

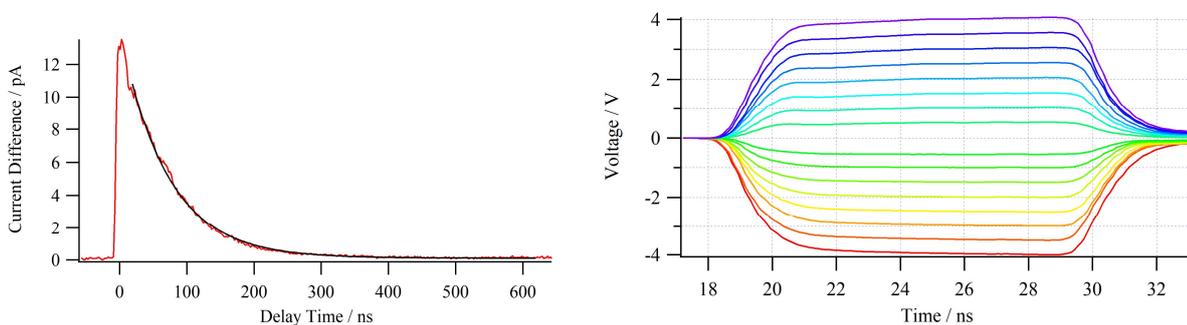
Light-assisted Timeresolved Scanning Tunneling Spectroscopy

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Scanning tunneling microscopy (STM) not only has an ultimate spatial resolution, but also an energy resolution of local density of electron states (LDOS), which allows us to precisely investigate spatially resolved individual molecular orbitals or band structure. Recently developed pump-probe STMs added time resolution to STM. They no more suffered from the slow response of STM preamplifier and can resolve ultrafast dynamics that occur from nanoseconds to even femtoseconds.[1,2] Except for some recent attempts, however, most of the time resolved STM studies so far do not utilize its energy resolution. Optical-pump optical-probe type STM is not compatible with the I-V curve measurements that energetically resolve instantaneous LDOS. Instantaneous I-V curve measurement with Terahertz-pulse probe STM is started to be discussed but it is not straightforward because of the complex shape of the probe pulse. Instantaneous I-V curve measurement with square shaped bias voltage pulse is a promising technique to do timeresolved STS measurements but has not been established.

In this study, we developed light-assisted timeresolved scanning tunneling spectroscopy and applied it to investigate ultrafast photocarrier dynamics from nanosecond evolution of STM I-V characteristics. The multilayer WSe₂ sample under STM was illuminated by an externally triggerable nanosecond pulse laser and the square shaped bias voltage pulses were applied to the sample with finite offset (DC) voltage while the STM tip was grounded via the STM preamplifier. The voltage pulses were periodically chopped for lock-in detection of time resolved tunnel current, which exhibited exponential decay after illumination with 72 ns time constant as shown in the left figure. The evolution of time resolved I-V characteristic measured with varying the pulse height as shown in the right figure and their physical interpretation with some technical details will be discussed at the presentation.



References:

- [1] Y. Terada, M. Aoyama, H. Kondo, A. Taninaka, O. Takeuchi and H. Shigekawa, *Nanotechnology* 18,044028 (2007).
- [2] S. Yoshida, T. Arashida, H. Hirori, T. Tachizaki, A. Taninaka, H. Ueno, O. Takeuchi, and H. Shigekawa, *ACS Photonics* 8, 315-323 (2021).