Cheiron2007
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Pump-Probe Experiment

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My conclusion will be …

- **Pump-probe method with synchrotron radiation enables us to make movies at atomic resolution.**
- **The time resolution is currently ~ 100 picosecond (sub-nanosecond) order, but soon will be ~ 100 femtosecond (sub-picosecond) order.**

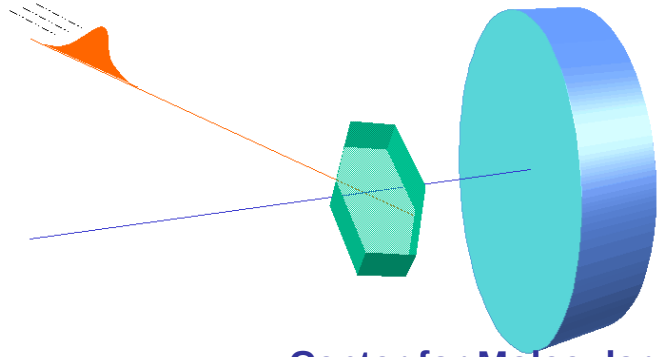
Outline of the talk

1. What is pump-probe method?
2. Pump-probe method with SR
   - Pulsed nature of SR
   - Synchronizing pump and probe
3. Applications
4. Concluding remarks

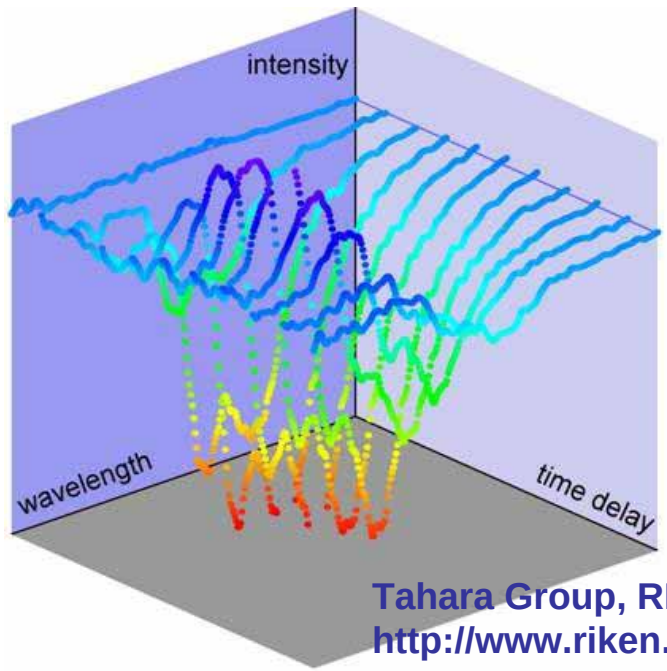
What is pump-probe method?

("Time-resolved spectroscopy" in Wikipedia)

In physics and physical chemistry, time-resolved spectroscopy is the study of dynamical processes in materials or chemical compounds by means of spectroscopic techniques. In a typical experiment, both the light for excitation ('pump') and the light for measuring the spectrum ('probe') are generated by a pulsed laser.



Center for Molecular Movies
http://cmm.risoe.dk/



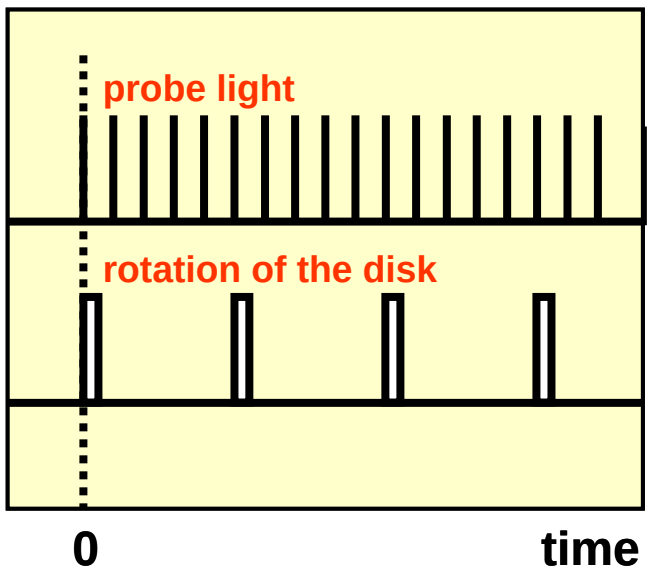
Tahara Group, RIKEN
http://www.riken.jp/

Pump-probe method as a tool for making movies (1)

We want to watch something moving!

Case 1: Continuous Light

Case 2: Pulsed Light



© Disney

Pump-probe method as a tool for making movies (2)

Watching the 6 guys dancing.

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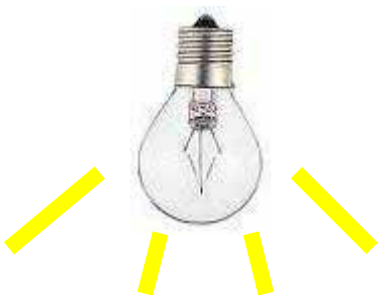
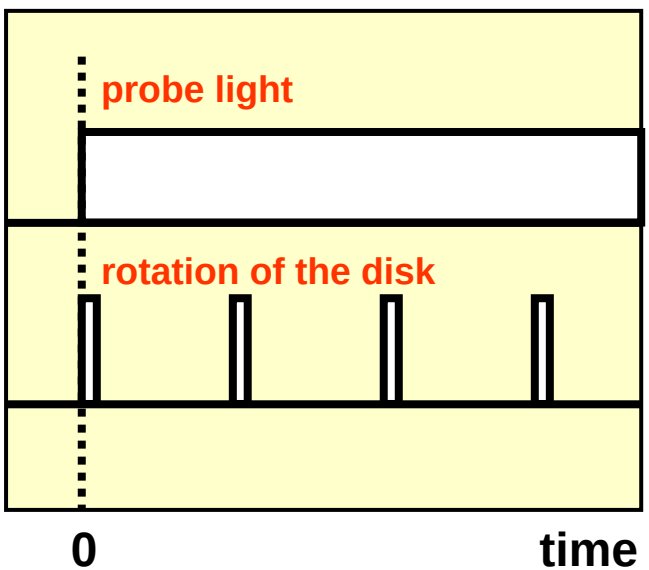
Pump-probe method as a tool for making movies (3)

