Toward Fourier-limited X-ray Science

Photon Factory, KEK
& PREST, JST
Shin-ichi Adachi
outline

• Time-domain X-ray science
  – with Storage Ring (Photon Factory Advanced ring, KEK)

• Current status of Energy Recovery Linac (ERL) project at KEK
  – 35-245MeV ERL test facility (under construction)
  – 3.5GeV ERL + XFEL Oscillator (not approved)

• Towards Fourier-limited X-ray Science with XFEL-O and seeded XFEL
  – Inelastic X-ray scattering
  – Nonlinear X-ray Optics
  – Two-photon correlation spectroscopy
  – Transient grating

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• Summary
<table>
<thead>
<tr>
<th><strong>TR x-ray applications at KEK</strong></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Picosecond photoresponse of perovskite manganite (NSMO) thin film ($\tau \sim 50\text{ps} \sim 2\text{ns}$)</td>
<td>Photo-induced spin-crossover transition of metal complex in solution (TR-XAFS: $\tau \sim 700\text{ps}$)</td>
</tr>
<tr>
<td><img src="image1.png" alt="NSMO thin film" /></td>
<td><img src="image2.png" alt="Metal complex" /></td>
</tr>
<tr>
<td>Photochemical reaction in liquid (TR-liquidography: $\tau \sim 100\text{ps}\sim 1\mu\text{s}$)</td>
<td>Ligand migration dynamics in protein crystal ($\tau \sim 800\text{ min}$)</td>
</tr>
<tr>
<td><img src="image3.png" alt="Liquid reaction" /></td>
<td><img src="image4.png" alt="Protein crystal" /></td>
</tr>
<tr>
<td>Laser shock-induced lattice deformation of CdS single crystal (TR single-shot Laue diffraction: $\tau \sim 1\text{ns}\sim 10\text{ns}$)</td>
<td></td>
</tr>
<tr>
<td><img src="image5.png" alt="CdS crystal" /></td>
<td></td>
</tr>
</tbody>
</table>
PF-AR (6.5GeV) Full-Time Single-Bunch Operation
~200 days/year

- multilayer mirror
- Si(111) monochromator
- undulator
- femtosecond laser system
- nanosecond laser system
- Jülich x-ray chopper

PF-AR (6.5GeV)
#1 TR-Diffraction

Picosecond photoresponse of perovskite manganite (NSMO) thin film

1 kHz rep rate
with mono X-ray ($\Delta E/E \sim 0.01\%$)
$\sim 10^9$ photons/sec
**Phase Transition in Manganite Thin Film**

\[ \text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3/\text{SrTiO}_3(011) \]  
\[ \text{(NSMO/STO(011))} \]

- **Mn\textsuperscript{3+}: (3d\textsuperscript{4})**
- \( \text{e}_g: x^2 - y^2 \)
- \( \text{t}_{2g}: 3z^2 - r^2 \)
- **Thickness**: 80 nm

\[ \text{(Nd, Sr)} \]
\[ \text{MnO}_6 \]

**Nakamura et al.**  
*APL 86* 182504 (2005)

**Ichikawa et al.**  

**CE-type Antiferromagnetic insulator (CE-AF)**

**Ferrometal (FM)**
Optical pump-probe results
Collaboration with K. Miyano Group (Univ. of Tokyo)

TR-Reflectivity (100K)

Kerr Rotation

Ichikawa et al.
Nature Materials, 10, 101-105 (2011)

Miyasaka et al. PRB 74 012401 (2006)
Temperature dependence of X-ray Diffraction

Nd$_{0.5}$Sr$_{0.5}$MnO$_3$/SrTiO$_3$(011)

CCD Image

Intensity (arb.)

Temperature (K)

Lattice parameter (Å)

Intensity (arb.)

Q (Å$^{-1}$)

Heating

CE-AFI
(charge/orbital order)

FM cluster
(charge/orbital disorder)
Layout of the laser-pump X-ray-probe experiment

Pump Laser
800 nm (1.55 eV)
FWHM: 150 fs

X-ray
Energy: 15 keV
FWHM: 100 ps

Sample
NSMO/STO(011) thin film
Film thickness: 80 nm

Detector
(CCD, Scintillation counter)

Ti:Sapphire
Time dependence of (004) reflection

Nd$_{0.5}$Sr$_{0.5}$MnO$_3$/SrTiO$_3$(011)

Difference Image
black : +  white : -

Delay Time = -200 ps

$T = 100$ K
Pump: 0.8mJ/cm$^2$
Time dependence of the (004) and (1/4 9/4 0) reflections

$\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3/\text{SrTiO}_3(011)$

$T = 100 \text{ K}$

Pump: 0.8mJ/cm$^2$
Photo-induced “hidden” state?

