A Coherent Case Study

Extreme Stability of Monaco is Instrumental in Supporting Cutting-Edge Nanoscale Metrology

User: Prof. Hidemi Shigekawa's Laboratory, Institute of Applied Physics, University of Tsukuba, Japan

Study theme: "Sub-cycle Transient Scanning Tunneling Spectroscopy with Visualization of Enhanced Terahertz Near Field"

About the Shigekawa Lab

The understanding and control of quantum dynamics, such as carrier transitions and transport in nanoscale structures are key factors that will enable the continued advancement of nanoscale science and technology. Professor Shigekawa and co-workers at the University of Tsukuba are driving research and development of new nanoscale metrology. This includes the world's first combination of scanning tunneling microscopy (STM) with ultrashort-pulse laser technology to obtain field measurements with extreme spatial and temporal resolution.

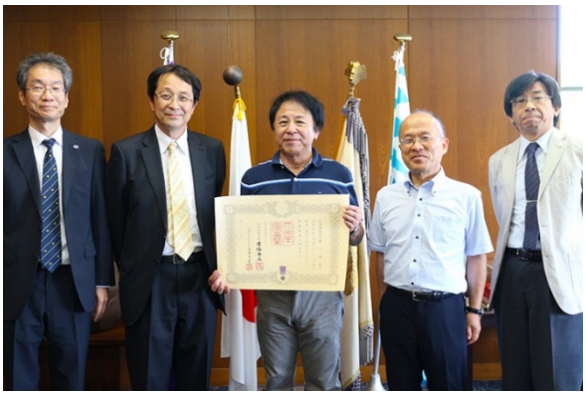

Figure 1. Prof. Shigekawa (center), at the award ceremony. (Source: http://www.tsukuba.ac.jp/)

Because of his significant achievements in applied physics, Prof. Shigekawa was awarded the Shiju-hosho (Medal of Honor with Purple Ribbon) in early 2019. This prestigious award is given for outstanding contributions in academic fields, arts, and sports.

Background and goal of this study

Recently, the terahertz scanning tunneling microscope (THz-STM) has gained increased interest. As shown schematically in figure 2, the electric field of THz radiation takes the place of the conventional bias voltage (between the tip and sample) traditionally used in an STM.

In the case of THz pulses, it is possible to obtain CEP-stable THz "light" consisting of nearly single-cycle pulses via optical rectification—by sending ultrashort optical pulses into one of various nonlinear optical crystals such as $LiNbO_3$. CEP-stable (Carrier Envelope Phase stable) refers to the phase offset (if any) between the pulse envelope and the oscillating electric field within the pulse. When combined with an STM, controlling the CEP provides control over the direction of the transient electric field applied between the tip and sample. In addition, when a nanoscale metal structure is irradiated by electromagnetic waves, the near field formed on the material surface is enhanced in a confined space far smaller than the diffraction limit. Therefore, in the case of the metal probe (nanotip) of an STM, it is possible to obtain an intense terahertz near field in a local region directly beneath the nanotip, owing to this field enhancement effect.

Furthermore, by the manipulation of THz pulses with controlled CEP, it is possible to apply both a positive and negative electric voltage – up to several volts – between the tip and sample. This results in a transient tunnel current between the tip and sample. With this approach, it is possible to interrogate the localized region directly beneath the nanotip with the spatial resolution of an STM, enabling THz-driven nanoscopy. Because the applied voltage between the tip and sample is on the femtosecond timescale, dynamic effects that can be induced and/or interrogated by light can be studied on this same timescale. This is done by using pump-probe techniques where one part of the laser pulse acts as the pump, and another part is converted to produce the THz pulse. Prof Shigekawa and coworkers have used this approach to obtain direct measurements of the transient electric field in THz-STM.

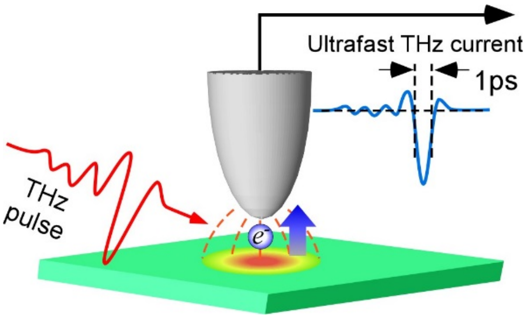

Figure 2. THz-STM

Why is it important to measure this electric field? The confinement and focusing of the THz-induced field results in an amplification and modulation of this field. To fully understand and exploit THz-STM measurements of dynamic effects in samples, it is essential to accurately measure the true waveform of the near field. In the experiments described in this case study, the researchers have used pump-probe methods to directly evaluate the waveform and the intensity of the enhanced near field in the tunnel junction in a THz-STM setup, with femtosecond temporal resolution. Moreover, this approach provides a platform for the observation of ultrafast dynamics at the atomic scale and for studying strong-field-driven dynamics.

