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Text for the class,
“Pump and probe technique for picosecond time-resolved x-ray diffraction”
at the Cheiron School

1. Introduction

1-1. Purpose

The pulsed nature of the synchrotron radiation (SR) and the synchronization technique between SR pulse and ultrashort laser pulse enable us to make pump and probe measurements with 40 ps time resolution. The time resolved x-ray diffraction experiments will be demonstrated for the lattice dynamics of a single semiconductor crystal. The course may help you with starting the picosecond time-resolved experiments at your stations for investigation on fast structural dynamics.

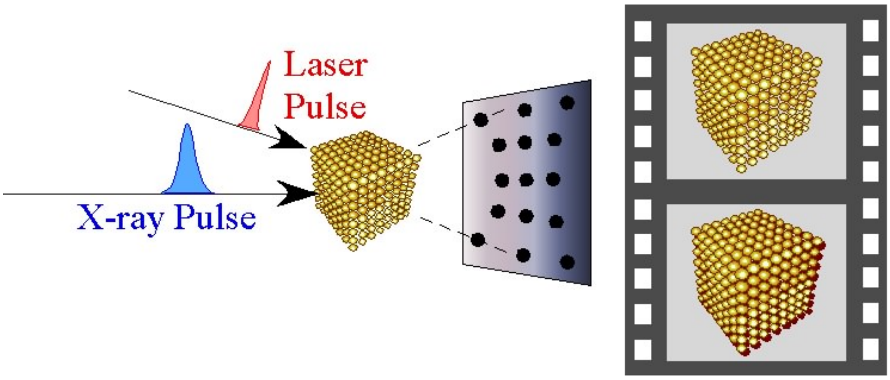


Fig. 1　Pump-probe method for time-resolved x-ray diffraction

1-2. Objectives

When a surface of GaAs single crystal is irradiated by a femtosecond pulse laser, the lattice expands with a response time of a few hundreds of ps. To observe the fast lattice response, a stroboscopic method in X-ray diffraction , i.e., laser-pump and SR-probe method is performed in the SPring-8 beamline.

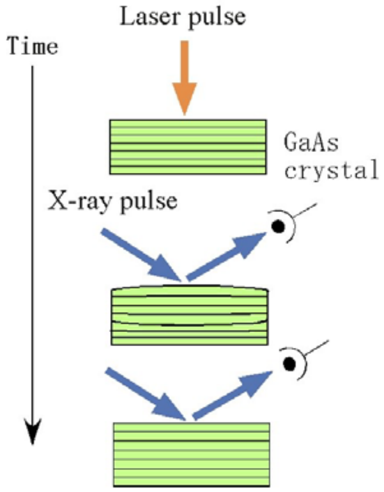


Fig. 2 Fast lattice expansion of a GaAs single crystal induced by femtosecond pulsed laser irradiation.

2. Experimental apparatus and equipments prepared

2-1. The hard X-ray beamline BL19LXU

The course will be opened at the beamline BL19LXU which can produce the most intense X-ray beam in the world at present. The SR beam generated by the 25 m-long undulator is guided to the experimental hutches through a Si 111 double-crystal monochromator (See ref. 1). The X-ray time-structure is determined by the electron bunches in the storage ring. The electron bunches have the pulse width of about 40 ps at full width at half maximum (FWHM), with a repetition rate of 1 MHz to 509 MHz. The repetition rate can be controlled by the ring operation with preferable filling pattern. In the class, the filling pattern operated is slightly complicated, which is called “hybrid mode”, to gain the bunch current for particular electron bunches as keeping a total current of 100 mA for the other users.

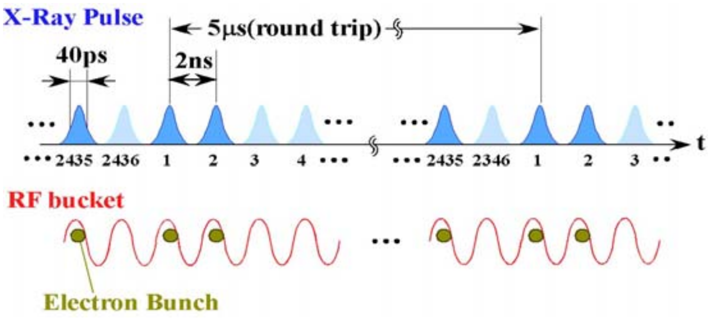


Fig. 3 Time structure of the SPring-8 SR pulses.

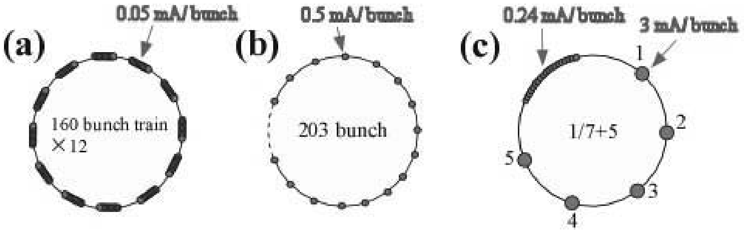


Fig.4 Examples of the filling patterns of electron bunches in the storage ring.