Continuous vs. Pulsed Light

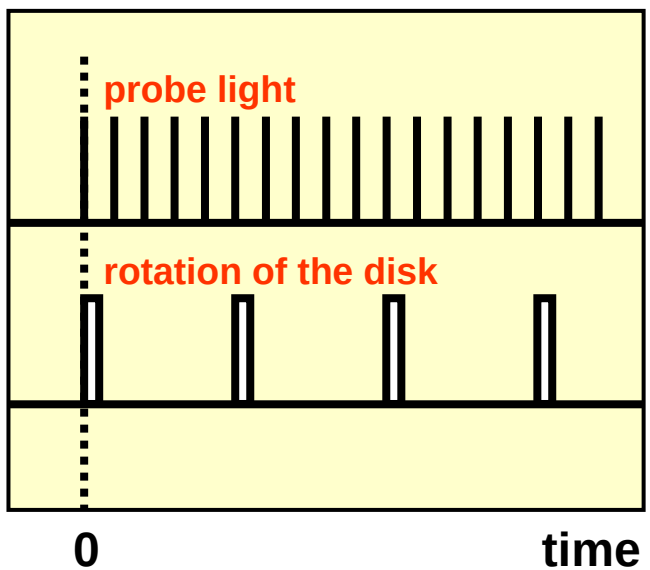
Case 1: Continuous Light





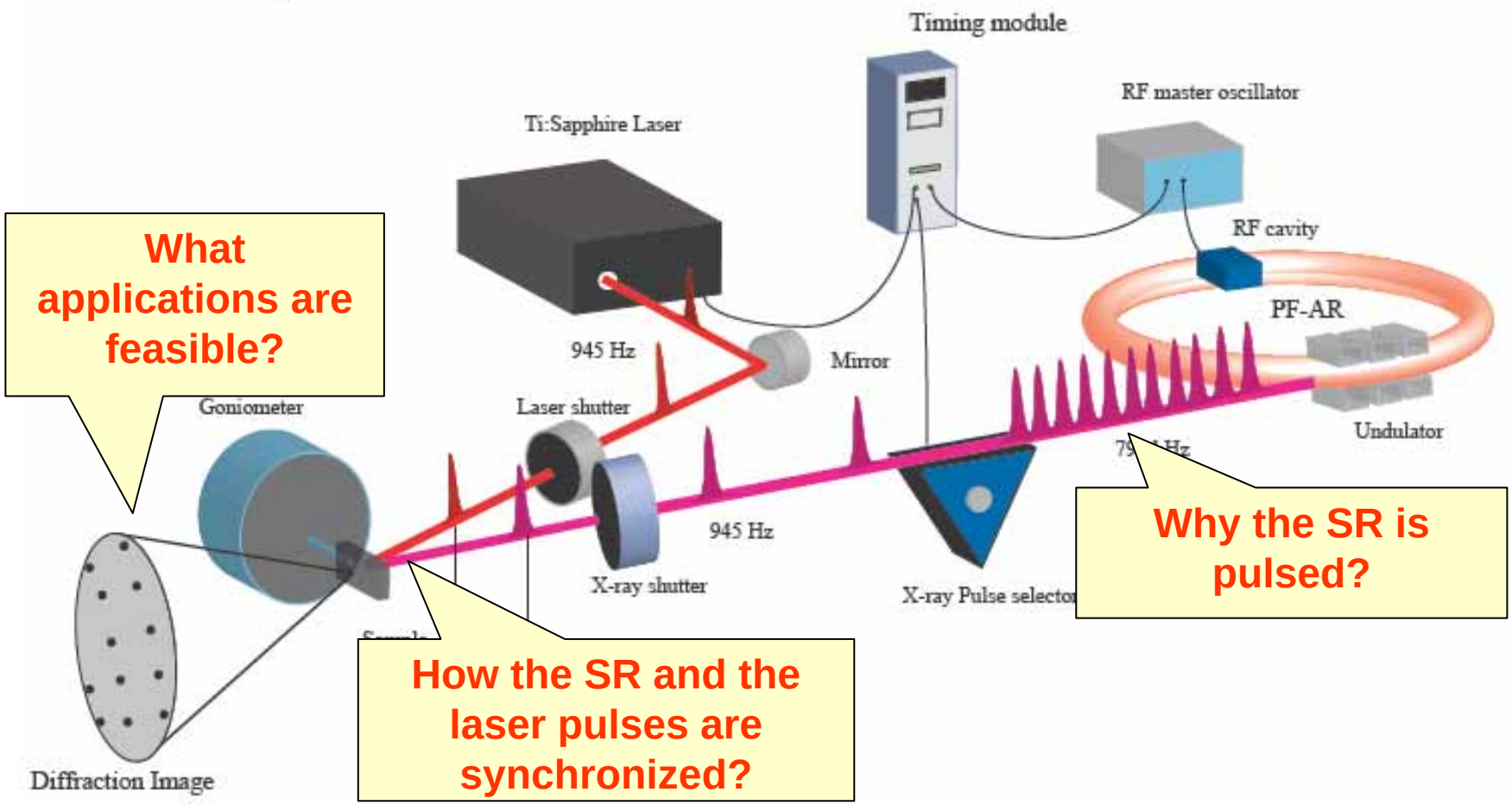
Case 2: Pulsed Light

Pump-probe movie!



Pump-probe method as a tool for making movies (4)

Pump-probe with SR



Summary #1
What is pump-probe method?

- **The pump-probe method enables us to make movies.**
- **We need pulsed light for it.**
- **Timing between the pump and the probe pulses will be one of the main issue.**

Summary #1

Still image	⟷	**Movie**
Static	⟷	**Dynamic**
Structure	⟷	**Mechanism**

Nicer?

Outline of the talk

1. What is pump-probe method?
2. Pump-probe method with SR
   - Pulsed nature of SR
   - Synchronizing pump and probe
3. Applications
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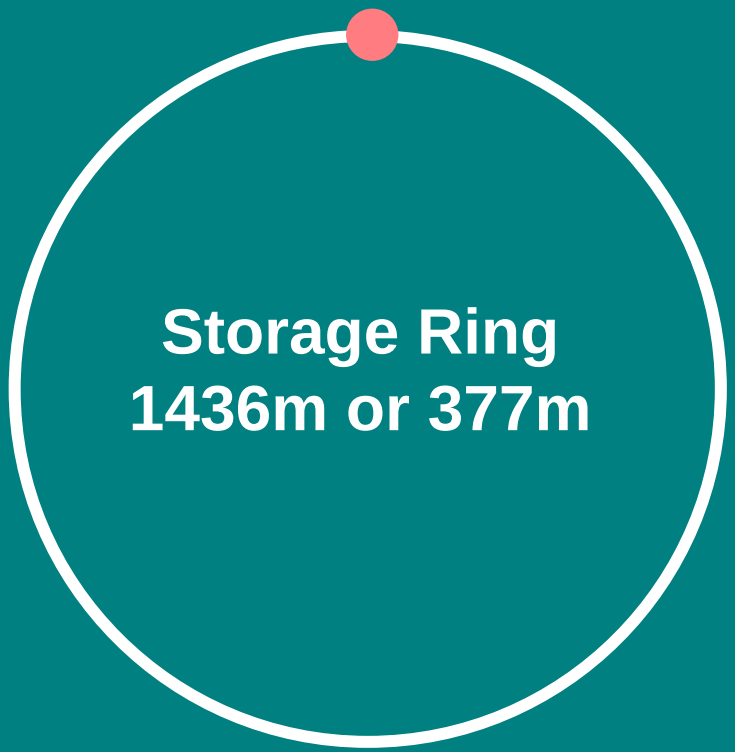
The pulsed nature of SR (1)