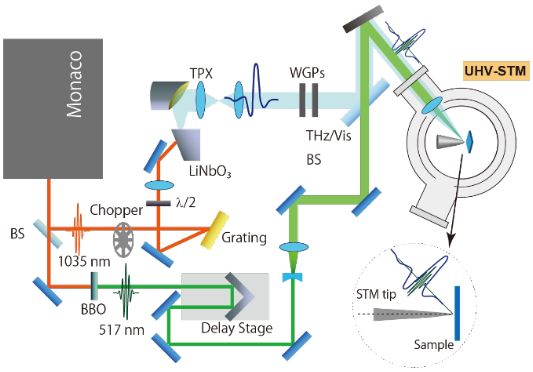

Figure 3. Key components of the time-resolved THz-STM setup

Method of this study

This study involved pump-probe experiments using a system that combines an ultrashort-pulse laser amplifier (Coherent Monaco) and an ultrahigh-vacuum STM system, which supports precise manipulation of the nanotip position. The electric field enhancement was achieved with a Platinum-Iridium-coated Tungsten STM nanotip. The nanotip was irradiated with two types of optical pulses: a THz pulse and an optical domain pulse. The THz pulse was generated by irradiating a $LiNbO_3$ crystal with an infrared (IR) pulse (1035 nm, 309 fs, 1 MHz) and it was nearly a single-cycle with a large first peak of 1 ps width. The probe pulse was either the beam at 1035 nm or its second harmonic at 517 nm, generated in a BBO crystal.

Figure 3 illustrates the two types of measurement methods – photoemission regime and tunneling regime. This way it is possible to compare the field generated at a typical (i.e. nanometer) STM tunneling junction with the field generated at the tip when tip and surface are well separated (~ a micrometer) and tunneling therefore cannot play a role.

For the tunneling regime measurements, a part of the 1035 nm laser output was used to create hot electrons. These electrons tunnel through the potential barrier at a rate determined by its height; since this is a direct function of the local field, by measuring the integrated current as function of the delay between the (pump) THz and (probe) 1035 nm pulses, this field can be measured on the femtosecond timescale. At larger distances between the surface and the nanotip, the THz field can still be measured by the current between the tip and the surface – in this case a photoemission current.

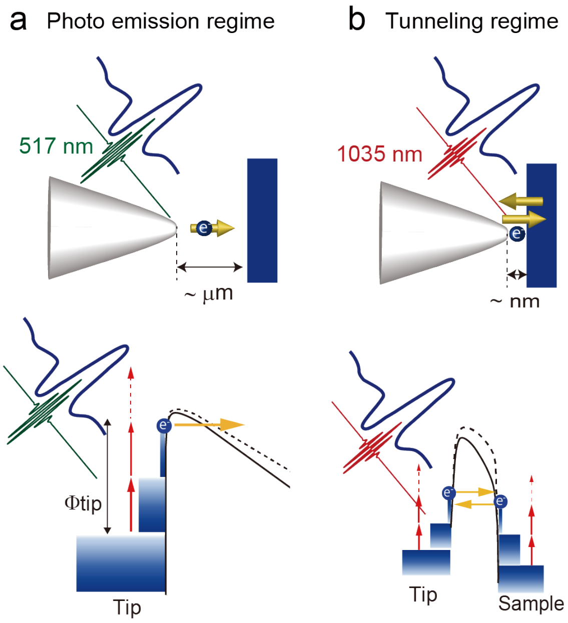

Figure 4. (a, b) Schematic illustrations of measurement methods in photoemission (photofield emission and above-threshold photoemission) regime and tunneling regime, respectively. The barrier height is modulated by the THz electric field and photoelectron emission (hot electron tunneling) is induced by the simultaneous irradiation by the probe light at 517 nm (1035 nm)

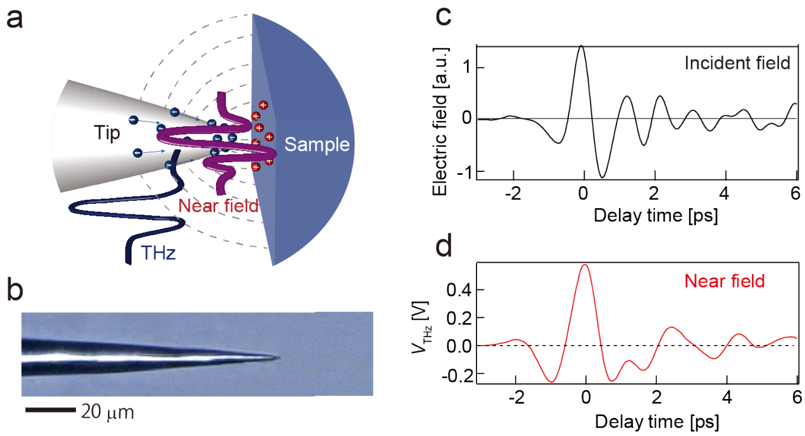

Figure 5. (a) Incident electric field and its modulation by a nanotip. (b) Nanotip. (c) Original waveform of the THz light. (d) Waveform of Enhanced THz electric field.

