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Scientific Article

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

Yasuhiko Terada, Shoji Yoshida, Osamu Takeuchi, and Hidemi Shigekawa
Institute of Applied Physics, University of Tsukuba, Tsukuba 305-8573 Japan

"Smaller and faster" are the key concepts in the progress of current nanoscience and technology. Thus, a method for exploring the transient dynamics in small organized structures is eagerly desired. Real-space imaging of nanoscale carrier dynamics with a wide range of lifetimes has been realized by pulse-laser-combined scanning tunneling microscopy (STM) with a noble delay time modulation method developed from a pulse-picking technique.

The real space observation of atomic-scale structures by STM has lifted the veil from various longstanding problems and is extending the frontiers of science and technology. However, since the temporal resolution of STM is less than 100 kHz, transient dynamics has been beyond its field of vision. In contrast, the development of ultrashort-pulse laser technology has enabled us to observe ultrafast dynamics, currently in the femtosecond range. Its spatial resolution, however, is generally limited by the pulse wavelength. Therefore, since the invention of the STM in 1982, one of the most challenging goals has been to combine STM with ultrashort-pulse laser technology.

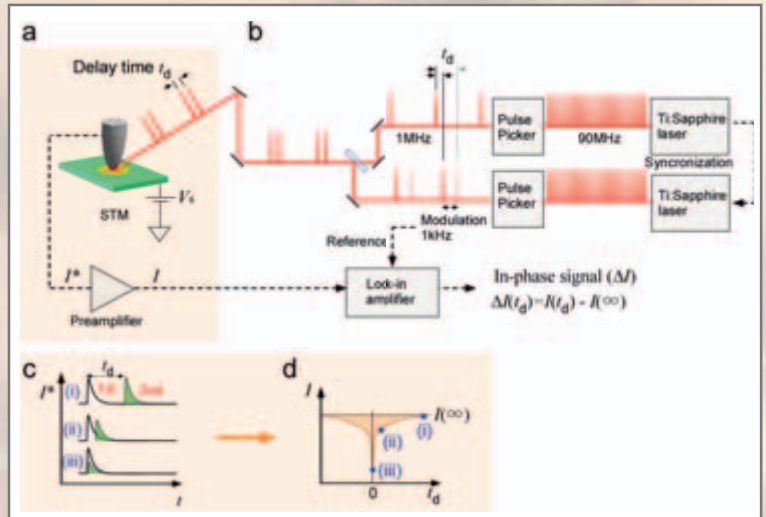
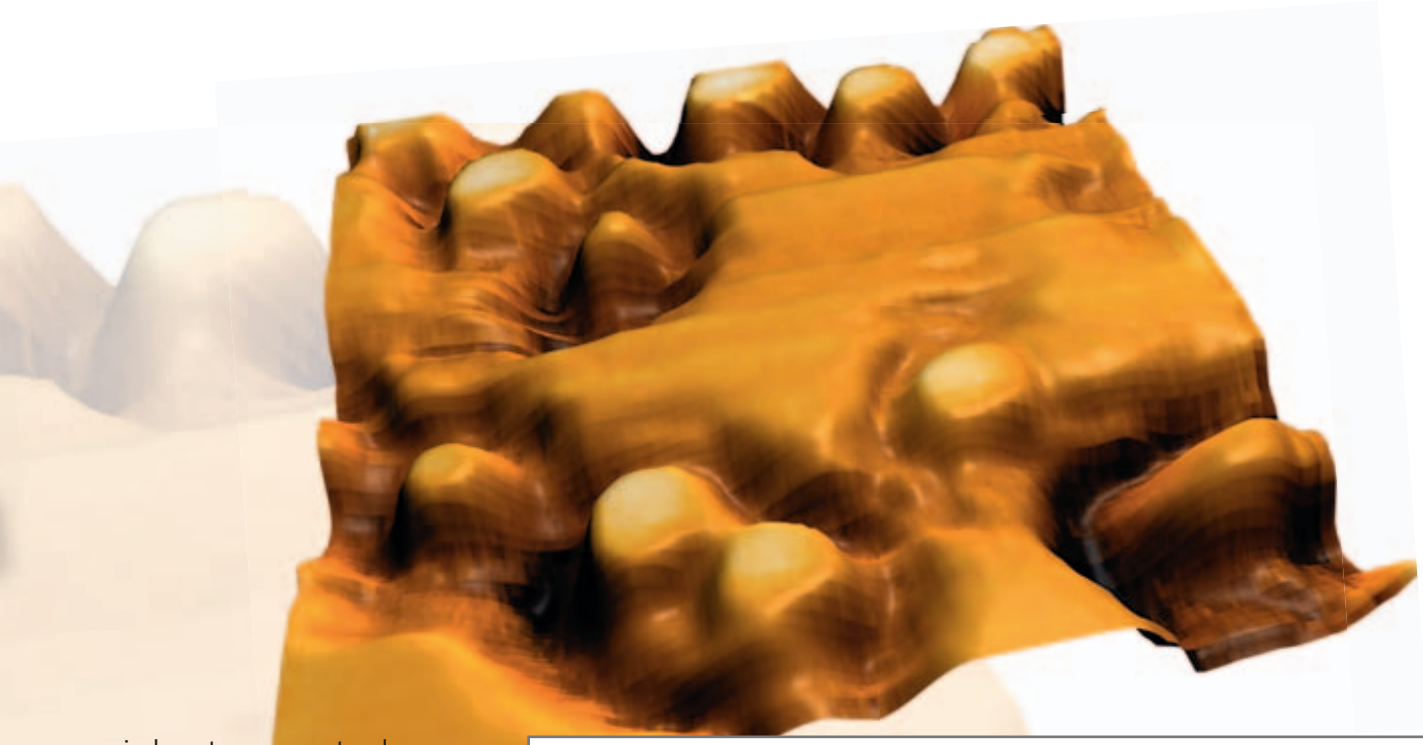