Temperature Dependence
(Without laser irradiation)

Pump Power Dependence
$T = 100 \text{ K}$
(Delay-time: +150 ps)

Ichikawa et al.
“Transient photoinduced ‘hidden’ phase in a manganite”
#2. TR-XAFS

Photo-induced spin-crossover transition of metal complex in solution

1 kHz rep rate
with mono X-ray ($\Delta E/E \sim 0.01\%$)

$10^9$ photons/sec
photo-induced spin-state transition by TR-XAFS

Fe²⁺(phen)₃

Open Jet system

Laser pulses 945 Hz

Fluorescence X-ray

X-ray pulses @ 794 kHz (Energy-scanned)


Shunsuke Nozawa (KEK)

Tokushi Sato (KEK)
picosecond time-resolved spin-crossover transition of $\text{Fe}^\text{II}(\text{phen})_3^-$

Fe$^{2+}$(phen)$_3$ cluster

$3d^6$ 

$\text{FeN}_6$ cluster

$e_g$ 

MLCT, LF

$1\text{MLCT}$ 

$\text{MLCT}, \text{LF}$ 

$1\text{A}_1$ 

$^{5}T_2$ 

$S=0,^{1}A_1$ 

$S=2,^{5}T_2$ 

Fe-N bond length

$R_{\text{Fe-N}}^{\text{LS}}$ 

$< R_{\text{Fe-N}}^{\text{HS}}$ 

Fe-N bond length

$\sim 700 \text{ps}$ 

$400 \text{nm}$ 

Energy
TR-XAFS: Experimental Setup
TR-Near Edge Structure

Fe K-edge XAFS
Fe(phen)$_3$
Aqueous Solution

Intensity (arb. units)

Photon Energy (eV)

Time Course
Feature B

$I = I_0 \exp\left[-(\Delta t/\tau)\right]$

$\tau = 691 \pm 30$ ps

X-ray
$\sigma = 60$ ps

Time Course
Feature D

Delay (ps)

Intensity (arb. units)

FWHM 142 ps

Low Spin

Transient difference at +50 ps

difference x 15
TR-XANES features in pre-edge region

Fe(phen)$_3$ : Low Spin
Fe(2-CH$_3$phen)$_3$ : High Spin
HS-LS

Transient difference

Intensity (arb. units)
Photon Energy (eV)

Fe K-pre-edge

difference $\times 2$

$\tau = 691$ ps
**excited state EXAFS**

EXAFS analysis summary

<table>
<thead>
<tr>
<th>Spectrum</th>
<th>$R_{Fe-N}$ (Å)</th>
<th>$\sigma^2$ (Å$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LS</td>
<td>1.98(1)</td>
<td>0.001(1)</td>
</tr>
<tr>
<td>Photo-excited HS</td>
<td>2.15(2)</td>
<td>0.011(3)</td>
</tr>
</tbody>
</table>
photoinduced structural change: a molecular movie!

Low Spin State

Photo-excited High Spin State

• TR-XAFS provides spin, electronic and structural information of photo-induced states, which enables to produce molecular movies.
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• Summary
Evolution of the synchrotron sources

Case 1: 3rd gen. synchrotron sources

Case 2: ERL & SASE-XFEL (Diffraction limit)

Case 3: XFELO & seeded XFEL (Fourier limit)
Diffraction Limit

$$\sigma_x \sigma_{x'} \geq \frac{\lambda}{4\pi}$$
Fourier Limit

$\Delta \omega \Delta t \geq \frac{\hbar}{4\pi}$
Fourier-limited X-ray
KEK Energy Recovery Linac (ERL) project

Linac based light source:
1) Diffraction-limited beam $\varepsilon \sim 15\text{pmrad} \sim \lambda/4\pi$
2) Short pulse capability 0.1~1 pico-second
3) High repetition rate 1.3 GHz
35-245MeV ERL test facility (Compact ERL)
- Plan and Status -
Compact ERL for developing and demonstrating ERL technologies