Experimental Setup

The Coherent Monaco femtosecond laser was used as the overall laser source for this study (40 W at 1035 nm, <350 fs pulse duration and 1 MHz repetition rate, later upgraded to reach 50 MHz). Figures 6 and 7 show the system configuration with THz-STM system and the Monaco laser. Briefly, the pulse light from the Monaco is split into two beams by beam splitter BS, with one beam used for THz generation and the other beam for the optical wavelengths. For efficient THz generation, we used the tilted pulse-front approach with a $LiNbO_3$ crystal. The THz pulse generated is adjusted in intensity by polarizer (WGPs), recombined with the pump light by a THz beam splitter, and focused between the STM tip and sample in the ultrahigh-vacuum chamber. The pump light is switched back and forth between fundamental wavelength (1035 nm) and second harmonic (517 nm) generated by BBO, depending on the experiment.

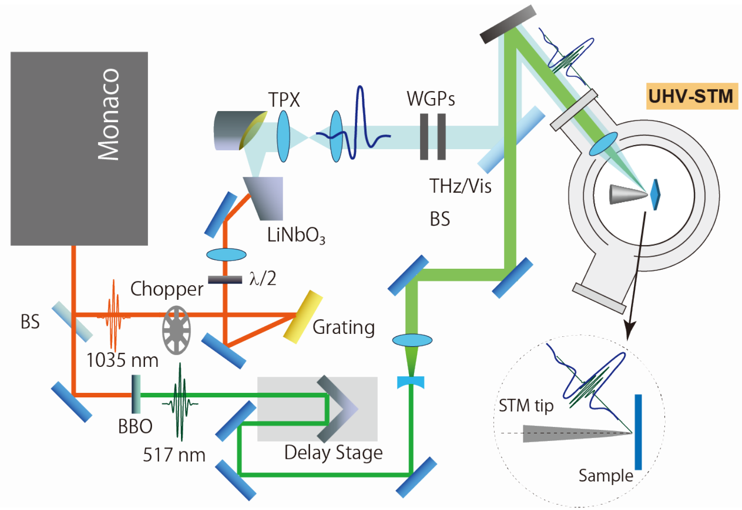

Figure 6. System configuration

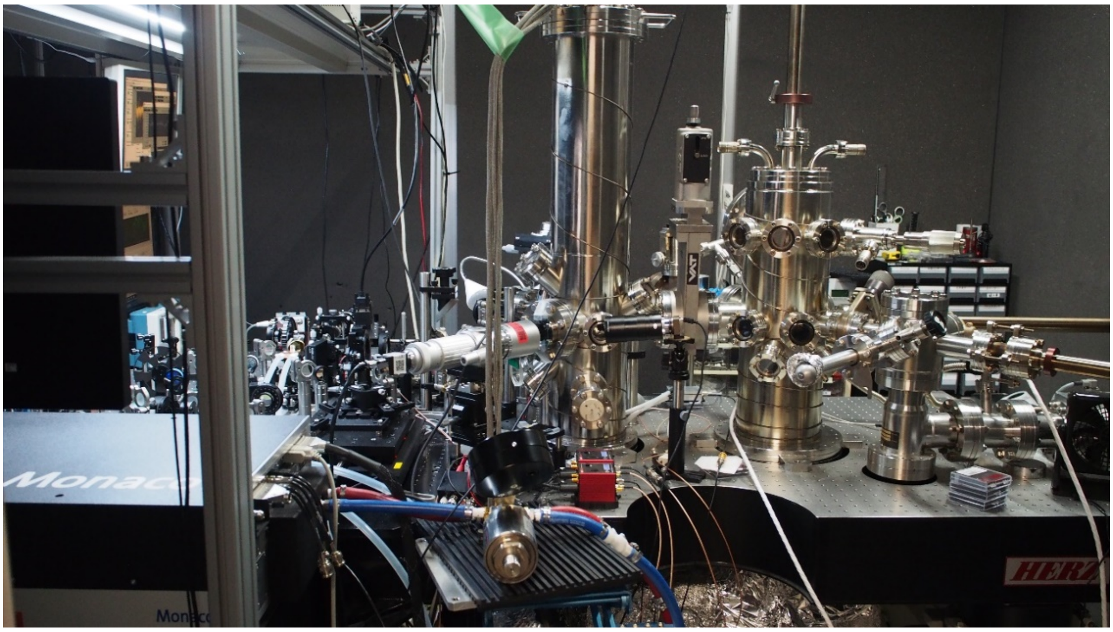

Figure 7. The Coherent Monaco femtosecond laser and THz-STM system.

