

## Supplementary Information

### Externally-triggerable optical pump-probe scanning tunneling microscopy with a time resolution of tens-picosecond

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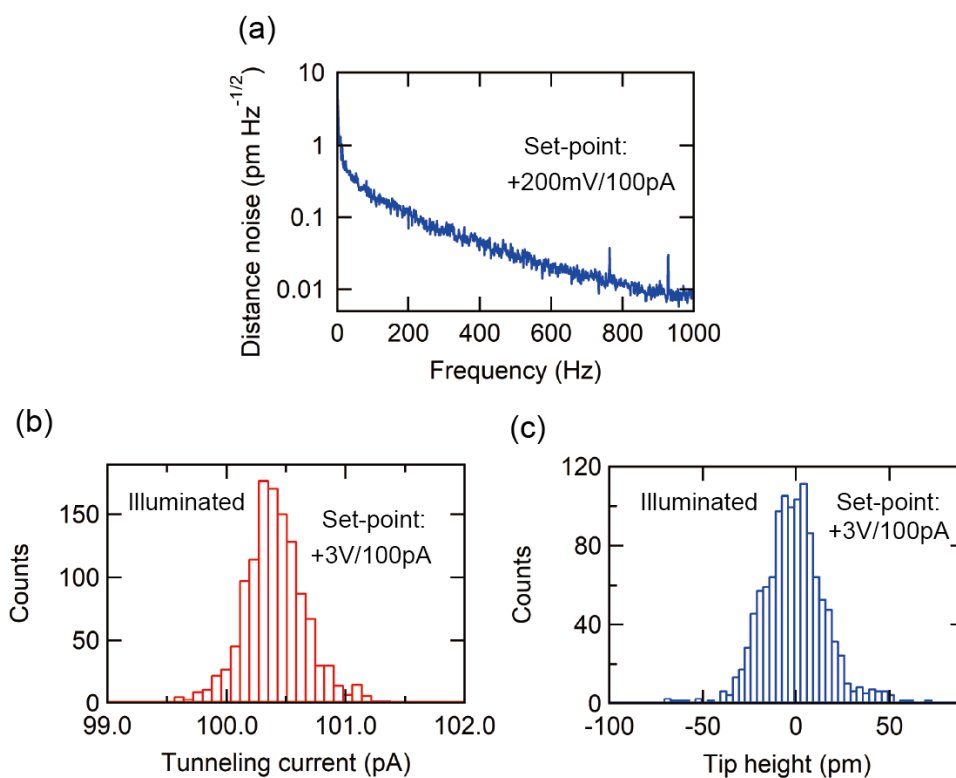


Figure S1. (a) Typical tip-sample distance noise spectrum taken when the feedback loop closed without laser illumination.  $T = 78$  K. The sample is highly ordered pyrolytic graphite (HOPG). Set-point:  $V = +200$  mV,  $I_t = 100$  pA. (b, c) Histogram of the tunneling current and the tip height with laser illumination on GaAs(110) surface for 12 hours, respectively.  $T = 6$  K. Set-point:  $V = +3$  V,  $I_t = 100$  pA. Laser power is 0.25 mW. The tunneling current and tip height were measured every 30 seconds. The average tunneling current (the tip height) and the standard deviation of tunneling current (tip height) are 100.4 pA (-2.2 pm) and 0.3 pA (20.1 pm), respectively.

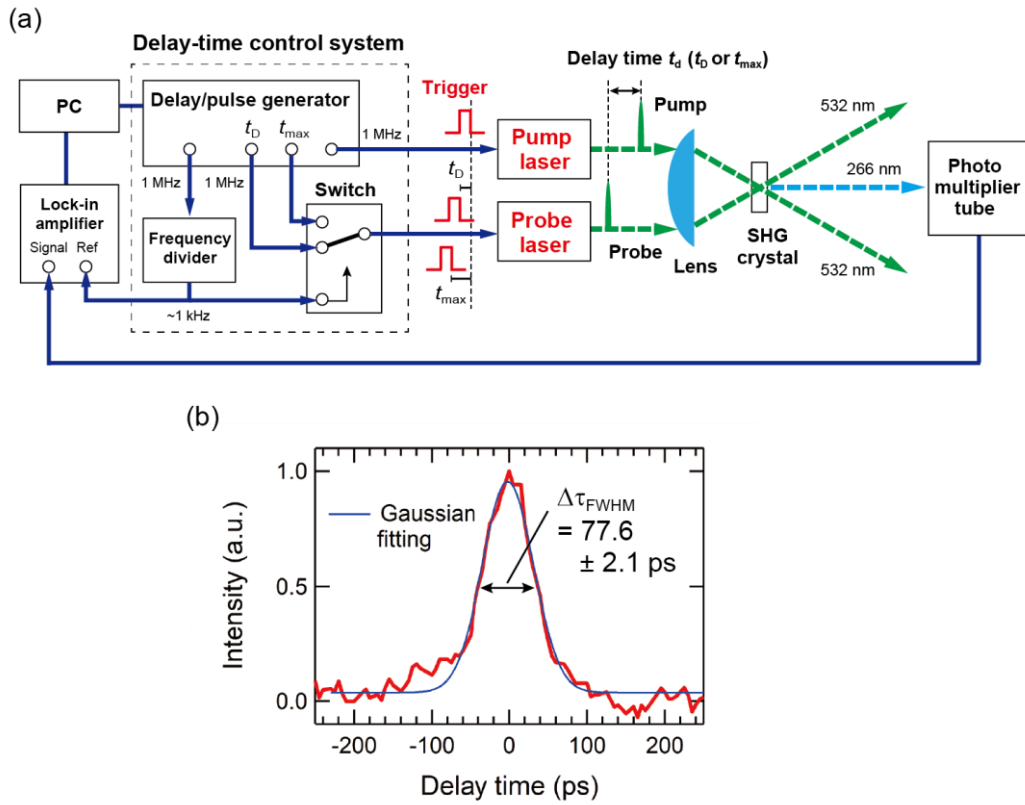


Figure S2. Cross-correlation measurement between pump and probe laser pulses using the sum frequency generation. (a) Schematic diagram of the system. SHG: second-harmonic generation. (b) Cross-correlation as a function of delay time. The correlation width estimated by Gaussian fitting is  $77.6 \pm 2.1$  ps.