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X-ray Pump-Probe Technique

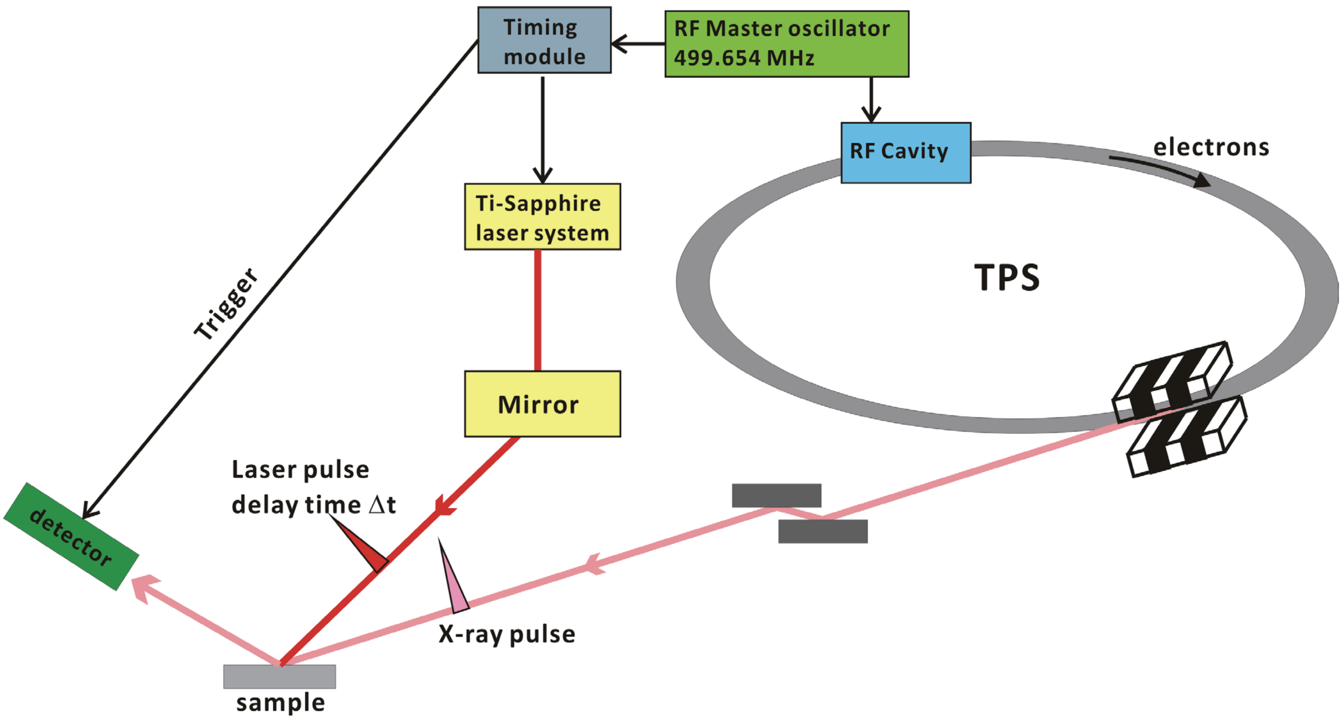

Fig. 1: Schematic diagram of the pump-probe experiment at **TPS 09A**.

An understanding of structural dynamics at a molecular level, such as the vibrations and rotations of single molecules or crystal lattices and the breaking and formation of chemical bonds, is most desirable in condensed-matter physics, chemistry, biology and materials science. At TPS, an X-ray source with pulse duration 20 ps rms can serve to perform a time-resolved experiment on a picosecond scale. At **TPS 09A**, the technique for time-resolved research involves using a laser as a pump and synchrotron radiation (SR) as a probe for time-resolved X-ray diffraction or scattering, allowing the structural dynamics induced by an ultrafast laser pulse to be studied. **Figure 1** shows a schematic diagram of the pump-probe experiment. The synchronization of the laser and X-ray pulses should be built up; on altering the delay between laser and X-rays we catch the scattering signal at varied timing to map an overall dynamic process. In addition, a particular filling pattern is necessary because of the limitation of the response time of the detector. In the next paragraph, we introduce what filling pattern is suitable and how to synchronize laser and X-ray pulses.

The TPS storage ring has circumference 518.4 m; the revolution period of the SR is 1.728 μs, corresponding to revolution frequency 578 kHz; the frequency of the RF cavity is 499.654 MHz, corresponding to 864 bunch buckets with interval 2 ns. In normal operation, electrons are injected into ~2/3 round of buckets. Under this condition, the pump-probe experiment cannot be executed because of the response time of the detector, which should be smaller than the bunch interval, but is actually 4 ns for our avalanche photodiode detector (APD). A pump-probe experiment hence requires a particular bunch-filling pattern to meet the requirement. At TPS, filling patterns of two kinds have been operated, a hybrid mode and a single-bunch mode, shown in **Fig. 2**. For the hybrid mode, about 2/3 round of buckets (300 mA) are filled with electrons and an isolated bunch of current 1.4 mA is injected at the middle of the 1/3 empty buckets. For the single-bunch mode, only one electron bunch with current 2 mA is injected into the bucket. As an experiment, an isolated X-ray bunch was chosen as the probe source, which provided a time resolution about 20 ps.