Examples of time-resolved measurement Results

Figure 8 shows time-resolved measurements performed with the THz-STM system. The sample material - 2H-MoTe2 - has similar properties to a semiconductor; its STM image in Figure 8a shows the intrinsic defects (bright spots), here resolved at atomic level.

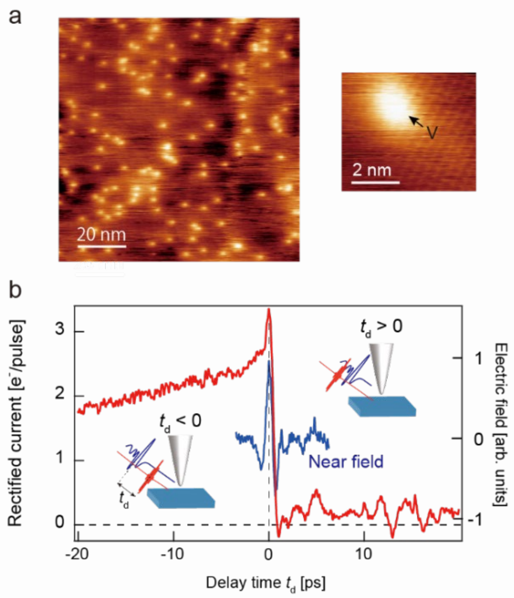

Figure 8. Transient electronic dynamics obtained by THz-STM for 2H-MoTe$_2$. (a) STM image of the sample. (b) Delay-time dependence of I THz

Figure 8b is a chart of this measurement result. The delay time td < 0 indicates that the THz beam reaches the sample after the optical beam excites it and creates hot photoelectrons. The rectified current reflects the dynamics of the excited state due to photocarrier generation. For more details, refer to the published article [1].

Conclusion

Professor Shigekawa summarizes, "In this study, we succeeded in developing the technology to experimentally acquire terahertz near-field waveforms at a nanotip in a STM setup. This was possible because of the combination of our own STM system and the Monaco ultrafast short pulse laser – see figures [6 and 7]. This pump-probe method is a complementary technique to other time-resolved techniques that have been developed previously. However, this new method is straightforward to use and provides unique and previously unachievable measurements on relevant samples. The relative simplicity of the method will allow many researchers to enter this research field more easily than before, which in turn will expand the range of applications. Looking forward, we therefore expect our method to play a valuable role in [the] development of science and technology."

Why the Coherent Monaco laser

A key member of the research team is Assistant Professor Yoshida. He explains the choice of laser source, "The most important prerequisites for a laser for these studies were high repetition rate and high power. To generate THz light most effectively, high pulse energy is necessary. And to achieve highly accurate measurements for this study, we need to use a high repetition rate. Until recently, it was very difficult to find such a laser which meets both of these requirements. However, Monaco meets these needs perfectly.

"We actually started these studies with a 1 MHz model of Monaco. But as we moved forward, we realized we could ideally use even higher repetition rates. So, we decided to upgrade the Monaco laser to 50 MHz repetition rate and at the same time we adjusted several system components to be compatible with the higher power this delivered. As a result of this upgrade, we can use the system with 40 W full power and higher repetition rate with high stability for these experiments."

Assistant Professors Arashida and Yoshida stated, "In addition, we have found that Monaco maintains its high stability over a wide range of operating conditions. This flexibility allows us to study an expanded range of samples. For example, we can independently adjust the pulse energy and pulse repetition rate. This allows us to optimize the signal-to-noise ratio, for example, to study processes spanning quite different timescales."

References

[1] ***[Sub-cycle transient scanning tunneling spectroscopy with visualization of enhanced terahertz near-field](#)***. Shoji Yoshida, Hideki Hirori, Takehiro Tachizaki, Katsumasa Yoshioka, Yusuke Arashida,Zi-Han Wang, Yasuyuki Sanari, Osamu Takeuchi, Yoshihiko Kanemitsu, and Hidemi Shigekawa, ACS Photonics, 6, 1356-1364 (2019).

Prof. Hidemi Shigekawa's Lab URL: http://dora.bk.tsukuba.ac.jp/