Supporting information

Subcycle mid-infrared electric-field-driven scanning tunneling microscopy with a time resolution higher than 30 fs

Yusuke Arashida, *Hiroyuki Mogi*, *Masashi Ishikawa*, *Ippo Igarashi*, *Akira Hatanaka*, *Naoki Umeda*, *Jinbo Peng*, *Shoji Yoshida*, *Osamu Takeuchi*, and *Hidemi Shigekawa*[*]

Faculty of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-8573, Japan

Analysis of thermal expansion problem

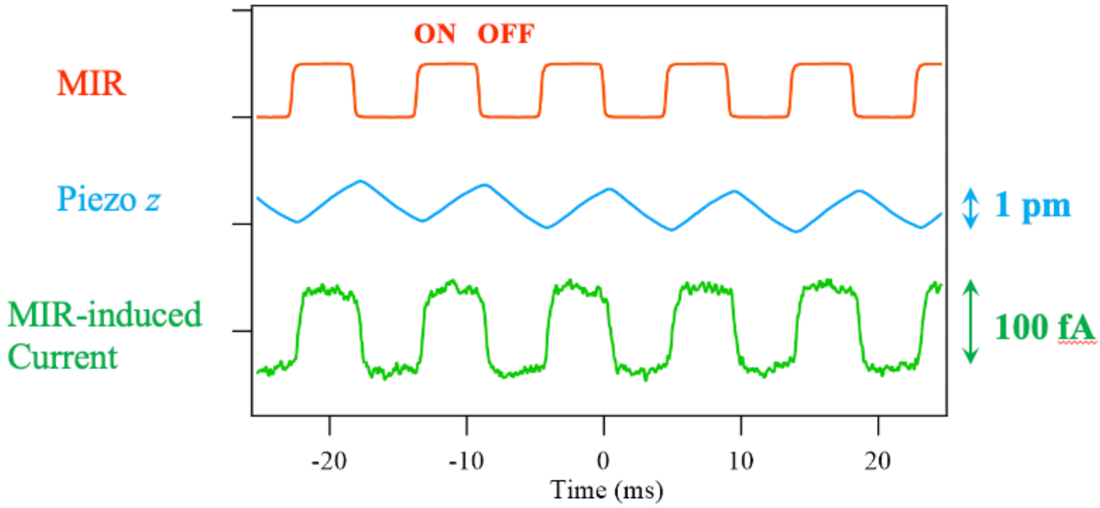

Fig. S1 Oscilloscope data (65536 integrations). Orange: MIR signal obtained when the light is turned on and off with an optical chopper. Blue: voltage proportional to the z displacement of the piezo element, measured using the change in orange signal as a trigger. Green: corresponding change in tunnel current.

Since the signal is weak in time-resolved STM, the lock-in detection method is used. Note that if the excitation light is in the visible or NIR region, the modulation of this intensity causes thermal expansion problems[1]. In THz-STM, the photon energy is small and, the therefore, thermal expansion problem does not occur, which is an advantage of this method[2]. In this study, the time resolution was improved by using MIR light whose frequency is about 30 times higher than THz; however, it was not obvious whether there would be thermal expansion due to the MIR light irradiation. Therefore, the MIR light intensity was modulated at a frequency of 120 Hz by an optical chopper, and the expansion and contraction of the piezo element and the change in tunnel current were measured with an oscilloscope. The change in the piezo element was 1 pm and the effect on the tunnel current was small. In fact, the measured MIR-induced current remained almost unchanged. It was thus confirmed that the thermal expansion problem could be ignored for this system.