Fig. 1 (a) (b) Schematic illustrations of SPPX-STM. Optical pulses from the two synchronized Ti-sapphire lasers are selectively transmitted using pulse pickers to produce a sequence of paired pulses with a certain delay time, td. Pulse pairs with a central wavelength of 800 nm and an average intensity of 1 mW are focused to less than 10 μm below the STM tip, and tunnel current is measured as a function of td. (c) Relationship between raw tunneling current I^* and delay time t_d. (d) Measured tunneling current I as a function of delay time, where the delay times labeled (i), (ii) and (iii) correspond to those of (i), (ii) and (iii) in (c).

Figure 1 shows schematic illustrations of our newly developed microscopy technique, shaken-pulse-pair-excited STM (SPPX-STM), and its basic probing mechanism.

The surface of a sample beneath the STM tip is excited by a sequence of paired laser pulses with a certain delay time t_d and the tunneling current I is measured as a function of t_d (Fig. 1a). The optical pulses give rise to current pulses in the raw tunneling current I^* (Fig. 1c), reflecting the excitation and relaxation of the sample. When t_d is sufficiently long, the paired optical pulses with the same intensity independently



induce two current pulses with the same magnitude in I^* (Fig. 1c-(i)). In contrast, when t_d is short and the second pulse illuminates the sample in the excited state induced by the first pulse, the second current pulse may have a different height, depending on t_d (Figs. 1c-(ii) and 1c-(iii)). The signal I therefore also depends on t_d, because the height difference in the second current pulse changes the temporally averaged value of the tunneling current (Fig. 1d). Accordingly, the relaxation dynamics of the excited state in the target material can be probed by STM. In principle, the spatial and temporal resolutions of a microscopy technique are limited by those of the probes used, that is, tunneling current (atomic scale) and optical pulse width (femotosecond range) in the case of SPPX-STM.
To realize SPPX-STM, we developed a method enabling the discrete and large modulation of the delay time by using a pulse-picking technique (Fig. 1b), which, for the first time, has allowed the probing of carrier dynamics in nanometer-scale structures over a wide range of time scales. Furthermore, this method reduces the measurement time and hence enables the spatial mapping of time-resolved signals, which has been desired for a long time.

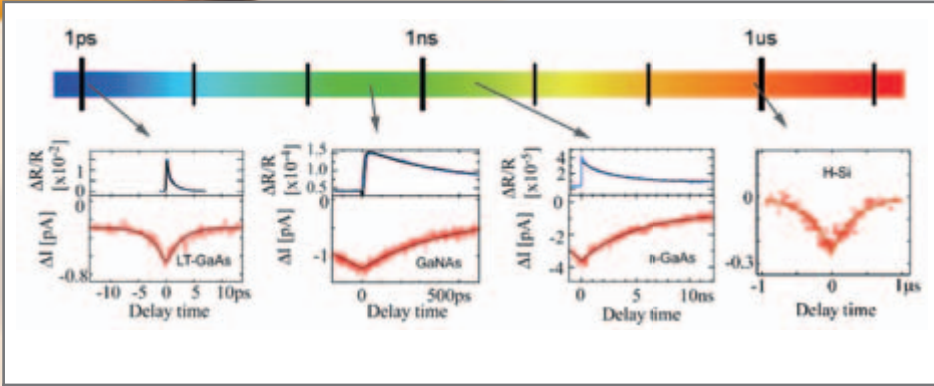


Fig. 2 SPPX-STM spectra obtained for low-temperature-grown GaAs (LT-GaAs), GaNAs, n-GaAs and hydrogen-terminated Si samples.