Parameters of the Compact ERL

<table>
<thead>
<tr>
<th>Parameters</th>
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</tr>
</thead>
<tbody>
<tr>
<td>Beam energy</td>
<td>35 - 245 MeV</td>
</tr>
<tr>
<td>Injection energy</td>
<td>5 MeV</td>
</tr>
<tr>
<td>Average current</td>
<td>10 - 100 mA</td>
</tr>
<tr>
<td>Acc. gradient (main linac)</td>
<td>15 MV/m</td>
</tr>
<tr>
<td>Normalized emittance</td>
<td>0.1 - 1 mm·mrad</td>
</tr>
<tr>
<td>Bunch length (rms)</td>
<td>1 - 3 ps (usual)</td>
</tr>
<tr>
<td></td>
<td>~ 100 fs (with B.C.)</td>
</tr>
<tr>
<td>RF frequency</td>
<td>1.3 GHz</td>
</tr>
</tbody>
</table>
Recent View in the ERL Development Hall (EDH)

Clean Room for SCC Assembly

Liquid Helium Refrigerator
3.5GeV ERL (1\textsuperscript{st} phase) 
+ XFELO (2\textsuperscript{nd} phase)
3.5GeV ERL Plan at KEK

Parameters of the ERL

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beam energy</td>
<td>3.5 GeV</td>
</tr>
<tr>
<td>Average current</td>
<td>10 - 100 mA</td>
</tr>
<tr>
<td>Normalized emittance</td>
<td>0.1 – 1.0 mm·mrad</td>
</tr>
<tr>
<td>Energy spread (rms)</td>
<td>(0.5 - 2) ×10^{-4}</td>
</tr>
<tr>
<td>Bunch length (rms)</td>
<td>1 - 3 ps (usual mode)</td>
</tr>
<tr>
<td></td>
<td>~ 100 fs (bunch compression)</td>
</tr>
<tr>
<td>RF frequency</td>
<td>1.3 GHz</td>
</tr>
</tbody>
</table>

Parameters of the light sources

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spectral range</td>
<td>30 eV - 30 keV</td>
</tr>
<tr>
<td>Average brilliance from insertion devices</td>
<td>10^{21} - 10^{23} ph/s/mm^{2}/mrad^{2}/0.1%bw</td>
</tr>
<tr>
<td>Average flux</td>
<td>&gt; 10^{16} ph/s/0.1%bw</td>
</tr>
<tr>
<td>Number of ID’s</td>
<td>20 - 30</td>
</tr>
</tbody>
</table>
ERL undulator spectra
(with 15pmrad ~ 1.5nmrad natural emittance)

E=3.5GeV
I=100mA
βx=βy=5m
K=1.0
σ_E/E=4e-5
L=5m
λu=16mm
$1^{st}$ harmonic of the undulator (@4024 eV) (with 15pmrad $\sim$ 1.5nmrad natural emittance)

Undulator radiation

$$I(\Delta \omega) \propto \left( \frac{\sin \left( \frac{\pi N \Delta \omega}{\omega} \right)}{\pi N \frac{\Delta \omega}{\omega}} \right)^2$$

E=3.5GeV
I=100mA
$\beta x=\beta y=5\text{m}$
K=1.0
$\sigma_E/E=4e^{-5}$
L=5m
$\lambda u=16\text{mm}$
1st harmonic of the undulator (linear scale)

$\Delta E/E = 12 \text{ eV} / 4024 \text{ eV} = 0.0030$

$\Delta \omega/\omega = 1/N$

$N = 5000/16 = 313$

$1/N = 16/5000 = 0.0032$
Hard X-Ray FEL Oscillator

- Store an X-ray pulse in a Bragg cavity → multi-pass gain & spectral cleaning
- Provide meV bandwidth ($\Delta \omega / \omega \sim 10^{-7}$)
- MHz pulse repetition rate → high average brightness

Originally proposed in 1984 by Collela and Luccio and resurrected in 2008 (KJK, S. Reiche, Y. Shvyd’ko, PRL 100, 244802 (2008))

Courtesy of K.-J. Kim
Low emittance and high rep rate of ERL matches the specs of XFEL Oscillator (XFELO)

