Femtosecond time-resolved photoemission spectroscopy by using high-repetition rate Yb-fiber laser system

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Abstract: Time-resolved photoemission spectroscopy using the 5th harmonic of a mode-locked, 95-MHz Yb-fiber laser enabled the detection of ultrafast electron dynamics in bismuth induced by a pump density as small as 30 nJ/mm^2.
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1. Introduction

A non-equilibrated state of matter triggered by an ultra-short light pulse is of great interest from both fundamental and application points of view. Interactions and collisions through pulse-induced excitations in electronic, vibronic, and spin degrees of freedom can result in cooperative phenomena such as ultrafast photo-induced phase transitions and coherent oscillations [1]. From an electronic perspective, Time-resolved photoemission spectroscopy (TrPES) utilizing a pump-probe method is a powerful tool to investigate the out-of-equilibrium dynamics, because it captures the electron distribution therein. Typically, TrPES on the femtosecond scale has been achieved using a Ti:Sapphire laser system operating at a repetition rate less than 300 kHz, and has successfully revealed the non-equilibrium electronic dynamics induced by an energy as high as>10 μJ/cm^2. However, One of the natural ways to understand out of equilibrium dynamics of systems is in the low-pump-density regime. To this end, achieving a higher repetition rate in TrPES is crucial so that small changes occurring in the spectrum are detected. The 5th harmonic of a Yb-fiber laser is suitable for realizing high-repetition-rate 6 eV light with femtosecond pulse duration, since power amplification can be achieved without a decrease in the repetition rate.

In this paper, we describe a femtosecond TrPES with low density pumping and investigate the electron dynamics of Bi, which shows coherent phonon oscillation as a consequence of electron-lattice interaction at high-pumping density [2].

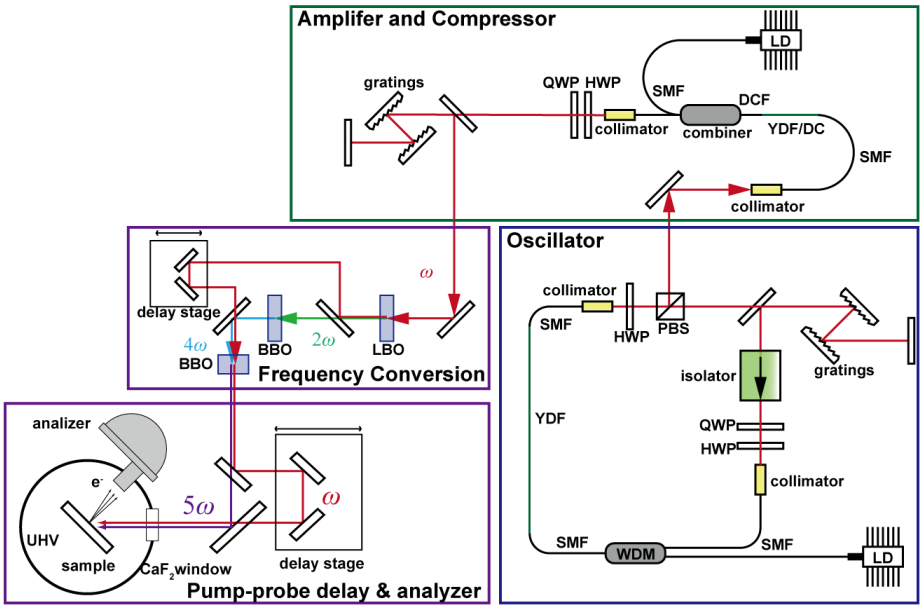

Figure 1: TrPES apparatus composed of a Yb-Fiber laser system and a hemispherical analyzer.

2. Experiment and results

Figure 1 is a schematic diagram of the experimental configuration used for TrPES, with the 95-MHz Yb-fiber laser system. A ring-cavity, nonlinear-polarization-evolution mode-locked Yb-doped fiber oscillator enabled the generation of femtosecond pulses centered at a pulse energy of 1.2 eV (1040 nm) with an average power of 15 mW. The seed laser pulse generated from the oscillator is amplified to 2 W, compressed by a grating pair, then directed to

the frequency conversion stage. The fundamental pulse energy of 1.2 eV is quadrupled by using successive LBO and BBO crystals, and the fourth harmonic light is added to the fundamental light to generate the 5th harmonic using a BBO crystal. The average power of the 6 eV pulse was 0.3 mW. The pulse duration of the fundamental and the 5th harmonic was estimated to about 100 fs and 250 fs, respectively. The fundamental pulse served as the pump, and the 5th harmonic pulse was used as a probe in the TrPES experiment described below.