- **You have already learned that “the synchrotron radiation is pulsed light source”…**
- **But I stress the importance of this feature for pump-probe method again.**

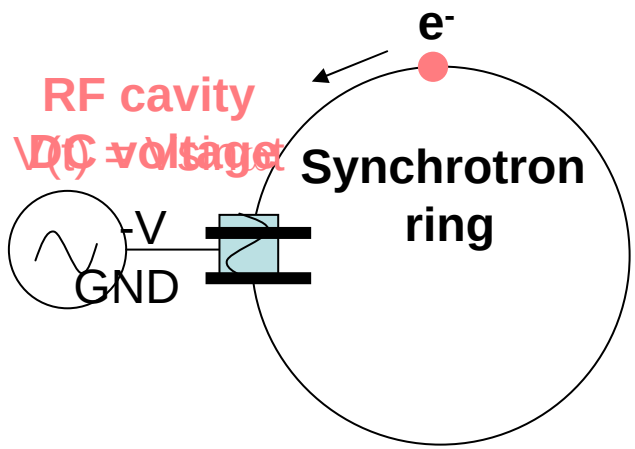
Question (1)



1. The circumference of the SPring-8 is 1436 m. What is the period of 1 turn?
2. What is the frequency of turns (revolution frequency)?
3. The circumference of the PF-AR is 377 m. What is the period and the frequency?

The pulsed nature of SR (2)

Acceleration of electrons with DC or AC voltage



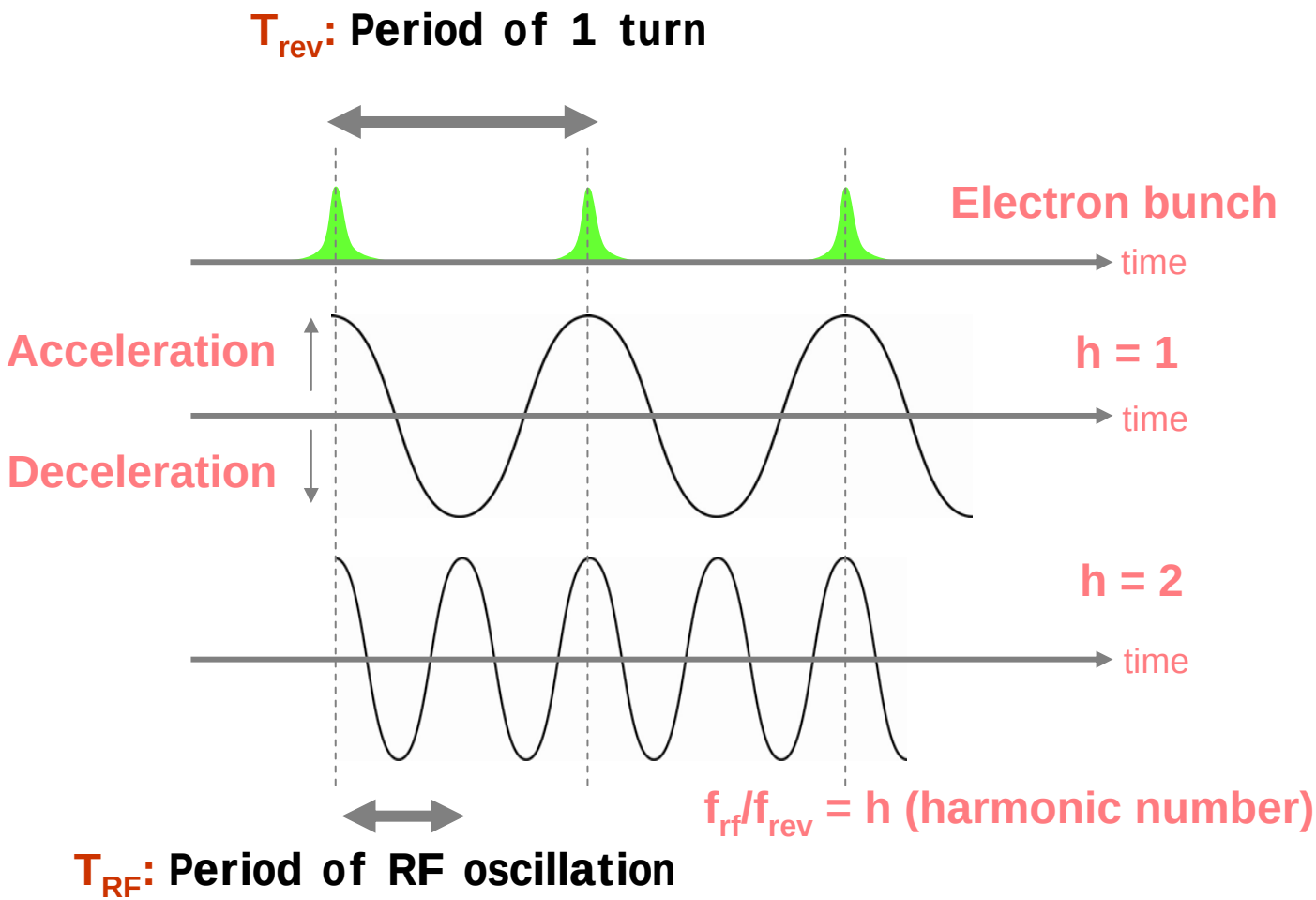
- **Electron emits SR and loses its energy. Continuous acceleration in the storage ring is needed.**
- **DC voltages does not make it.**
- **AC voltage and revolution must be synchronized.**
- **(Period of 1 turn) / (Period of RF) = N :natural numbers**

