are summarized in Fig. 3(a) and 3(b). Similar to the GaAs substrate, bipolar behaviors of the reflectivity are observed with both devices, i.e. $\Delta R/R$ displays positive changes at some wavelengths and negative changes at other wavelengths. Carrier decay behaviors, which are indicated by the tails of the reflectivity traces, also appear to be different for different wavelengths, indicating rich physics underlying the involved processes.

Further insight can be gained by plotting the traces in Fig. 3 into waterfall graphs with a logarithm time scale, as shown in Fig. 4. Such time-wavelength mapping of transient reflectivity offers a comprehensive picture of the behaviors of dielectric function after optical excitation by femtosecond pulses. Comparing Fig. 4(a) and Fig. 4(b), it is evident that photoelectron accumulation on device surface occurs quicker in the gradient-doped photocathode than in the uniform-doped photocathode (indicated by the earlier appearance of the bipolar feature near the GaAs band gap in Device#2). This confirms the projected advantage of gradient-doped photocathodes, i.e. the doping profile can assist carrier transportation to enhance device performance. Such complex carrier dynamics would not be revealed without the ability to make wavelength-resolved measurements.

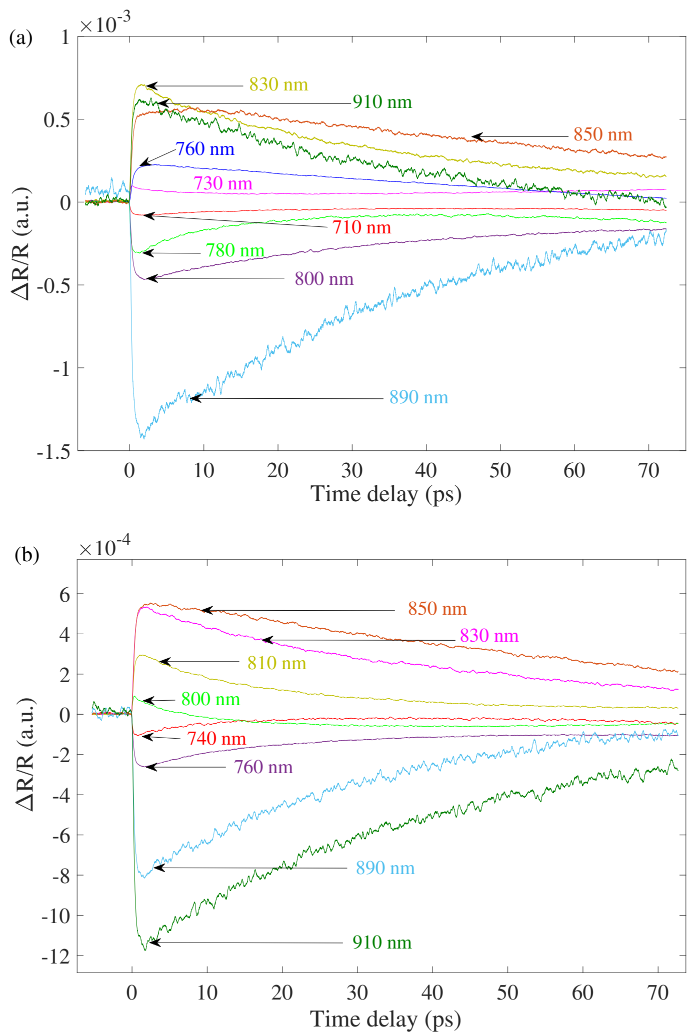
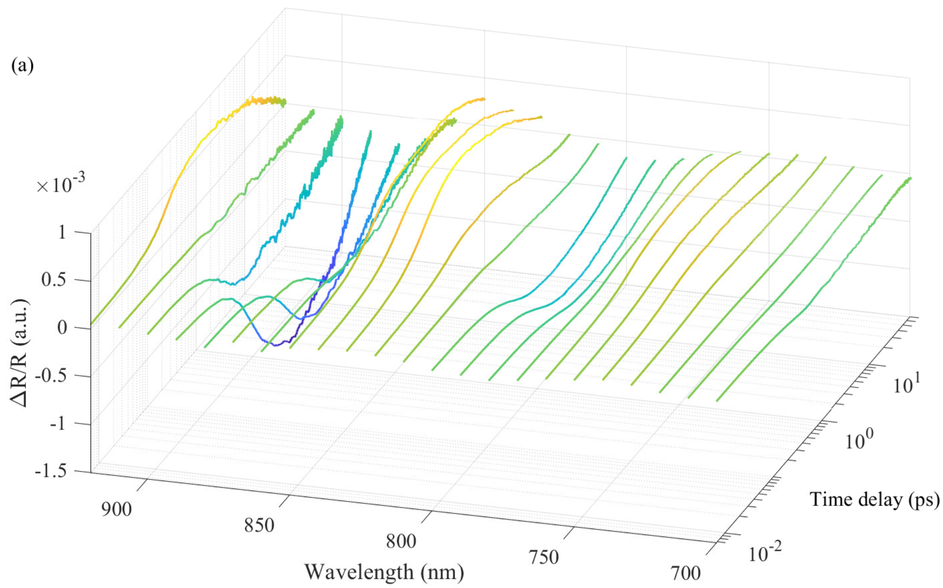


