Real-Space Imaging of Photogenerated Carrier Dynamics via 4D Scanning Ultrafast Electron Microscopy

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Abstract

Understanding the behavior of photogenerated carriers at semiconductor surfaces is important to improve photovoltaic and optoelectronic devices. In this project, we used 4D Scanning Ultrafast Electron Microscopy (4D-SUEM) to image the surface dynamics of GaAs and CdTe in real space and time. Then we analyzed the time-resolved images using intensity profiles and two-component exponential fitting to study their spatial and temporal responses after optical excitation. GaAs showed a dark photo-induced contrast with time constants of 4.69 ps and 256.65 ps, while CdTe showed a bright contrast with time constants of 5.72 ps and 645.09 ps. The different responses and time constants demonstrate distinct surface charge dynamics in the two materials. Overall, our results show that 4D-SUEM provides a useful approach for directly studying ultrafast photoinduced surface processes in semiconductor materials.

Date: August 25, 2026

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1 Introduction

The increasing global demand for clean and sustainable energy has accelerated research into advanced semiconductor materials for renewable energy technologies, particularly photovoltaic and optoelectronic devices.[1] The efficiency of a photovoltaic cell is largely determined by its generation, recombination and transport properties. These physical properties occur at a time span of femtoseconds to nanoseconds [2]. It has become essential to investigate these carrier dynamics with high spatial and temporal resolution since they directly influence device efficiency, which motivated the development of 4D Scanning Ultrafast Electron Microscopy (SUEM) that offers a combination of the nanometer scale spatial resolution of a Scanning Electron Microscope (SEM) synchronized with a femtosecond pumped laser for sample excitation, adding ultrafast pump-probe techniques that allow direct imaging of photogenerated carrier dynamics in both space and time [3][4]. While there are other techniques with good spatiotemporal resolution that can be used to image photovoltaic cells including Transient Absorption/Reflectivity Microscopy (TAM/TRM) and Time-resolved Photoemission Electron Microscopy (TR-PEEM). They tend to measure mixed bulk and surface carrier responses because the optical pump probe penetrates deeply into the sample unlike a SUEM which probes only the surface of the sample [5].

Gallium Arsenide (GaAs) and Cadmium Telluride (CdTe) are important direct bandgap semiconductors, with band gap energies of approximately 1.42 and 1.5 eV, respectively, widely used in photovoltaic and optoelectronic devices because of their excellent optoelectronic properties. However, the performance of GaAs and CdTe-based devices is strongly influenced by carrier behavior near the surface, where surface band bending, defects, and recombination affect carrier diffusion and lifetime, while surface oxidation further affects the surface carrier dynamics in CdTe. Therefore, understanding the spatial and temporal evolution of photogenerated carriers at GaAs and CdTe surfaces is important for improving device efficiency [6][1][7].

In this study, using 4D-SUEM, we characterized the spatial evolution and temporal dynamics of photogenerated carriers in GaAs and CdTe, providing insights into their diffusion and recombination behavior by analyzing time-resolved secondary electron images using Gaussian fitting and intensity analysis. This analysis provides a deeper understanding of the behavior of semiconductor materials for optoelectronic and photovoltaic applications, contributing to the development of more efficient renewable energy technologies.

2 Methodology

The experimental measurements were carried out using a Scanning Ultrafast Electron Microscopy (SUEM) system, shown in Figure 1, which combines a modified FEI Quanta 650 SEM with a ClarkMXR femtosecond fiber laser. The laser produces infrared pulses centered at 1030 nm with a pulse duration of 270 fs. The infrared beam is split by a beam splitter, entering a second harmonic generator and a third harmonic generator, that convert the infrared beam into a 515 nm wavelength green beam and a 343 nm wavelength ultraviolet beam, respectively. The laser repetition rate can be adjusted according to the experimental conditions; however, all measurements in this work were performed at 8.33 MHz, corresponding to the pulse interval of 120 ns. This interval allows the excited carriers to relax before the arrival of the subsequent excitation pulse [3][8].

