A Coherent Case Study

Real Space Imaging of Transient Carrier Dynamics by Femtosecond Time-Resolved STM

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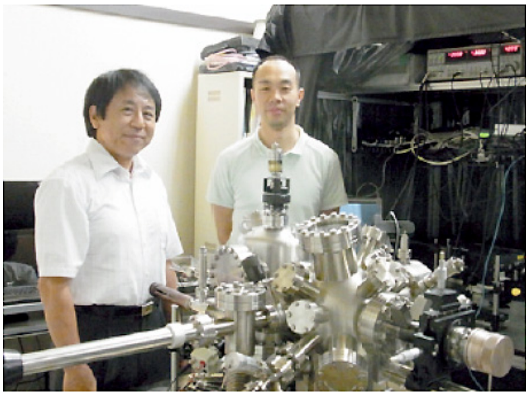
Main Research Details



From left to right: Prof. Hidemi Shigekawa, Asst. Prof. Shoji Yoshida

"Smaller and faster" are the key words for nanoscience and nanotechnology. Therefore there is a strong need for methods able to explore transient dynamics in small organized nanostructures. In this white paper we discuss nano-scale imaging of carrier dynamics over a wide range of lifetimes by femtosecond lasers combined with scanning tunneling microscopy (STM).

The real space observation of atomic-scale structures by STM has revealed answers to various long-standing questions and is pushing the frontiers of science and technology. However, since the temporal resolution of STM is less than 100 kHz, ultrafast transient dynamics have been well beyond its limits. Conversely, the development of ultrashort-pulse laser technology enabled us to observe dynamics in the femtosecond timescale. However, spatial resolution with conventional pump & probe techniques is generally limited by the laser wavelength employed. Therefore, since the invention of the STM in 1982, one of the most challenging goals has been to combine the spatial resolution of STM with the time-resolution afforded by femtosecond lasers.

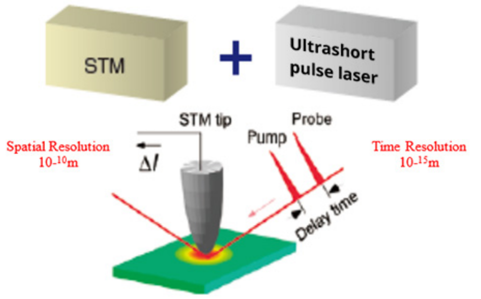
In the Shigekawa Lab, we are always looking for different approaches to tackle new research themes. And one of the ground-breaking results we have achieved has been the extension of STM to the ultrafast domain by developing a time-resolved set-up that combines a STM and femtosecond lasers by Coherent (Chameleon and Mira).

By adopting new ways to combine these two devices, we have realized angstrom-level microscopy with unprecedented femtosecond- time resolution.

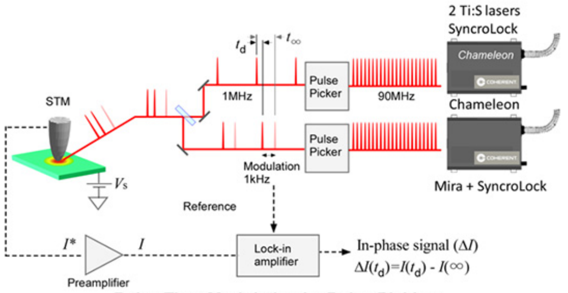
Solutions & Benefits

Professor Shigekawa explains, "An important and enabling factor in achieving our goal – combining an STM with a femtosecond laser – is the stability of the laser over extended periods of time as we needed to synchronize the two lasers and control the delay time over a wide time delay range, from picosecond to microsecond. This was achieved by using two Pockels cells, to reduce the repetition rate as required when measuring delays longer than a few nanoseconds.

Furthermore, to analyze various dynamics it's important that the wavelength can be changed easily. The Chameleon and Mira femtosecond lasers have the appropriate stability, and are easily synchronized and controlled thanks to their onboard SyncroLock feature. In addition, Chameleon provides high output power and a wide wavelength range, making it well suited for our objectives. Presently, we are moving forward with research to further expand upon different laser combinations with STM."



High Spatial and Temporal Resolution



Delay Time Modulation by Pulse Picking

Figure 1. Optical Pump Probe STM

Examples of Research Results

Confirming temporal resolution

As a first demonstration, we looked at measurement by tunneling current (with atomic-level spatial resolution possible) of photo-induced dynamics caused by femtosecond pulses spaced over a wide time domain.

Figure 2 compares the results by conventional optical pump probe method (a, c, e), i.e., with an optical signal, and the results by time-resolved STM (b, d, f, g), i.e., relying on a tunneling current signal. These confirm that the STM method has the same time resolution as an all optical method, over several very different time scales.