N: Harmonic Number

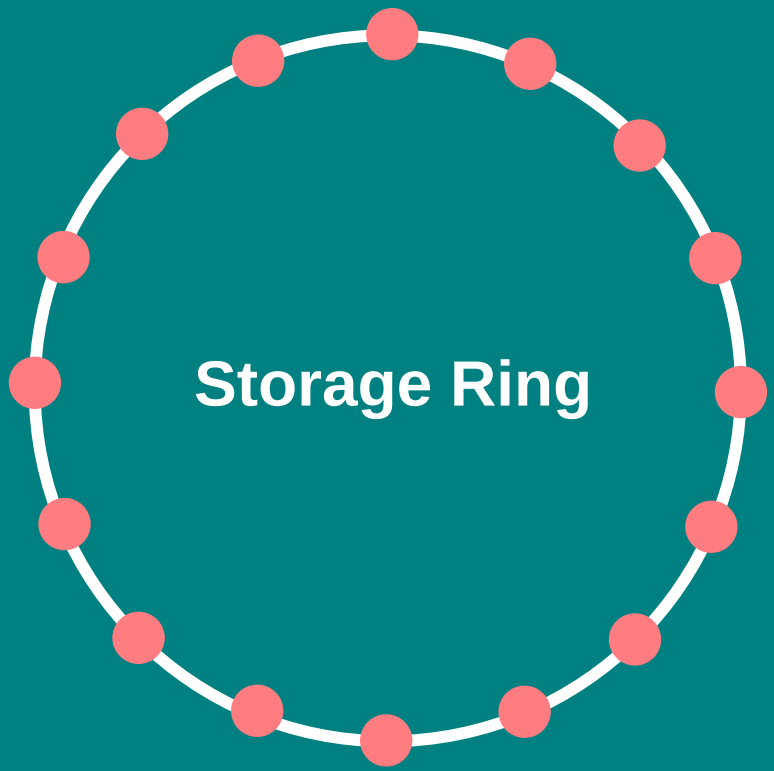
RF: radio-frequency

The pulsed nature of SR (3)

Acceleration of electrons with AC voltage at radio frequency (RF)



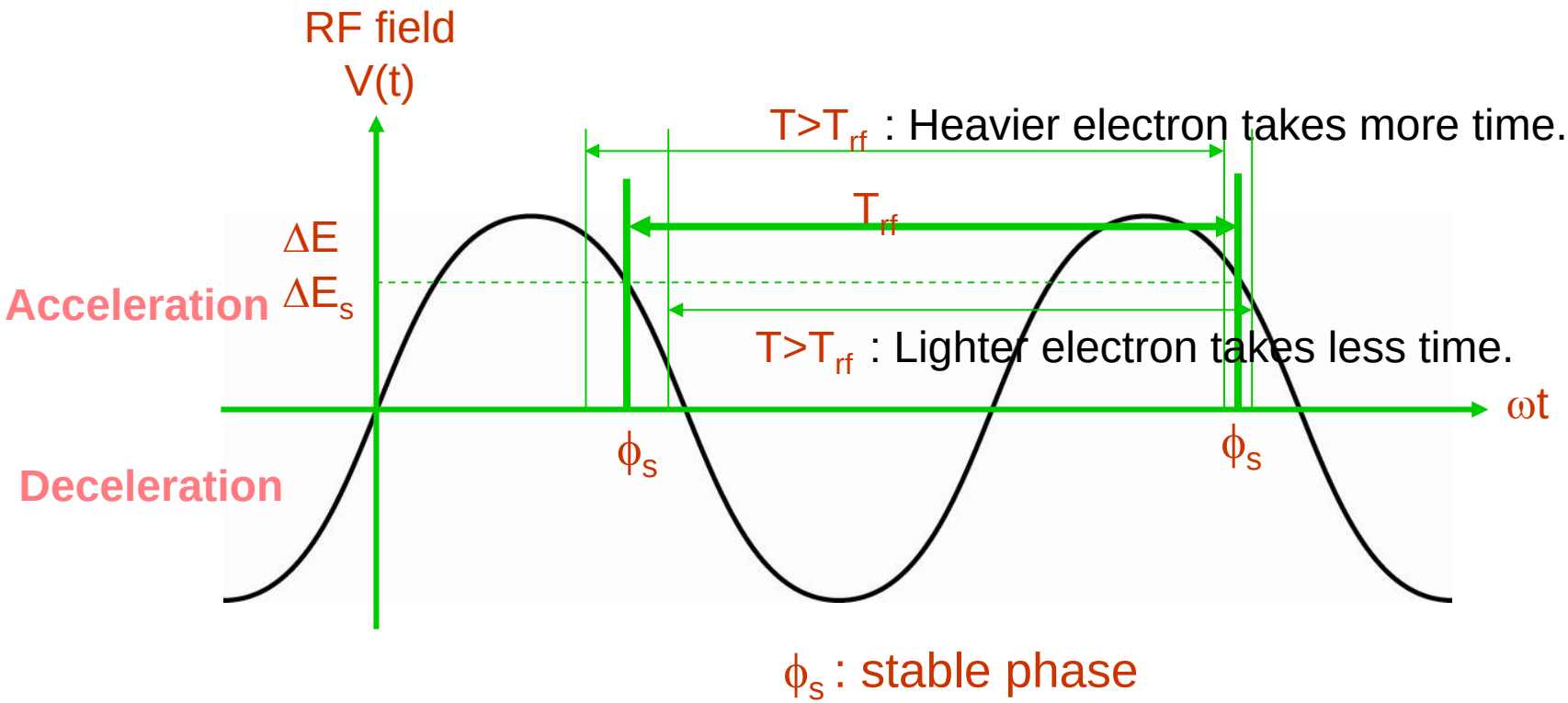
Question (2)



1. The RF frequency of SPring-8 and PF-AR is 508 MHz.
2. What is the harmonic number of SPring-8?
3. What is the harmonic number of PF-AR?

The pulsed nature of SR (4)

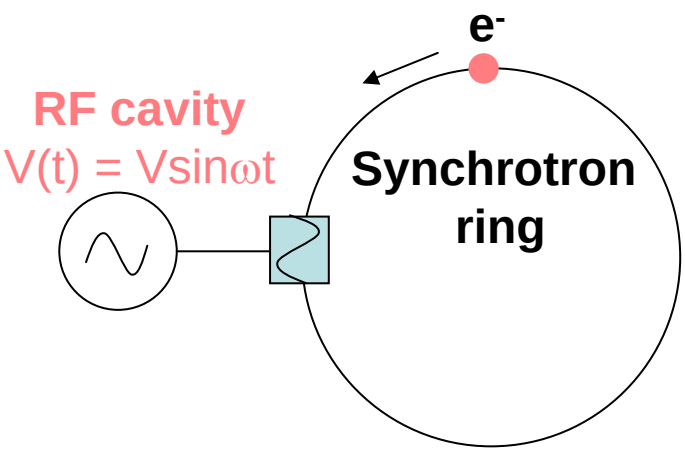
Why are electrons bunched?
(1) stable phase