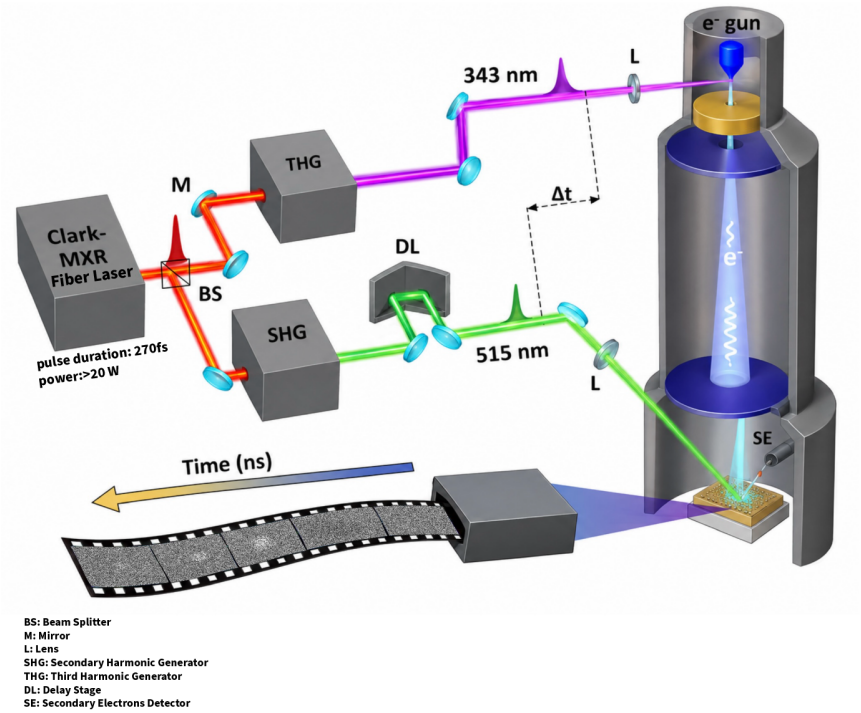


Figure 1 Schematic illustration of the 4D-SUEM system and its operating principle. A pulsed optical pump laser excites the sample, while a delayed ultrafast electron probe scans the surface. The resulting changes in the secondary electron (SE) signal are recorded to visualize the spatial and temporal dynamics of photogenerated charge carriers[8].

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During operation, the ultraviolet beam is directed at the SEM electron gun, generating ultrashort electron pulses in a photoemission process, replacing the conventional continuous thermionic electron source. Simultaneously, the green beam is focused on the sample surface for optical excitation. An optical delay stage was used to control the time difference between the electron probe and optical pump, which allowed the images to be recorded at different pump-probe delay times[3]. The microscope was operated at a fixed working distance of 10 mm and an acceleration voltage of 30 keV. For both GaAs and CdTe samples, the green laser power was kept at 1.7 mW while taking measurements. The secondary electrons (SE) emitted from the sample surface were collected by a positively biased Everhart-Thornley detector. Each final image was generated by averaging 64 individual frames, with an exposure time of 100 ns for each frame, to improve the signal-to-noise ratio.

Following the recorded images, the time-resolved carrier dynamics were analyzed by comparing the images taken before and after the optical excitation. The pre-excitation images were used as the baseline and subtracted from the corresponding post-excitation images using MATLAB. This produced contrast images in which the changes caused by the laser excitation could be more clearly observed. The resulting images were used to study the photogenerated carrier dynamics at the surfaces of the GaAs and CdTe samples. Both samples were used as received without additional surface treatment; however, they were cut immediately before characterization to

expose fresh, clean edges for the measurements.

3 Results and Discussion

We used Time-resolved SUEM to investigate the spatial and temporal evolution of photo-induced surface charge dynamics in GaAs and CdTe. We analyzed the spatial distributions using a two-dimensional Gaussian fitting, while the temporal dynamics was fitted using the two-component exponential function (eq 1):

$$y = A_1 \exp\left(-\frac{t}{\tau_1}\right) + A_2 \exp\left(-\frac{t}{\tau_2}\right) + C \tag{1}$$

where t is the pump-probe delay time, τ_1 and τ_2 are the fast and slow time constants, respectively, A_1 and A_2 are the corresponding amplitudes, and C is the constant background offset.

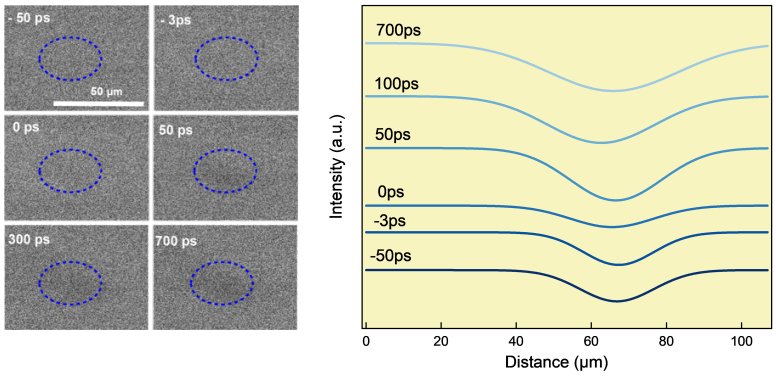


Figure 2 (a) Time-resolved SUEM images of the GaAs sample at different pump-probe time delays and (b) Line-profile of the same SUEM images at different time delays.