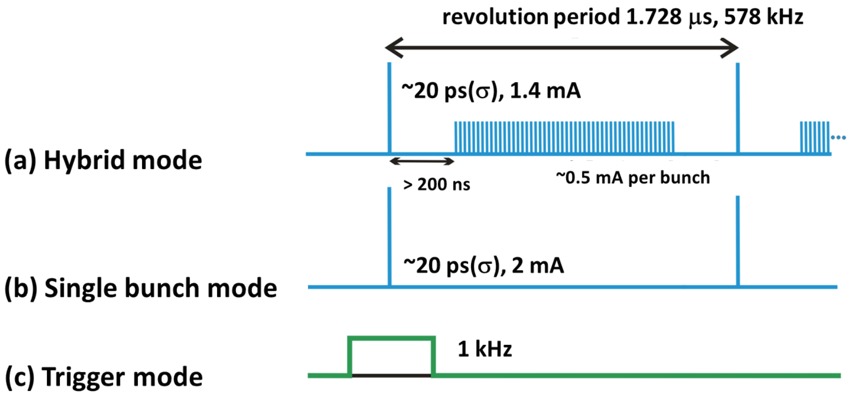

Fig. 2: Filling patterns for (a) hybrid mode and (b) single-bunch mode; (c) 1-kHz trigger signal sent to the APD detector to extract the synchronized signal.

The laser pulse is generated with a mode-locked Ti-sapphire laser, which produces a 35-fs pulse of wavelength about 790 nm. The repetition rate of the amplifier is 1 kHz; the timing is synchronized to the X-ray pulse. The laser system was housed in a laser hutch between endstation 1 (ES1) and endstation 2 (ES2) and guided to ES1 by reflecting mirrors in a series. In ES1, a laser pulse was focused to 1 x 1 mm^2 with a lens kit to irradiate a sample. Typical laser characteristics are listed in **Table 1.**

Table 1: Characteristics of Ti-sapphire laser system

Mode-lock IR laser	Micra (oscillator)	Legend (amplifier)
Average power	450 mW	4 W
Wavelength	790 nm	790 nm
Pulse width	< 35 fs	35 fs
Repetition rate	83.276 MHz	1 kHz
Pulse energy	5.4 nJ	4 mJ

The laser system must be synchronized with the synchrotron X-rays; the detailed signal control is shown in **Fig. 3**. An initial RF (radio frequency) master oscillator transmits main frequency 499.654 MHz to a RF cavity, a laser-timing system and a detector-trigger system. For the laser-timing system, a 1/6 divider divides the main frequency to 83.28 MHz; the divided signal is sent to the delay generator and synchronizer. The delay generator (CANDOX84DgR5CO2, trigger & clock delay module) can adjust the delay of the laser pulse; the synchronizer can synchronize these instruments including the delay generator, the oscillator (Micra) and the amplifier (Legend). The amplifier pro-

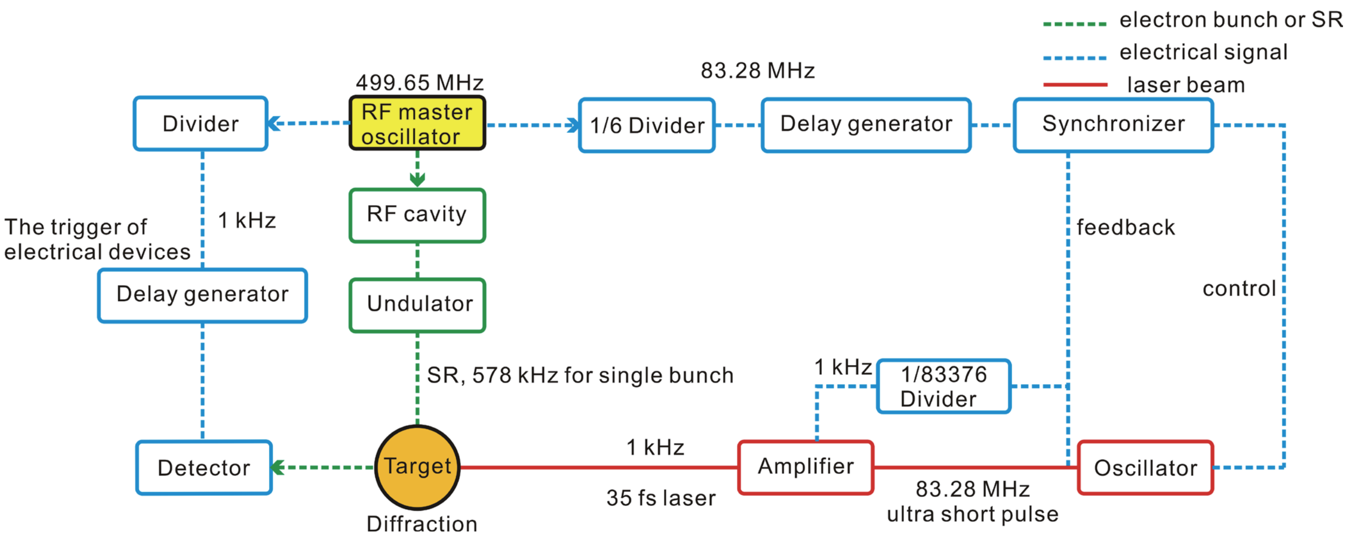

Fig. 3: Scheme of synchronization and delay control.