Figure 2 shows the TrPES results of bismuth(Bi) acquired at room temperature. The pump density was set to 30 nJ/mm^2, which is more than two orders of magnitude lower than the usual pump-probe experiments done at < 300 kHz. Figure 2(a) reveals the population of unoccupied side by the pump, and Figure 2(b) shows the subsequent recovery of the excited carriers. The variation occurring in the unoccupied side is five orders of magnitude smaller than the intensity at the Fermi energy. The time resolution of TrPES is estimated through fitting the leading edge of the transient change. The recovery time depends on electron energy as shown in Figure 2(c) and becomes shorter at higher energy. This is compatible to the cooling of the excited-electron ensemble. A phonon bottleneck effect was not observed, and the recovery time can be described by an exponential-type decay with the time constants of around ps. This indicates that there was little energy transfer from the electrons to the lattice, presumably due to the low-density pumping. The decay time being approximately a picosecond reveals that the ultrafast dynamics in a Bi semimetal are similar to those of a metal rather than those of a semiconductor, the latter typically showing transient change lasting for >100 ps.

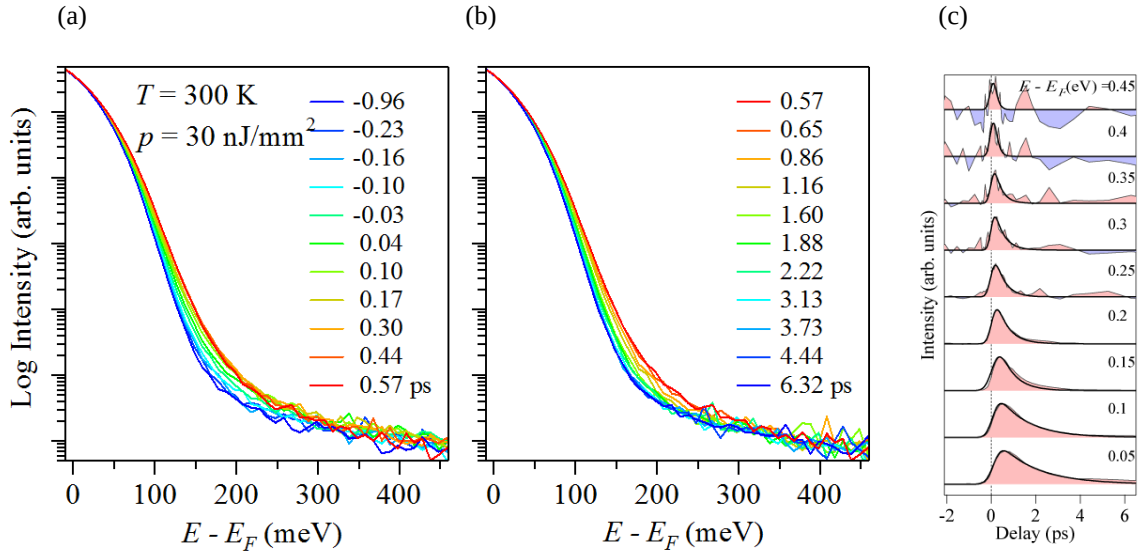

Figure 2: TrPES of Bi. Spectra at t < 0.57 ps (a) and at t > 0.57 ps (b). (c) Transient variation at each energy. The bold line is an exponential-type decay function broadened with a Gaussian of 380 fs full width at half-maximum, representing the time resolution.

3. Conclusion

In conclusion, we developed a 95 MHz, 6 eV Yb-fiber laser system for TrPES and investigated the carrier dynamics of Bi. We successfully recorded the spectral changes induced by a pump density as low as 30 nJ/mm^2 with a time resolution of 380 fs. We observed an exponential-type recovery of ~3 ps in the electronic excitation, where the recovery did not exhibit signatures of coherent phonon excitations or phonon bottleneck effect. This indicates that the non-equilibrium electronic dynamics in a semimetal Bi at low-density pumping are similar to those of a metal.

4. Reference

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[2] H.J. Zeiger *et al*. Phys. Rev. B. 45, 2 (1992)