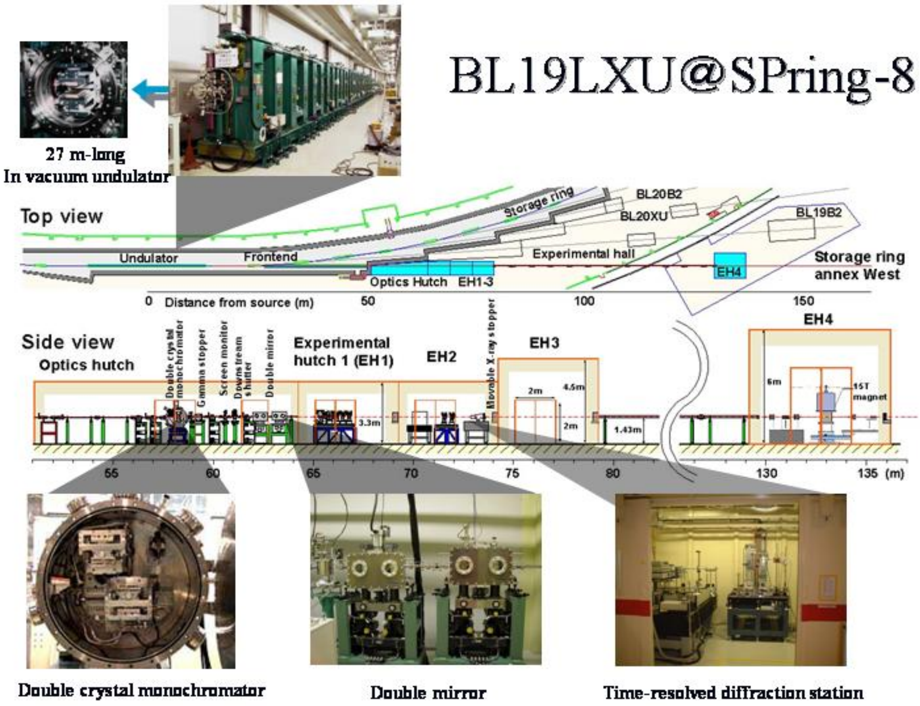


Fig. 5 Overview of the RIKEN beamline of BL19LXU.

2-2. Femtosecond pulsed laser system

A femtosecond laser system for irradiation of a GaAs crystal surface has been installed in the experimental hutch of the beamline. The laser system is composed of the mode-locked laser oscillator, and a regenerative amplifier to produce laser pulses with a pulse width of 130 fs and a pulse energy of about 1mJ/pulse. The laser pulses are synchronized with the master oscillator of the storage ring controlling the acceleration timing of the electron bunches. (See ref. 2)

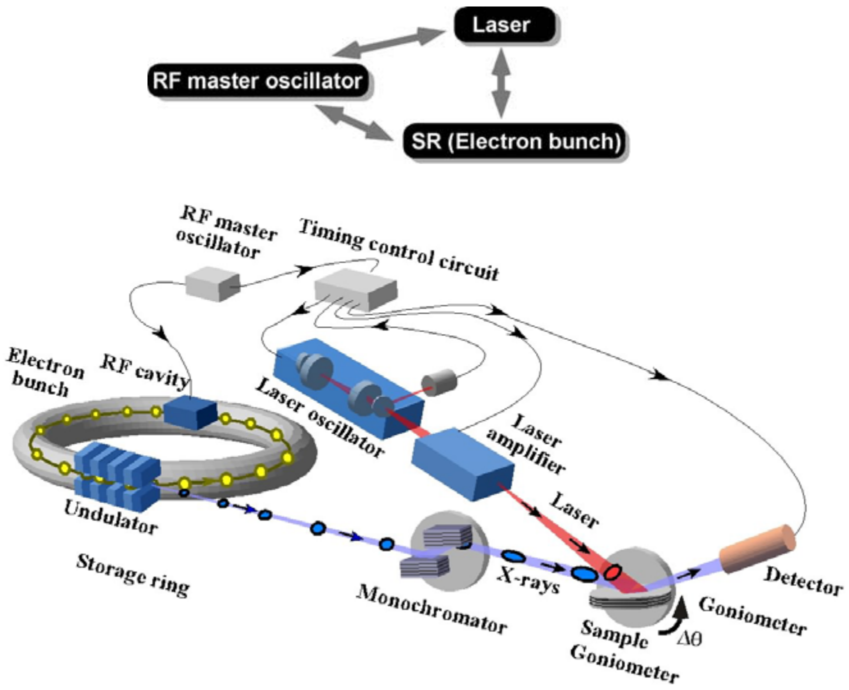


Fig. 6　Synchronization system of femtosecond laser and SR pulses.