Figure 3. Wavelength resolved transient reflectivity (a) Device#1. (b) Device#2.



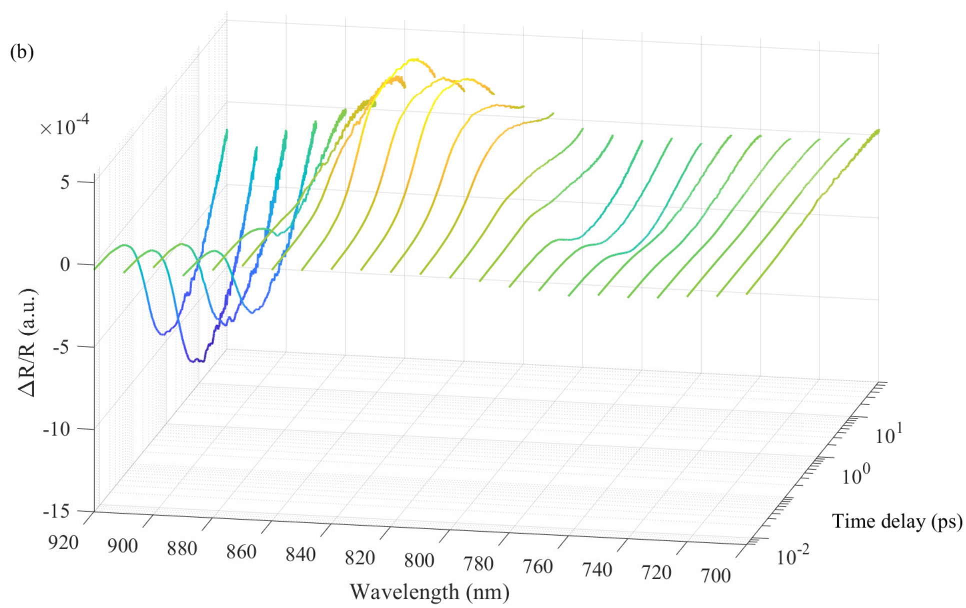


Figure 4. Time-Wavelength mapping of transient reflectivity (a) Waterfall plot for Device#1 with logarithmic time scale. (b) Waterfall plot for Device#2 with logarithmic time scale.

4. CONCLUSION

In conclusion, we report here wavelength-resolved pump-probe transient-reflectivity measurements with a GaAs substrate and GaAs/AlGaAs NEA photocathodes. In both cases, the technique offers insights into transient carrier dynamics that otherwise would not be available. These results demonstrate the feasibility of performing both time domain and spectral domain PPRM simultaneously and the effectiveness of such an approach in characterizing broadband optoelectronic devices.

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation (NSF) under Grant ECCS-1254902 and ECCS-1606836.

The authors acknowledge the key support to this research by Dr. Yunsheng Qian of Nanjing Institute of Science and Technology, China, and Dr. Liang Chen of Institute of Optical & Electronic Technology, China Jiliang University, China, who supplied the photocathode devices.

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