For GaAs, the time-resolved images in Figure 2(a) show a localized dark contrast within the laser-irradiated region after excitation. The corresponding spatial intensity profiles in Figure 2(b) show a clear decrease in intensity around the center of the excited region, which gradually becomes less pronounced at longer time delays.

In SUEM, a bright contrast generally results from an increase in SE emission when photoexcited electrons gain enough energy from the optical excitation to contribute more effectively to SE generation. In contrary to a dark contrast that can result from processes that reduce the energy or number of secondary electrons emitted, including electron-electron scattering, electron-phonon scattering, electron-impurity scattering, carrier trapping, recombination, and surface oxidation or surface states that promote carrier trapping and energy loss[6][1].

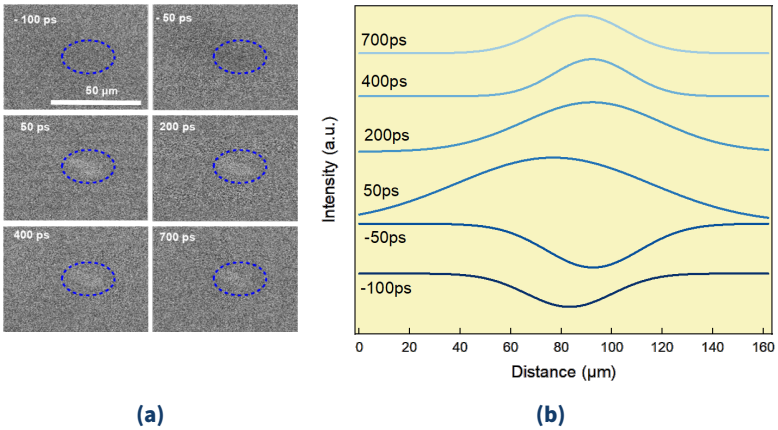


Figure 3 (a) Time-resolved SUEM images of the CdTe sample at different pump-probe time delays and (b) Line-profile of the same SUEM images at different time delays.

For CdTe, Figure 3(a) shows a clear transition from the pre-excitation state to a bright photo-induced feature at positive time delays. The bright region becomes clearly visible at 50 ps and remains present at later times. This behavior is also evident in the spatial profiles in Figure 3(b), where a clear intensity peak develops after excitation and persists over the measured time range.

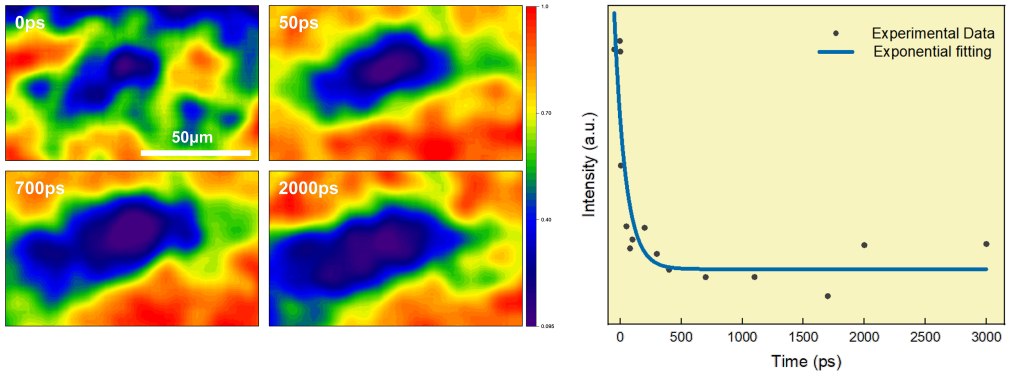


Figure 4 (a) colored maps showing the spatial evolution of the photo-induced region in the GaAs sample. (b) Temporal evolution of the SE intensity in GaAs, yielding τ_1 = 4.69 ps and τ_2 = 256.65 ps.