3. Experimental procedure

(1) Set up for diffraction measurement of GaAs 400.

In order to observe the 400-Bragg reflection of a GaAs wafer, set the sample on the goniometer, and find the Bragg reflection with an ion chamber.

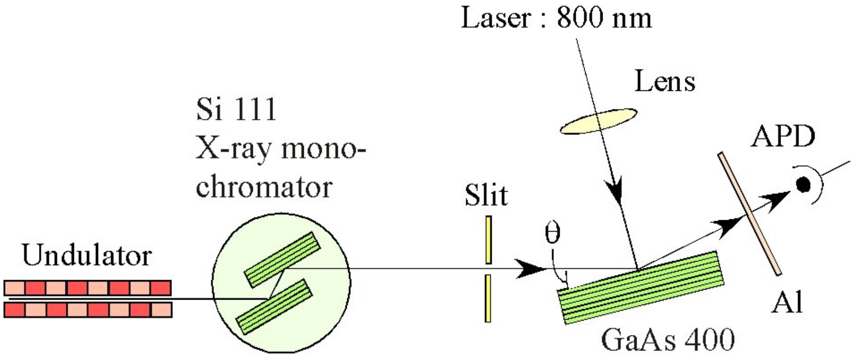


Fig. 7 Experimental setup for time-resolved measurement of fast lattice expansion of a GaAs wafer.

(2) Alignment of a femtosecond laser beam onto the sample.

Guide the femtosecond laser beam onto the sample surface so as to achieve the spatial overlap with the SR beam whose footprint is observed by a burned paper.

(3) Monitor of the synchronization between the laser pulse and the SR pulse.

An avalanche photodiode is used to monitor the timing of the laser and the SR pulses. Tune the pulse timing of the laser to be close to the target SR pulse as monitoring with an oscilloscope. Then, adjust the timing of electric gate to cover the target SR pulse signal.

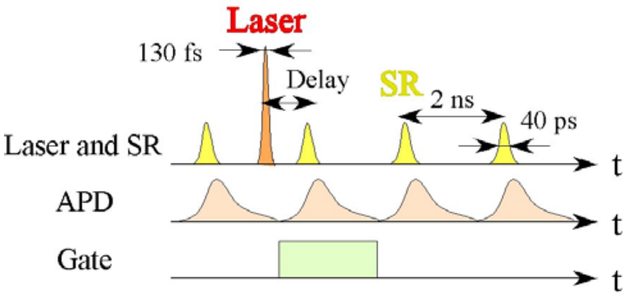


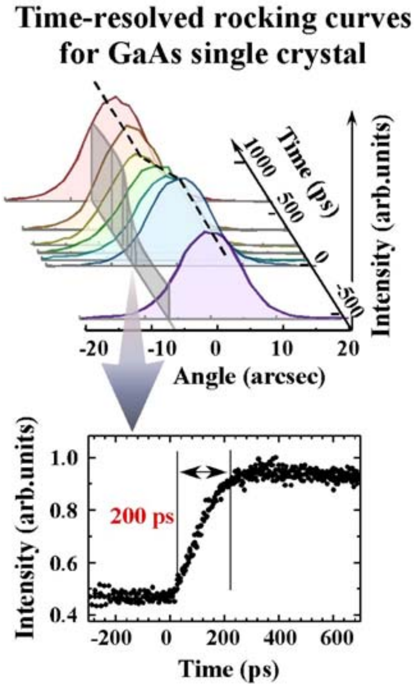
Fig. 8 Time chart of laser and SR pulses, together with electric gate signals.

*(4) **Control of the timing delay between the pulses.***

Scan the timing of the trigger signal for the femtosecond laser by an electronic circuit including a phase shifter. Then you can obtain the diffraction intensity dependent on the delay timing (Ref. 3).

4. Expected results

The X-ray diffraction intensity becomes large with a response time of about a few hundred ps at the smaller angle side of the Bragg diffraction profile, due to the Bragg peak shift. (See refs. 4 -6)


Time-resolved rocking curves for GaAs single crystal

5. References

(1) http://www.spring8.or.jp/wkg/BL19LXU/instrument/lang-en/INS-0000000361/instrument_summary_view

(2) Y. Tanaka *et al.*, Rev. Sci. Instrum., **71**, p.1268 (2000), “Timing control of an intense picosecond laser to the SPring-8 synchrotron radiation pulses”

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(5) Y. Hayashi *et al.*, Phys. Rev. Lett., **96**, p.115505 (2006), “Acoustic pulse echoes probed with time-resolved x-ray triple-crystal diffractometry”

(6) Y. Tanaka *et al.*, J. Phys. Conf. Ser., **278**, p.012018 (2011), “Time-resolved X-ray diffraction studies of laser induced acoustic pulse generation in semiconductors using synchrotron radiation”