Evaluation of MIR far-field

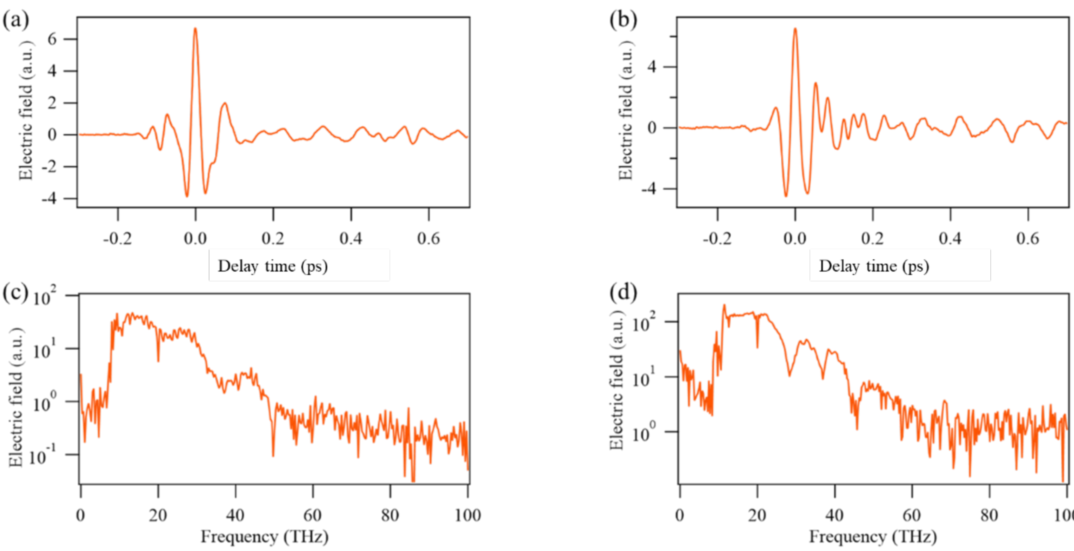

Fig. S2 (a) Temporal waveform of the MIR far-field measured by EO sampling in air. A GaSe crystal with the thickness of 30 μm was used. (b) Temporal waveform of the MIR far-field measured by using a photoconductive antenna (PCA) in the vacuum chamber. (c) and (d) the Fourier spectra of (a) and (b), respectively.

To carry out time-resolved MIR-STM measurement correctly, it is first necessary to confirm that the incident MIR far-field is reproduced at the STM tip. Therefore, the electric field waveforms of the incident far-field and the far-field at the STM tip were measured by EO sampling and photoconductive antenna (PCA) methods, respectively (Fig. S2). The PCA is an element in which an antenna structure such as a dipole antenna is patterned with a metal on a semiconductor substrate. The dipole antenna has two convex structures of gold electrodes facing each other with a gap of 6 ~~5 to 10~~ μm between them. When the PCA is irradiated with NIR light, electrons are excited from the valence band to the conduction band in the LT-GaAs (Low temperature grown GaAs) substrate, thereby generating optical carriers. Without an electric field, no current flows because the circuit is closed owing to the gap, but when an electric field is applied in this state, a current proportional to the electric field strength flows. Therefore, the MIR waveform can be obtained by measuring the change in current while changing the delay time between the MIR light and the NIR light similarly to the EO sampling method. This element was placed at the position of the STM tip in the STM measurement setup.

Reconstruction of the near field voltage $V_{NF, MIR}$ (t) from $I_{NF, MIR}$ (t)