Figure 2 shows time-resolved spectra obtained for various semiconductors. With the new method, carrier dynamics can be measured using a single microscope over a wide range of time scales. We can evaluate the transient dynamics in small organized structures consisting of composite materials with the spatial resolution of STM.

Figure 3 shows the first real-space imaging of carrier dynamics in the nanosecond range modulated on the nanoscale. Photoexcited minority carriers (holes) captured at the surface of GaAs are recombined with electrons tunneling from the STM tip via the gap states formed at the Co nanoparticle/GaAs interface (Fig. 3(a)). Understanding such a charge transport mechanism involving nanoparticles is of great importance not only for the development of nanoscale electronic devices but also for their application to the finer control of chemical reactions in catalysis.
Figure 3(c) shows the overlap of the STM image in Fig. 3(b) with the map of the decay constant of photoinduced carrier density obtained over the surface. As shown in the cross section in Fig. 3(d) obtained along the → P.8

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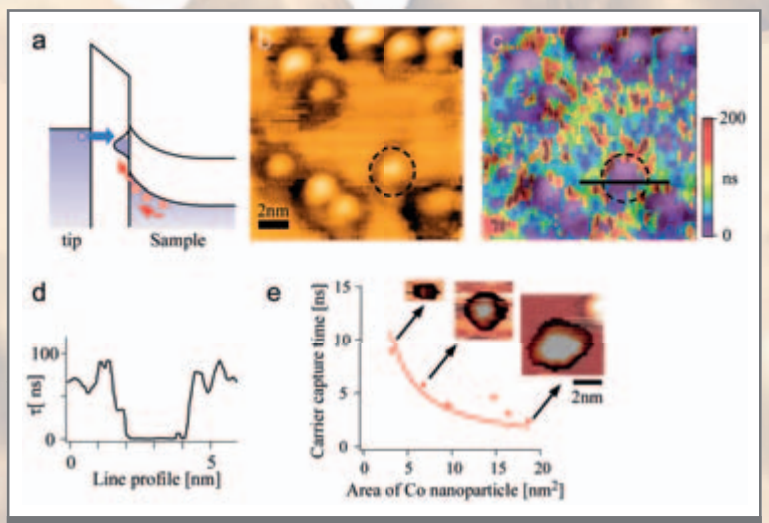
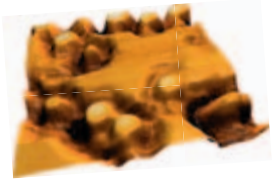


Fig. 3 (a) Schematic of the carrier recombination mechanism via the gap states. (b) STM image of Co nanoparticles/GaAs. (c) Overlap of color-scale two-dimensional mapping of decay constant obtained over the surface in (a). (d) Cross section of the decay constant along the line in (b). (e) Hole capture time as a function of nanoparticle size.

line in Fig. 3(c), the decay is rapid in the Co regions owing to the enhancement of carrier recombination via the gap states.
Figure 3(e) shows the decay constant as a function of the Co nanoparticle size. The time constant increases with decreasing nanoparticle size as expected.



3D representation of the STM image of Co nanoparticles/GaAs

SPPX-STM enables us to study ultrafast dynamics simultaneously with the observation of local structures by STM. In general, phenomena such as dipole formation, charge transfer, changes in conductance, modulation of local potential, and vibration are possible mechanisms for producing signals. Using an electromagnetic field combined with polarized light, SPPX-STM also has the potential to be used to investigate spin dynamics.

(Instrument:
Variable Temperature SPM)

The Research Group

Prof. Hidemi Shigekawa
e-mail:
hidemi@ims.tsukuba.ac.jp

Associate Prof.
Osamu Takeuchi

Assistant Prof.
Shoji Yoshida

Professor H. Shigekawa leads a group specializing in nanoscale science and technology based on scanning probe microscopy and related techniques. The group also has experience in synchrotron radiation experiments and has been engaged in research on ultrashort-pulse laser technology during the last 10 years. On the basis of their research backgrounds, group members have been developing new microscopy techniques and investigating applications of these techniques. The current research interests of group members include semiconductor physics, molecular physics, low-temperature physics and low-dimensional physics.
Web: http://dora.bk.tsukuba.ac.jp
Corresponding author: hidemi@ims.tsukuba.ac.jp

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