The colored maps in Figure 4(a) further show that the spatial evolution of the dark contrast with increasing delay time, in these maps the red regions correspond to high SE intensity while blue regions correspond to lower SE intensity. The kinetic trace shows a fast initial change followed by a slower evolution toward a nearly constant intensity, as shown in Figure 4(b). The fitting using eq1 gives τ_1 = 4.69 ps and τ_2 = 256.65 ps. The fast component represents the initial ultrafast response, while the longer component describes the slower evolution of the photo-induced SE signal.

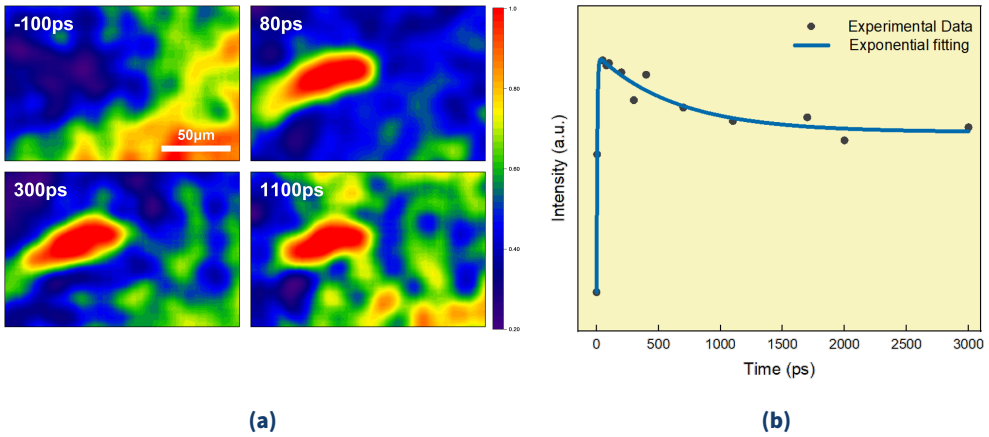


Figure 5 (a) colored maps showing the spatial evolution of the photo-induced region in the CdTe sample. (b) Temporal evolution of the SE intensity in CdTe, yielding $\tau_1 = 5.72$ ps and $\tau_2 = 645.09$ ps.

The colored maps in Figure 5(a) show the spatial evolution of the bright contrast in CdTe at different delay times. The bright contrast becomes clearly visible after excitation and remains observable at longer delay times, indicating the persistence of the SE signal. The corresponding kinetic trace in Figure 5(b) shows a fast increase in SE intensity followed by a slower decay. The fitting using eq1 gives $\tau_1 = 5.72$ ps and $\tau_2 = 645.09$ ps, representing the fast and slow temporal components of SE response, respectively.

These findings reveal how GaAs and CdTe exhibit different photo-induced surface charge dynamics, with differences in both the observed SE contrast and time constants.

4 Limitations and Future Work

One of the main limitations faced during this internship was the limited access time we had when using the 4D-SUEM system. Each session lasted 1 hour, followed by a 45-minute refresh period, limiting our experimental time. We faced challenges in processing and analyzing the experimental data using MATLAB and Origin, particularly when extracting the spatial profiles and kinetic traces. Choosing the suitable data fit for the kinetic data required extra time to accurately represent the results of our data.

For future work, the cause behind the dark contrast could be investigated further by combining 4D-SUEM with surface characterization techniques such as XPS and AFM. Comparing untreated and etched surfaces could help determine the role of surface oxidation and surface states in the observed contrast. Measurements at different electron energies could provide more information about the surface and subsurface carrier dynamics. Numerical modeling could also help to understand how scattering processes affect the SE signal. Finally, studying other photovoltaic materials such as perovskites could provide a broader comparison of photoinduced surface dynamics in different semiconductor materials [6][1].

5 Conclusion

In this study, we used 4D Scanning Ultrafast Electron Microscopy (SUEM) to investigate the spatial and temporal evolution of photogenerated carrier dynamics in GaAs and CdTe. The resulting images showed different photo-induced responses in the two materials, with GaAs showing a dark contrast in response to photoexcitation and CdTe showing a bright contrast. The fast and slow components were identified for both materials, with time constants of $\tau_1 = 4.69$ ps and $\tau_2 = 256.65$ ps for GaAs, and $\tau_1 = 5.72$ ps and $\tau_2 = 645.09$ ps for CdTe. These differences demonstrate that the two materials undergo distinct surface processes after photoexcitation.

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