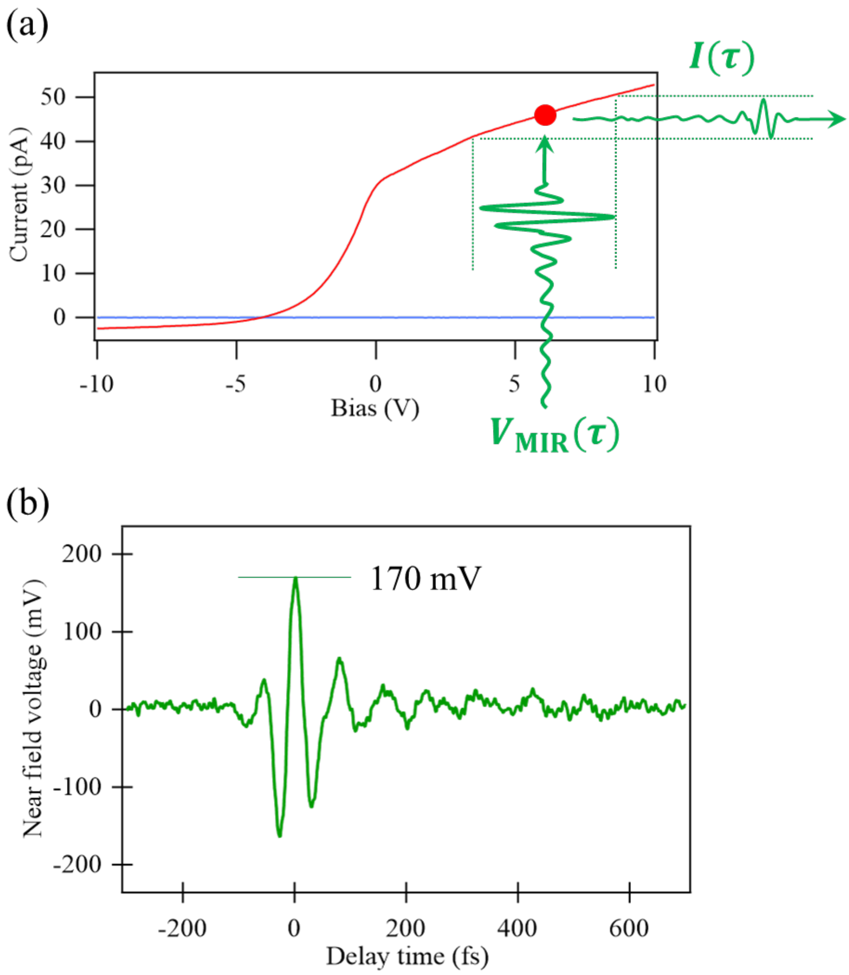

Fig. S3 (a) Relationship between the applied electric field (voltage) $E_{NF,\ MIR}(t)$ ($V_{NF,\ MIR}$(t)) and the current $I(t)$ obtained from I- V_{DC} curve shown in Fig. 2(b). (b) $V_{NF,\ MIR}(t)$ calculated using this method from the $I_{NF,\ MIR}(t)$ waveform for V_S = 6 V shown in Fig. 2(c).

FDTD calculation of MIR near-field waveform

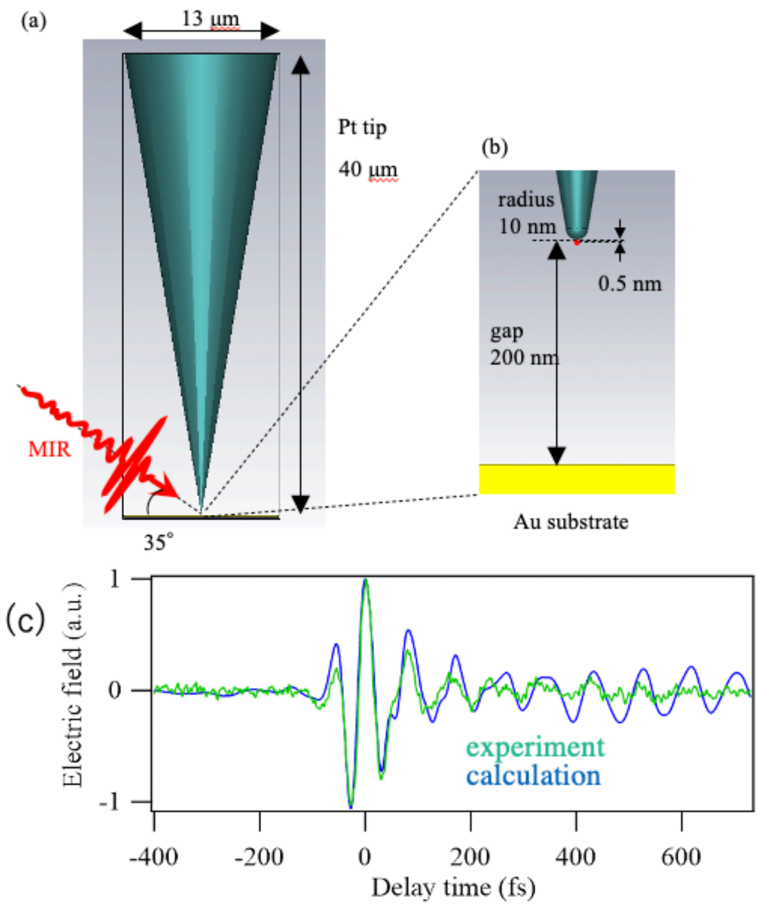

Fig. S4 To evaluate the MIR near-field waveform experimentally obtained by the evaluation method using photoelectrons, the near-field electromagnetic field was calculated in the time domain by the finite-difference time-domain (FDTD) method. (a) Shape of the tip and the arrangement of the tip and sample used in the calculation. A Pt cone with a length of 40 µm and a bottom diameter of 1 nm was used as the tip model for the calculation. (b) Magnified image of part of (a). A gold substrate was placed 200 nm from the tip. A subcycle MIR pulse was irradiated at an angle of 35 ° with respect to the gold sample. The component of the electric field in the direction perpendicular to the gold substrate was calculated at the position 0.5 nm from the tip shown by a red point in (b). The minimum mesh size was 0.6×0.6×0.13 nm^3. (c) Result obtained by the calculation (blue). The far-field electric field waveform used in the calculation was measured using the PCA shown in Fig. S3 and is reproduced in the figure (green).

Reference

1. You, A., Be, M. A. Y. & In, I. Laser-induced thermal expansion of a scanning tunneling microscope tip measured with an atomic force microscope cantilever. **2521**, 1–4 (2002).
2. Cocker, T. L. *et al.* An ultrafast terahertz scanning tunnelling microscope. *Nat. Photonics* **7**, 620–625 (2013).