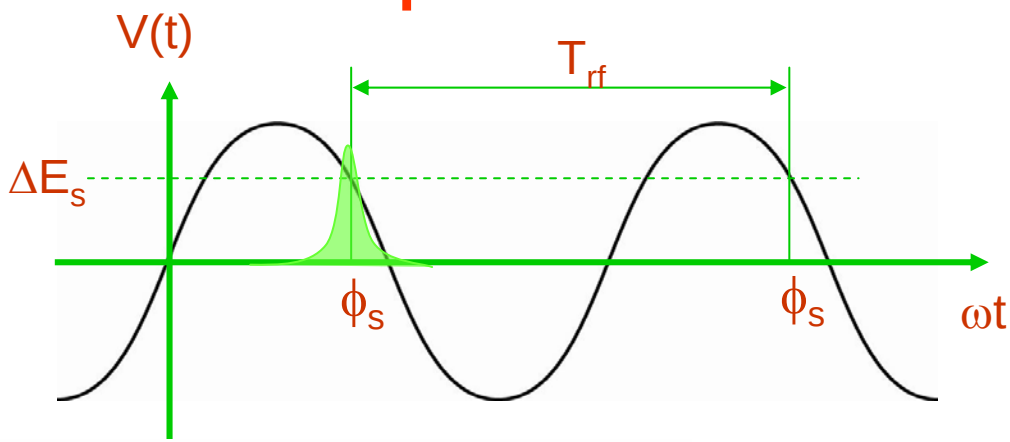
ϕ_s : stable phase

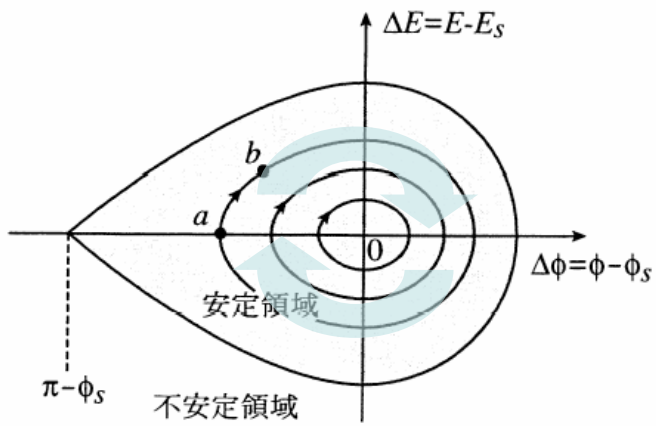
The pulsed nature of SR (5)

Why are electrons bunched?
(2) synchrotron oscillation



Δt ~ 50 picoseconds

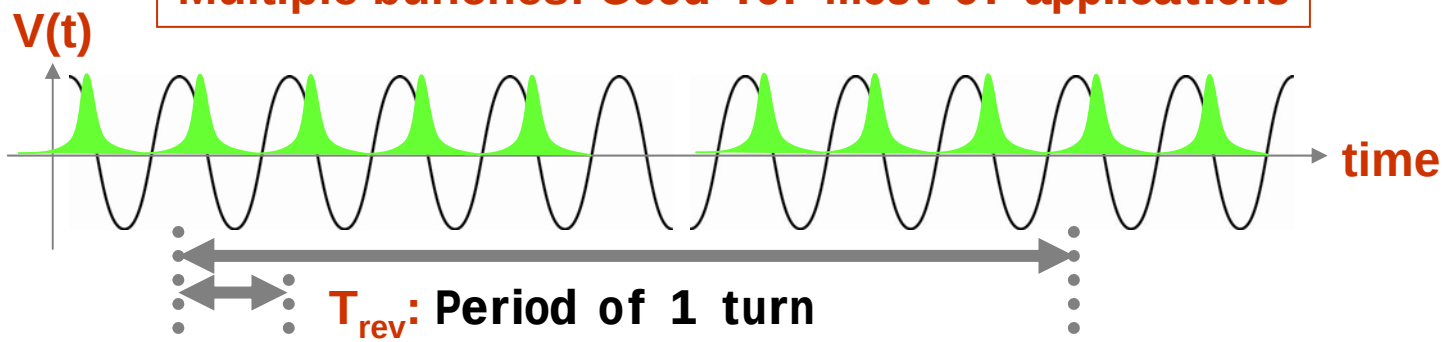




The pulsed nature of SR (6)

