Supplementary information

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Supplementary note 1

In time-resolved STM, initially, absorption bleaching-type time-resolved STM, similar to the optical pump-probe method, was realized[1–3]. Subsequently, the development of electric-field-driven time-resolved STM, which utilizes subcycle light with a controlled carrier envelope phase (CEP) as an instantaneous bias voltage, was achieved[4–10]. In the absorption bleaching-type time-resolved STM, by modulating the delay time or polarity of circularly polarized light, one can perform stable measurements with a high signal-to-noise (S/N) ratio by eliminating the effects of heat. This technique enabled the observation of phenomena in real space such as the impact of single atomic-level defects on carrier dynamics and spin dynamics within individual quantum wells[11,12]. Extensions to multiprobe STM are also advancing[13–15]. In electric-field-driven time-resolved STM, it is possible to evaluate the energy levels and densities of states of instantaneous excitation states[4–9]. Initially, when using 1 ps THz pulse light, the time resolution was approximately 500 fs[4,6–10,16]. However, a time resolution of less than 30 fs has been achieved by employing mid-infrared pulse light, enabling the analysis of band structure changes in non-equilibrium states[5].

Supplementary note 2

For instance, in the case of p-type GaAs (doping concentration, 3.0 x 10^{17} cm^{-3}; depletion layer width, 45 nm), it has been revealed by time-resolved photoelectron spectroscopy that it takes a rise time of about 50 ps[17]. For the bulk WSe2 used in this study, considering the very low mobility in the crystal c-axis direction (conductivity anisotropy, ~1200 [18]) and the possibility that the depletion layer width might be around ~120 nm[19], it can be assumed that the rise time of SPV is longer than the 45 ps laser pulse width used in this experiment.

Supplementary figure

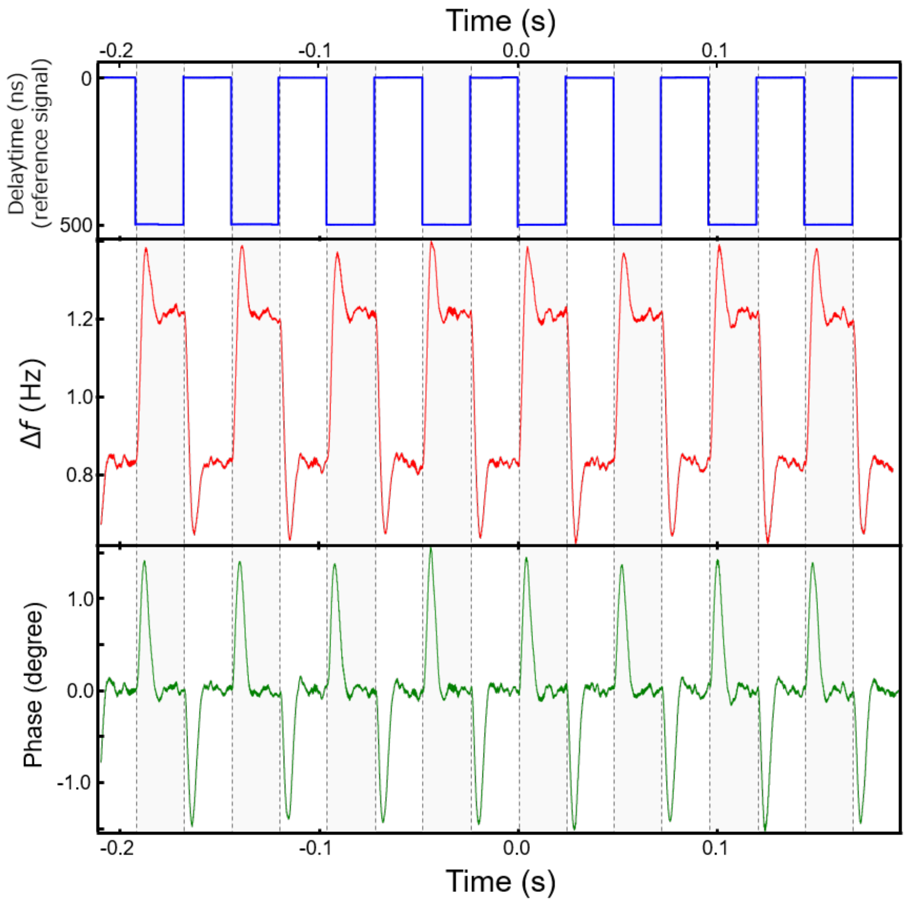

Fig. S1 Results of measuring Δf responsive to the delay time modulation and the time variation of the phase on an oscilloscope ($V_s = 0$ V, Δf setpoint = +1 Hz, delay time modulation frequency = 20 Hz). Because we did not perform lock-in measurements, the effect of noise was pronounced. To increase the S/N ratio, we increased the irradiation light intensity to approximately 7 mW to boost the signal strength and averaged 256 signal curves. First, it was confirmed that the phase-locked loop (PLL) feedback adequately tracks the frequency shift changes (PLL gain setup: PI gain = 1.3 Hz/deg, PI integral cut off frequency = 80 Hz). As the delay time switching occurs within one pulse cycle (2 μs), which is considerably faster than the PLL feedback bandwidth, an overshoot peak appears depending on the PLL feedback system's phase delay and gain settings.

This effect can be minimized by measuring only the in-phase component with a lock-in amplifier. The attractive force was stronger when the delay was shorter, indicating that we captured the time-resolved signal through the image dipole force. Using a tuning fork and setting the measurement conditions to the repulsive region with a small amplitude of 4 nm, the measurement of the near-field forces by PiFM became straightforward.

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