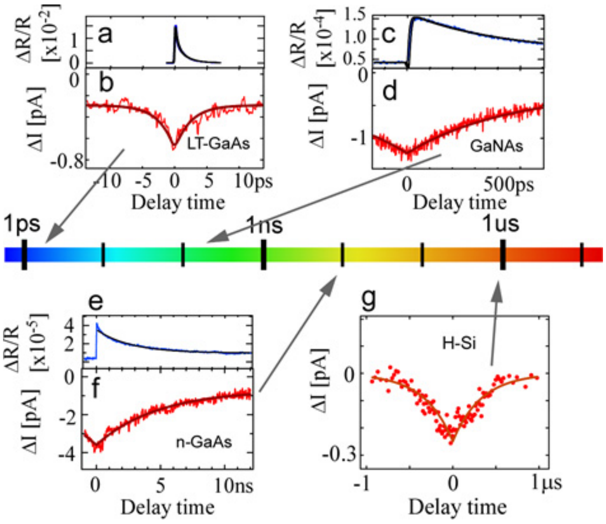


Figure 2. Time Resolved STM Spectra

Measuring localized carrier dynamics

PIN structures are basic and common semiconductor elements used for example in solar cells. To better understand and improve the performance of PIN devices, it is essential to resolve in real space the effects on carrier dynamics of changes in potential at interfaces and local structures. Figure 3 maps the location and density of a small number of photo-excited carriers (shown in red) over time.

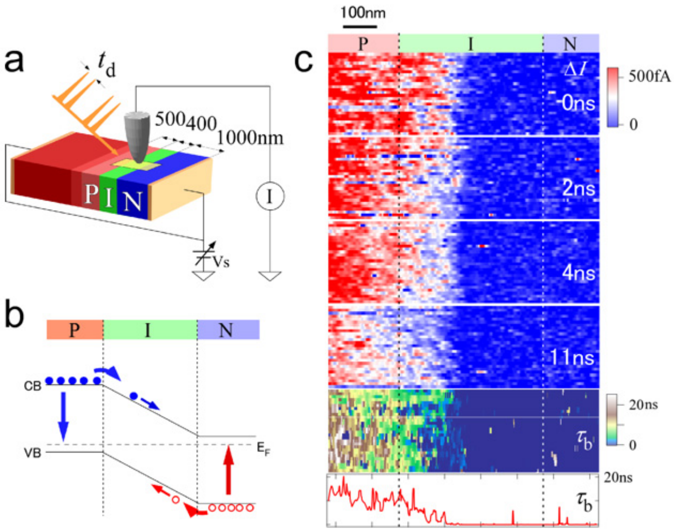


Figure 3. GaAs-PIN Structure Time Resolved Maps

The top four panels are at different time intervals, showing that in just a few nanoseconds, most of the carriers have decayed. The fifth panel replots this data in the form of a plot of decay time vs. location. And the bottom panel is a cross-sectional plot of decay time located along the white line shown in the fifth panel.

Charge transfer on the nano scale

Charge transfer and related carrier dynamics play an extremely important role in device development and control of catalysis. So in another study, we used our ultrafast STM instrument to look at charge transfer of metal nanoparticles/semiconductor structures. In Figure 4, (a) is an STM image of a cobalt nanoparticle/GaAs structure; (b) is a lifetime map of the capture (recombination) time of the holes generated by photo-excitation at each point in (a), and (c) is a superposition of a and b. It is assumed that for this type of nanoparticles, recombination is promoted at the metal/semiconductor interface (via a gap state). This mapping data confirms that indeed a shortening of carrier lifetime occurs in the cobalt domain. Moreover, the graph (d) shows that carrier lifetime Tcap decreases as particle size increases, specifically a Tcap∝1/S relationship is observed.

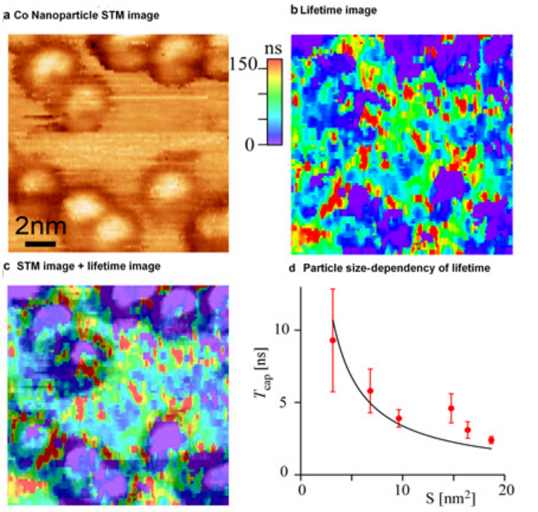


Figure 3. Time-Resolved Signal Real Space Mapping

Summary

We have shown that femtosecond optical pulses can be combined with STM technology to enable the dynamics of materials to be studied with an unprecedented combination of temporal and spatial resolution. This type of data can lead to improved understanding of fast nanoscale excitation events, potentially enabling better electronic and photonic devices. The availability of stable femtosecond lasers with stable timing control is a key facilitating technology in this work.