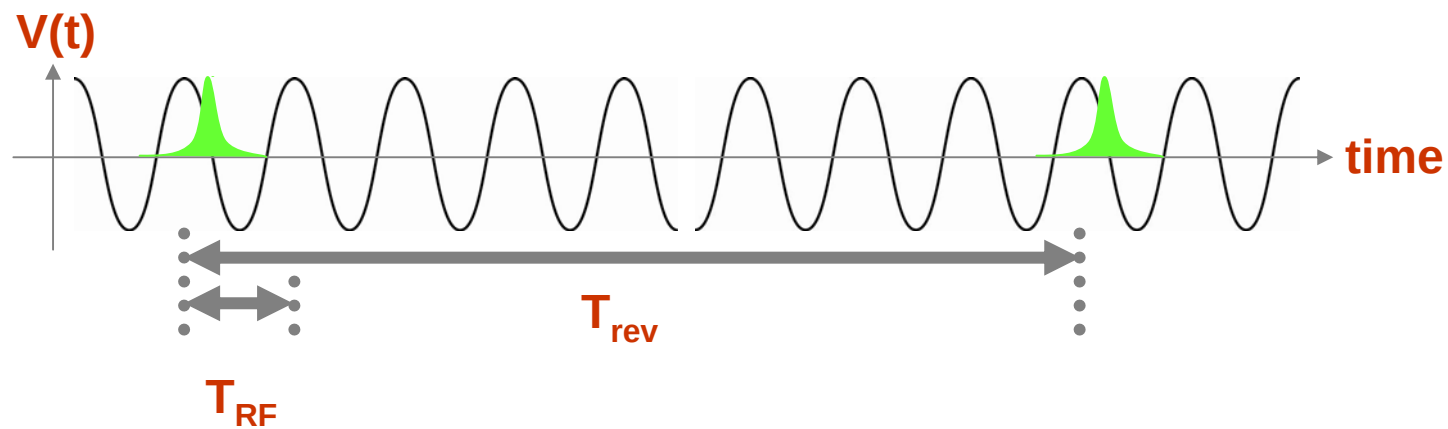
Multiple vs. few bunches mode

Multiple bunches: Good for most of applications



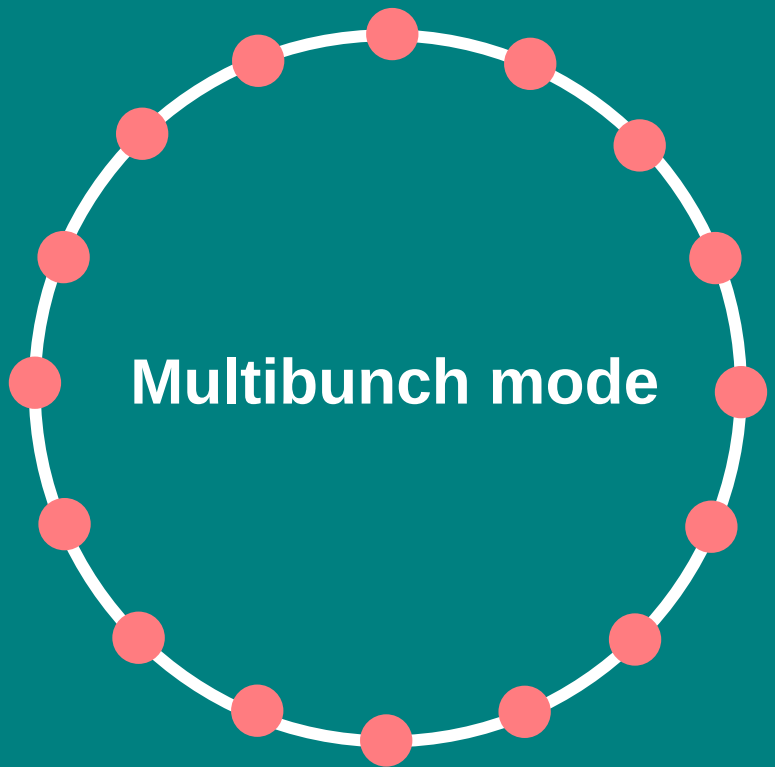
T_{RF}: Period of RF oscillation (1.97 ns @ 508 MHz)

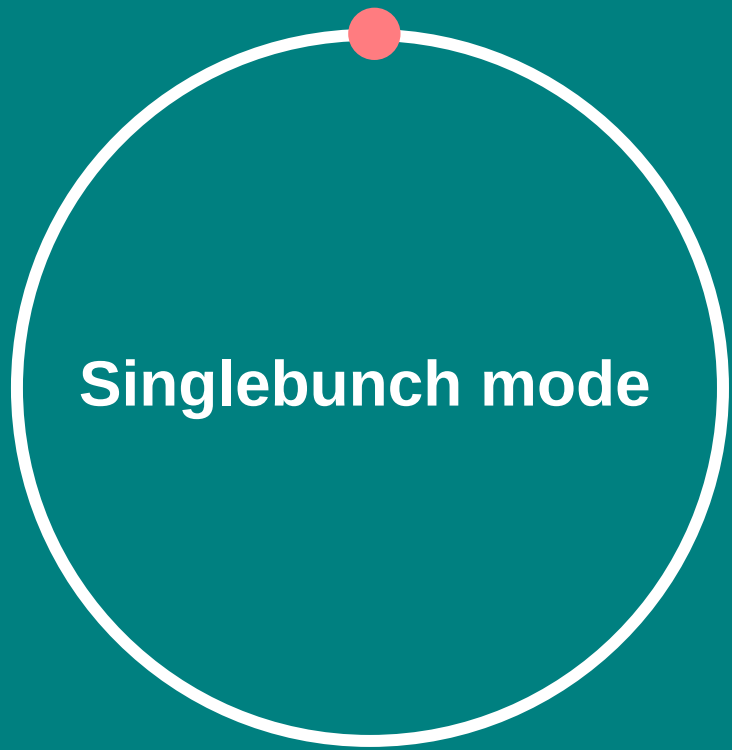
Few bunches: Good for pump-probe method



Questions (3)

1. The ring current of the SPring-8 is 100 mA. What is the electron charge in 1 bunch, if only single RF bucket is used (single bunch mode)?
2. What is the electron charge in 1 bunch, if all the RF buckets are used (multibunch mode)?
3. Assume the ring current of the PF-AR is 100 mA, and calculate the same values.





Bunch modes for pump-probe method at SR facilities

	PF-AR, KEK	ESRF	APS	SPring-8
Ring energy	6.5 GeV	6.0 GeV	7.0 GeV	8.0 GeV
Pump-probe mode/year	~5000 hours (100% single bunch)	~1700 hours (~30% 4-, 16-bunch, hybrid mode)	~800 hours (hybrid mode)	~1000 hours (~20%, D-mode)
Max. current/bunch charge/bunch	60 mA 75.5 nC	16 mA 40 nC	60 / 16 nC	3 mA 16 nC
Bunch duration (RMS)	62 ps	73 ps	65 / 40 ps	20 ps
Beam life time	20 h	6 h (s.b.)	∞ (top-up)	∞ (top-up)
Horizontal emittance	290 nmrad	4 nmrad	3 nmrad	3 nmrad

Summary #2
Pulsed nature of the SR

- **SR is pulsed because electrons are bunched.**
- **Electrons are bunched by acceleration with AC voltage.**
- **Synchrotron oscillation limits the bunch length ~ 50 picoseconds.**
- **Few-bunches mode is highly needed for pump-probe type experiment**

