

## Time Resolved Scanning Tunneling Microscopy: Instruments

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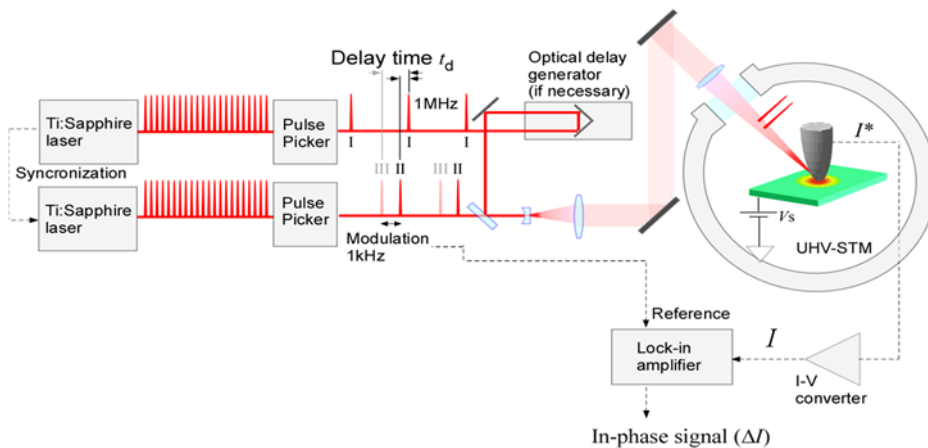
In order to explore the transient carrier dynamics in organized nanostructures, we recently developed shaken-pulse-pair-excited STM (SPPX-STM) [1]. In SPPX-STM, the tunnel junction is illuminated by a sequence of ultrafast laser pulse pairs. While the delay time between the two pulses in the pulse pairs is periodically modulated, the response in tunnel current is detected by a lock-in amplifier. In the first generation SPPX-STM, we modulated the delay time mechanically by using a piezo device. However, the method suffered from large noise because the modulation frequency was limited to less than 30Hz.

To improve the signal-to-noise ratio, we here present a new method which generates and modulates delay time by pulse picking as illustrated below. Laser pulses from two Ti:Sapphire lasers were introduced into two pulse pickers, respectively. With each pulse-picker, we transmitted one pulse out of every 90 pulses by blocking other 89 pulses. Consequently, the periodicity of the transmitted pulses became 90 times of the periodicity of original laser pulses,  $T_{\text{rep}0}$ . By transmitting pulses with displacement of  $n$  pluses between two pulse lines, we could generate delay time of  $n$  times of the original pulse periodicity  $T_{\text{rep}0}$ . Simultaneously, delay time shorter than the repetition periodicity  $T_{\text{rep}0}$  was realized by displacing the timing of oscillation of two laser oscillators.

Adopting this method, the delay time could be modulated with a large amplitude,  $\sim 1 \mu\text{s}$  at a high frequency,  $\sim 1 \text{ kHz}$ . Thus, the signal-to-noise ratio was much improved and we could finish, for instance, a measurement that took 10 hours by the first generation SPPX-STM, in a few minutes. In addition, since the delay-time could be changed discretely, the time resolution became independent of the modulation amplitude. Consequently, a better time resolution was obtained as well [2].

[1] O. Takeuchi et al., Appl. Phys. Lett. 85(15), 3268-3270 (2004).

[2] Y. Terada et al., Nature Photonics DOI :10.1038/NPHOTON.2010.235 (2010).



Conceptual illustration of SPPX-STM using delay-time modulation by pulse-picking