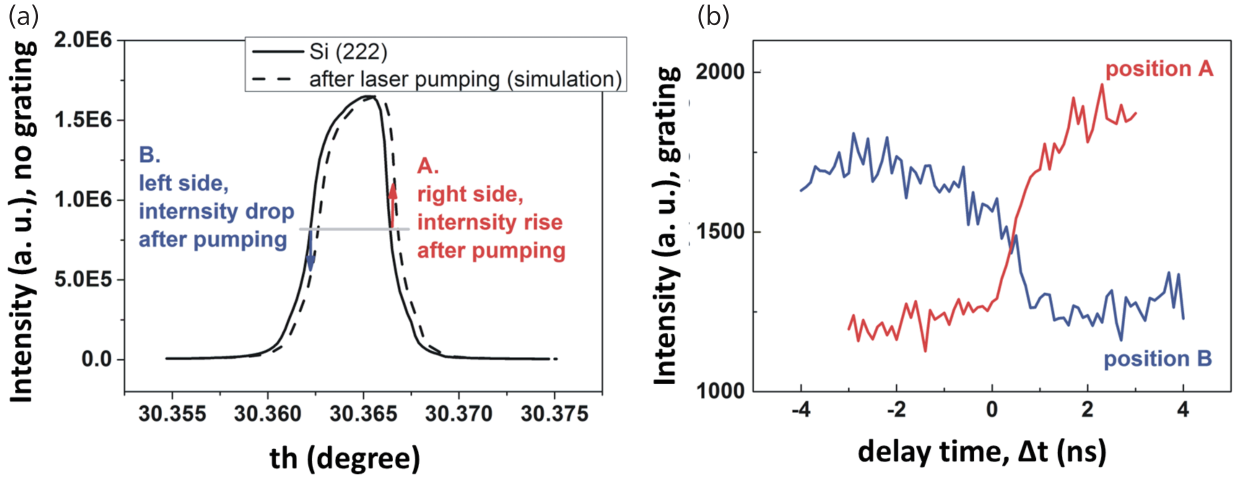

Fig. 4: (a) The black line is the rocking curve for the Si(222) reflection. After laser pumping, the diffraction angle was shifted to a larger angle (dotted line) through the lattice compression. (b) Time responses of the Bragg peak at positions A and B.

vides a 1-kHz laser pulse to irradiate a sample; as the repetition rate of the single-bunch X-ray pulse is 578 kHz, which is unmatched to the amplifier frequency, we use a 1-kHz trigger signal (**Fig. 2(c)**) to catch a synchronized signal from the detector to achieve the overall timing control.

Here we demonstrate a basic pump-probe experiment to ensure that the entire setup is ready. The temporal overlap of the laser pulse and the X-ray pulse can be precisely determined from this demonstrated experiment. In the experiment, using a short laser pulse (35 fs) with energy 0.73 mJ/pulse to irradiate a silicon crystal, the laser pulse induced a strain field in the crystal promoting the lattice expansion. Referring to Bragg's law, $2\,d \sin \theta_B = \lambda$, a lattice perturbation, Δd, leads to a deviation of the Bragg angle, $\Delta\theta = -(\Delta d/d) \tan \theta_B$.[1,2] After the laser pump, we can thus observe the deviation of the Bragg angle from the diffraction pattern with a X-ray probe. In **Fig. 4(a)**, the black line is the rocking curve of the Si(222) reflection before laser pumping. After laser pumping, the peak was shifted to larger q angle through compression of the lattice. Two positions A and B at half maximal intensity were chosen to measure the time response of the Bragg peak. The response time of the lattice compression was several picoseconds; we can thus observe the intensity rising and falling simultaneously at positions A and B. The measurement result is shown in **Fig. 4(b)**; the expected intensity rising and falling coincidently has been observed. The initial rising (falling) point at position A (B) was decided as the temporal overlap point, at which the delay time is equal to zero.

In conclusion, a time-resolved X-ray diffraction experiment, which combines 35-fs laser pulses and 20-ps X-ray pulses in a pump-probe technique, has been realized at **TPS 09A**. We have observed a lattice compression induced with an ultrafast laser pulse. Recently an optical parametric amplifier (OPA), which can extend the wavelength of the Ti-sapphire laser from 800 nm to the range 240-2600 nm has been installed in the laser hutch. In the future, a chopper system will be installed at the beamline to extract the X-ray pulse to decrease the repetition rate (~1 kHz) to be consistent with the frequency of the laser pulse.[3,4] This upgrading will increase the flexibility to study structural dynamics with a laser-pump/X-ray probe technique. (Reported by Ying-Yi Chang, Yi-Wei Tsai and Wei-Rein Liu)

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