Outline of the talk

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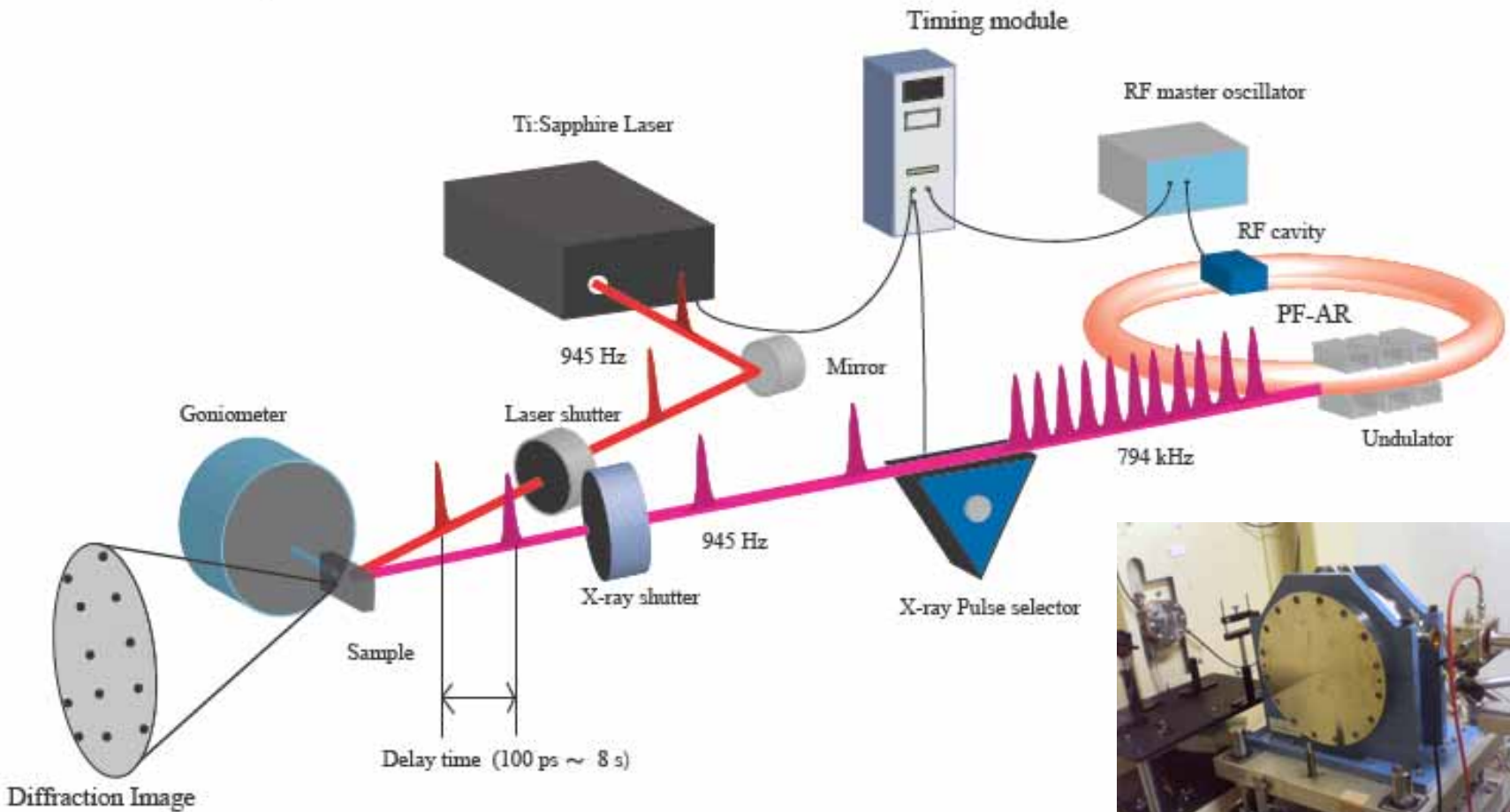
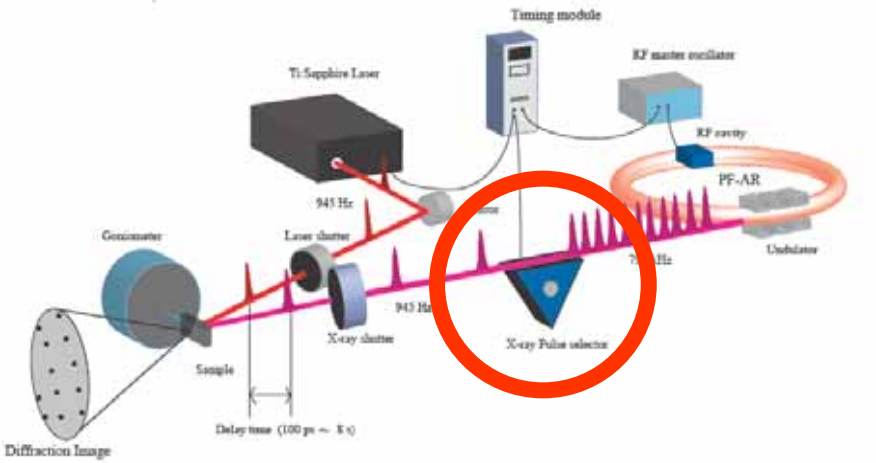
Synchronizing pump and probe pulses (1)

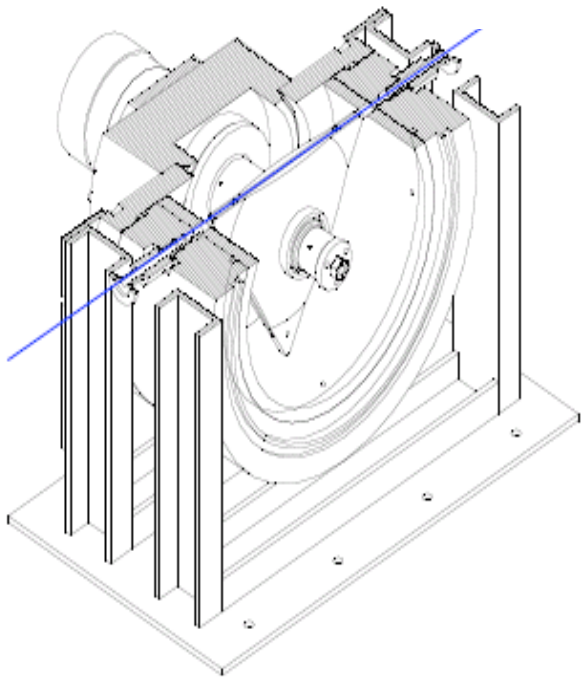
Overview of the typical setup
“pump by laser, probe by SR”



Synchronizing pump and probe pulses (2)

