Generation of sub-cycle mid-infrared pulses with high-repetition rate for ultrafast time-resolved scanning tunneling microscopy

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Combination of phase-controlled sub-cycle terahertz (THz) pulses with scanning tunneling microscopy (STM), called as THz-STM [1-3], has enabled to study exotic surface phenomena with sub-picosecond (ps) temporal resolution and atomic resolution. Recent progress in femtosecond (fs) laser technology has made it possible to generate intense mid-infrared (MIR) pulses that further improve the time-resolution of STM, reaching to several tens of fs. The MIR-STM is therefore capable of observing and manipulating ultrafast physical and chemical properties such as intramolecular vibration and charge transfer at the single molecular level. To develop the MIR-STM, there are technical requirements on the MIR pulses: (a) broadband sub-cycle pulses with stable carrier-envelope phase, (b) high-peak electric field which can induce electron tunneling at a single tunnel junction, and (c) high-repetition frequency to achieve sufficient signal-to-noise ratio. In this study, we demonstrate the generation of intense sub-cycle MIR pulses with high-repetition frequency, which satisfy the all requirement above, using an optical parametric chirped pulse amplifier (OPCPA) as a fundamental light. The OPCPA has the pulse duration of 8.2 fs, the repetition frequency of 4 MHz, the average power of 4.3 W, and the wavelength range of 660-940 nm. To generate MIR pulses, we utilized an optical rectification with type I phase matching by using a GaSe crystal with a thickness of 20 μm. Figure 1 shows the waveform of the MIR pulses measured by an electro-optic sampling using a GaSe crystal. The pulse duration of the envelope of the electric field was 31 fs only with 0.85 cycle oscillation, while the peak field was 190 kV/cm which is sufficient to drive the electron tunneling. We also confirmed that the CEP of MIR pulses is very stable with the fluctuation of 48 mrad for 5.6 hours. These features are ideal for lightwave-driven MIR-STM [4].

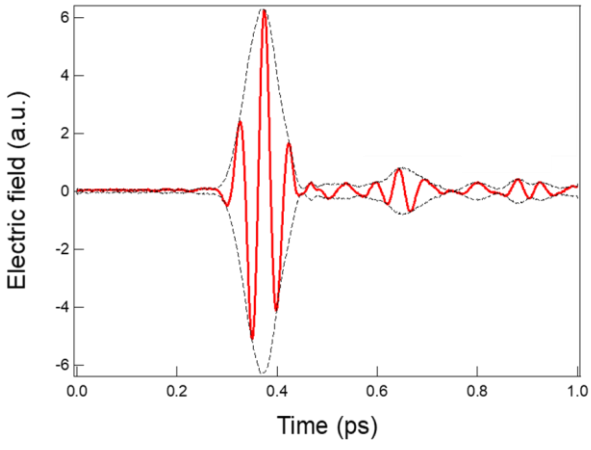

Fig. The waveform of MIR pulse.

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