# Development of soft x-ray time-resolved photoemission spectroscopy system at SPring-8 BL07LSU

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## **Introduction**

Synchrotron radiation (SR) has been one of the most important light sources for material science. Varieties of measurement methods have revealed structures, electronic states and spin characters of many materials. Recently, there have been growing interests in extending these experimental techniques to time-resolved measurements to trace dynamical phenomena, such as chemical reactions and phase transitions, in real time. Since a pulse width of SR is typically several tens picoseconds (ps), picosecond time-resolved experiment can be carried out by the pump-probe method with ultrafast pulsed laser.

Time resolved photoemission spectroscopy (TRPES) is one of most powerful tool to directly understand electronic states in dynamical processes. However, in TRPES the repetition rate of measurement should be low to wait for relaxation of sample to initial state from excited state. In this condition, high efficiency analyzer has been needed.

In this reports, we show the detail of developed TRPES system and show the result of TRPES measurement of surface photovoltage effect of  $Si(111)7 \times 7$ .

#### **Experimental setup**

We have developed TRPES system with a time-of-flight (TOF) type analyzer, ARTOF10k (VG Scienta)[1]. Our system is composed of four components, (1) SR beamline, (2) TOF type analyzer, (3) laser system equipped with synchronizing feedback circuit and (4) timing control unit. Since a TOF type analyzer is slit less, for the analyzer, high electron transmission was reported as compared with a hemispherical type analyzer at the same energy resolution. Laser pulses are synchronized with SR within the 10 ps jittering by feedback circuit. The most important part is the timing control unit. It controls the delay between laser and SR with in the

1 ps accuracy, and makes trigger signal for analyzer. The SR pulse has complex patterns. As an example, the operation mode called "D-mode" (5 bunches and 1/7 filling part) is shown as the bottom signal in Fig.1. In such operation mode, the ARTOF10k can measure only the 5 bunches part. The signal from the filling part cannot be resolved due to the short interval time. To measure spectra only from the bunches, the timing control circuit makes the three signals: Ch1, Ch2 and synthetic signal. With the synthetic signal, the analyzer can utilize the bunch part of the operation mode of SPring-8 and be blind to the filling part.



Fig 1 Time dependence of signals for the analyzer created in the timing control circuit, and SR pulses detected at the 0th order light monitor in the beamline. Ch1 is for gating SR light from the filling part. Ch2 is 7 signals per a cycle, and its frequency corresponds to each bunch and the start and end of the filled section. A synthetic signal is made by Ch1 and Ch2 passing through the "and" circuit. The SR monitor (bottom) is used to determine the timing of gating.

The timing between laser and SR was measured by photodiode at sample position. The measured signal is shown in Fig.2(a). The highest signal came from laser pulse. By the photodiode, the delay time between laser and SR can be determined in the tens ps accuracy, which corresponds to pulse width of SR.

## **Results**

To demonstrate the performance of the system, we measured surface photvoltage (SPV) of Si(111)7 $\times$ 7 surface. The sample is n-type silicon wafer with 0.01 Ωcm resistivity. The SPV effect is photo-induced reduction of bulk band bending near a semiconductor It is caused by creation surface. of electron-hole pairs after the light irradiation, followed by accumulation of one of the carriers at a surface. Relaxation time has been known to take in nanosecond-to-microsecond time-scale, depending mainly on doping concentration in a semiconductor wafer. We measure the phenomenon with the peak shift of Si 2p spectra with photon energy hv=252eV since the energy difference between valence band and Si core level is rigid. The laser pump power is set to  $70\mu$ J/cm<sup>2</sup> and the



Fig 2 (a) SR and laser signals detected by a PD at the sample position. The PD signal of SR pulses reproduces the bunch structure of SPring-8. Each single bunch is labeled in the order from the bunch train of SR. The additional signal with the highest intensity originates from the laser pulse. (b) Energy shift of Si 2p core level as a function of delay time for the time-resolved photoemission measurement of the surface photo voltage (SPV) effect on Si(111)  $7 \times 7$ . Circles, triangles, inverted triangles, and squares correspond to the data points measured with the second, third, fourth and fifth electron bunch, respectively. The solid line represents the result of fitting.

pump energy is hv=1.55 eV. The repetition rate of measurement is ~1kHz. The result is shown in Fig.2(b). At one measurement, we can take 5 points of data from different bunches. For fitting these data, we set two exponentials and get two relaxation time,  $\tau_{\text{fast}} = 3.3\pm0.80$  ns and  $\tau_{\text{slow}} = 500\pm25$  ns.

## **Conclusion**

We developed time-resolved photoemission system at SPring-8 BL07LSU. The synchronization between laser and SR was achieved within 10 ps jittering. The timing between laser and SR was measured by photodiode at sample position and determined within the accuracy of SR pulse width. The time-resolved measurement of surface photovoltage effect on Si(111)7×7 surface was performed. The result showed two relaxation processes.

## References

[1] M. Ogawa, S. Yamamoto, Y. Kousa, F. Nakamura, R.Yukawa, A. Fukushima, A. Harasawa, H. Kondo, Y.Tanaka, A. Kakizaki, and I. Matsuda, Rev. Sci. Instrum. **83**, 023109 (2012).