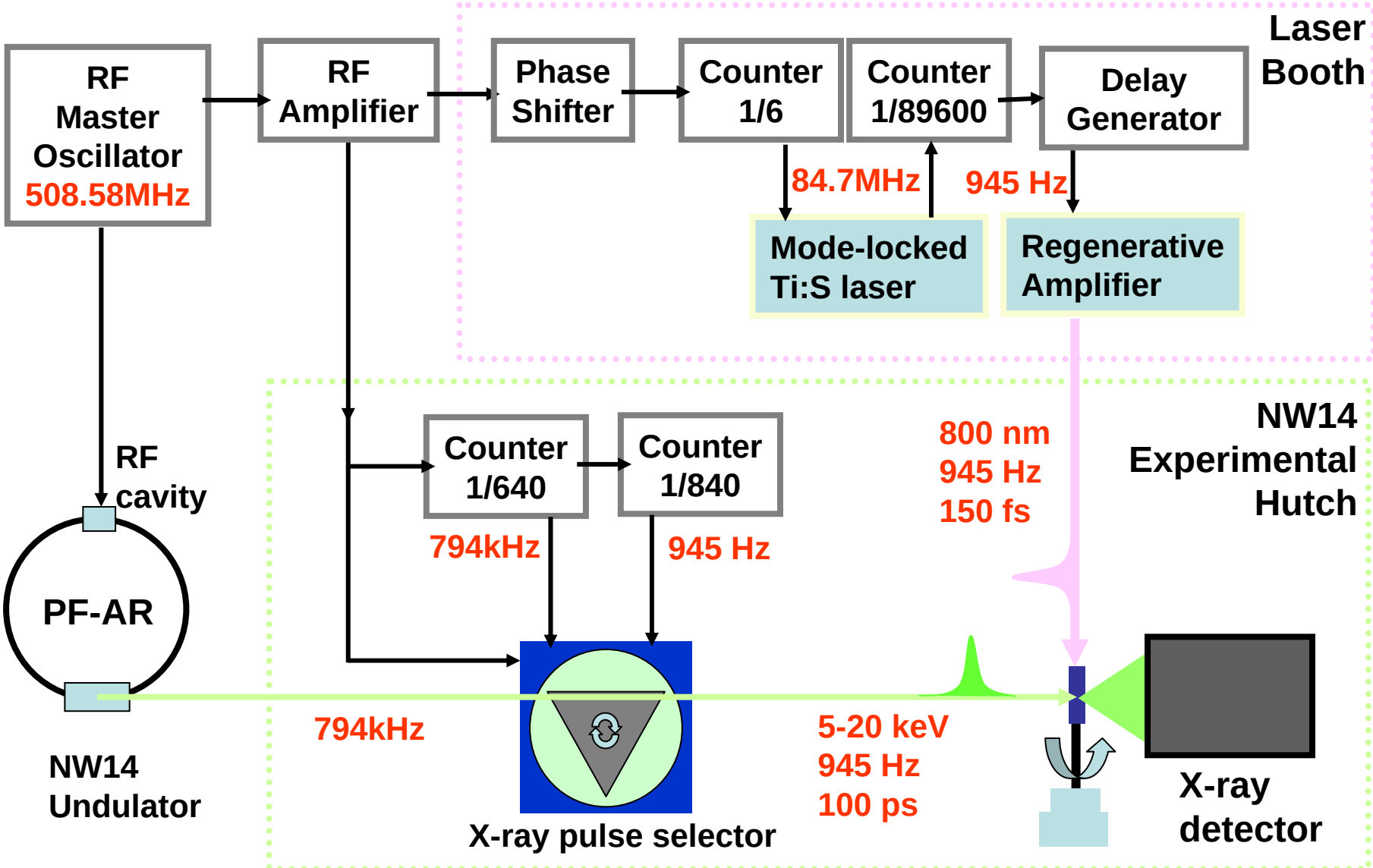
Pulse Selector





Synchronizing pump and probe pulses (3)

Syncing scheme



Synchronizing pump and probe pulses (4)

Timing scheme

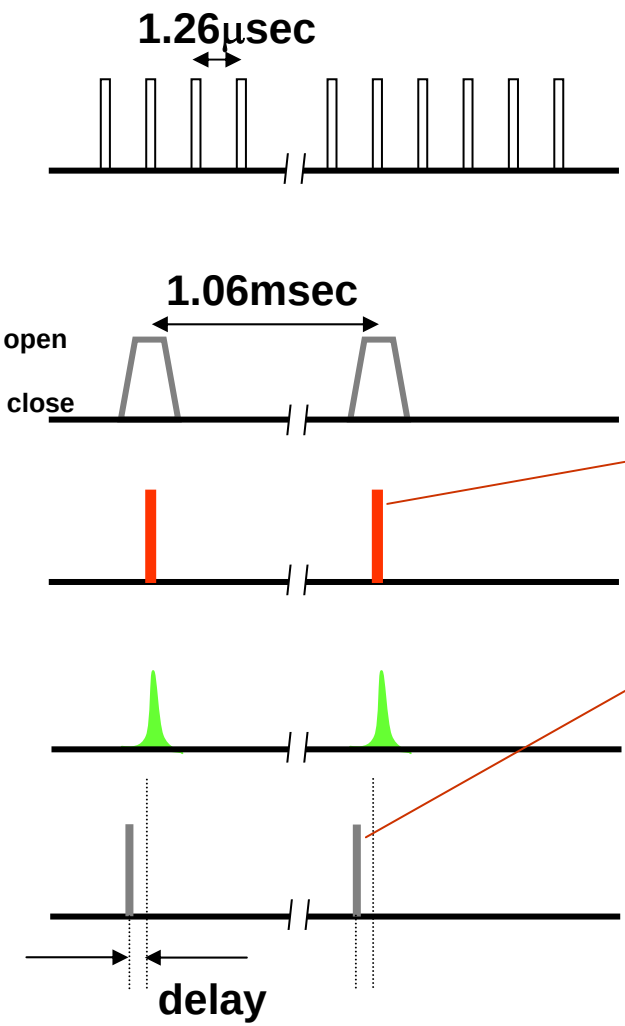
X-ray from PF-AR
(794 kHz = 508 MHz / 640)

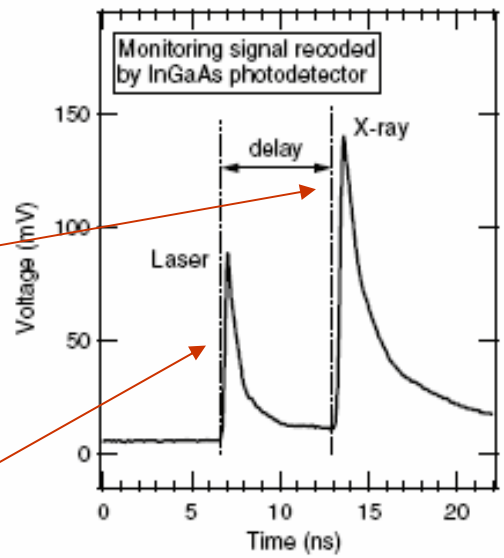
X-ray Pulse Selector
(945 Hz = 794 kHz / 840)

X-ray at Sample
(945 Hz)

Diffraction signal
(945 Hz)

Laser pulse (945 Hz)





Summary #3
Synchronizing pump and probe pulses

- **Synchronization of the pump and the probe pulses is feasible based on the RF frequency.**
- **In order to reduce the repetition rate of the SR (probe) pulses, the pulse selector is very useful.**

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The concept of pump-probe experiment with SR

Synchrotron Radiation：

Powerful tool to explore atomic and electronic structures

×

Pulsed Nature of SR：

Time-resolution down to 100 ps

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Movies of atomic and electronic structures at 100-ps and angstrom resolution will be feasible!!

Some tips of pump-probe experiments

- **Mismatch of the penetration depth between the X-ray and visible light to the sample must be solved.**
  - **X-ray penetrates deeper.**
  - **Sample must be optically thin.**
- **Repetition frequency of the pump-probe experiment is limited by sample.**
  - **Pump-probe experiment is photon demanding.**

Examples

Type of experiments	sample	Typical repetition rate
Single-crystal diffraction	Charge transfer complex crystal	1kHz
	Molecular excited state	10Hz
	Transition metal oxides	1kHz
	Protein crystal	1 Hz
Liquid scattering	Organic & inorganic solution	1kHz
	Protein solution	1kHz
XAFS	Transition metal complex	1kHz
Shock wave	Inorganic crystal	single shot
Grazing incidence diffraction	Organic thin film	1kHz

Summary #4
Applications

- **Pump-probe method can be applied to most of SR experiments.**
  - **Diffraction**
  - **XAFS**
  - **Solution Scattering**
  - **etc.**
- **Pump-probe method adds another dimension (time) to your research.**

Concluding remarks

- **Pump-probe method with synchrotron radiation enables us to make movies at atomic resolution.**
- **The time resolution is currently ~ 100 picosecond (sub-nanosecond) order, but soon will be ~ 100 femtosecond (sub-picosecond) order.**
- **This will be fun!**

Femtosecond X-ray Sources

The X-FEL under construction

http://www-xfel.spring8.or.jp/index.htm