K.-J. Kim et al. PRL (2008) 100, 244802

- **Electron beam:**
  - Energy 7 GeV
  - Bunch charge ~ 25 - 50 pC
  - Bunch length (rms) 0.1 - 1.0 ps
  - Normalized rms emittance < 0.2 - 0.3 mm-mr
  - Energy spread (rms) ~ 2x10^{-4}
  - Constant bunch rep rate @ ~1 MHz

- **Undulator:**
  - \( L_u = 20 - 60 \) m, \( \lambda_u = 2.0 \) cm, \( K = 1.0 - 1.5 \)

- **Optical cavity:**
  - 2- or 4- diamond crystals and focusing mirrors
  - Total round trip reflectivity > 50 - 85 %

- **XFELO output:**
  - 5 keV - 25 keV
  - Bandwidth: \( \Delta \omega / \omega \sim 1 \times 10^{-7} \), pulse length (rms) = 80- 500 fs
  - # photons/pulse ~ 1x10^9
Energy Recovery Linac (ERL) and XFEL Oscillator

3.5 GeV main SC linac

7 GeV XFEL-O
Diamond backscattering: High reflectivity and narrow bandwidth

Reflectivity vs. Photon energy (keV)

C @ 300 K

E_e = 12.0404 keV

C(444); L = 0.2 mm; T = 300 K

C(444); L = 0.042 mm; T = 300 K

Courtesy of Yuri Shvyd'ko
Seeded XFEL & XFEO

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Fourier-limited X-ray

\[ \Delta E \]

\[ \Delta t \]

\[
\begin{array}{cccc}
\mu eV & meV & eV & keV \\
ns & ps & fs & as
\end{array}
\]

Seeded XFEL?
1. inelastic X-ray scattering

Current High Resolution IXS @ APS, ESRF, SPring-8

10^9-10^{10} photons/sec, ΔE≈1meV

XFEL O X-ray beam characteristics

1x10^9 photons/pulse @ 1MHz
1x10^{15} photons/sec
Δω/ω ≈ 10^{-7}

Ideal for IXS, NRS, HXPES, etc...

APS XFELO Workshop 2010
Presentation by Clement Burns
(Western Michigan Univ.)
2. Nonlinear X-ray Optics

- Quantitative and systematic studies will require Fourier-limited X-rays
- Sum- & difference-frequency mixing applications
- Parametric down-conversion
  - $X$ (pump) $\rightarrow$ $X'$ (signal) + EUV or SX (idler)
3. two-photon correlation spectroscopy in radio wave domain

Radio wave domain
Multi-dimensional NMR

Correlation of nuclear magnetic spin
two-photon correlation spectroscopy (2)
in IR domain

Multi-dimensional IR spectroscopy (photon echo)

Correlation of vibrational modes
two-photon correlation spectroscopy \((3)\)
in X-ray domain

<table>
<thead>
<tr>
<th>Frequency</th>
<th>MHz ((10^6\text{Hz}))</th>
<th>GHz ((10^9\text{Hz}))</th>
<th>THz ((10^{12}\text{Hz}))</th>
<th>PHZ ((10^{15}\text{Hz}))</th>
<th>EHz ((10^{18}\text{Hz}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>0.3km ((10^3\text{m}))</td>
<td>0.3m ((10^0\text{m}))</td>
<td>0.3mm ((10^{-3}\text{m}))</td>
<td>0.3(\mu)m ((10^{-6}\text{m}))</td>
<td>0.3nm ((10^{-9}\text{m}))</td>
</tr>
</tbody>
</table>

X-ray region

Multi-dimensional X-ray spectroscopy

Correlation of electronic states, Wave packet motion?
4. Transient grating

Transient X-ray standing wave without perfect crystal

Proposed by Keith Nelson
Summary

• SC Linac-based light source enables electron beam with high rep rate and low emittance suitable for Fourier-limited X-ray sources.
• Fourier-limited X-ray may open new X-ray applications in inelastic X-ray scattering, nonlinear X-ray optics, two-photon correlation spectroscopy and transient grating.
members @ Beam Line NW14A, KEK

<table>
<thead>
<tr>
<th>Name</th>
<th>Institution</th>
<th>Name</th>
<th>Institution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shunsuke Nozawa</td>
<td>KEK</td>
<td>Tokushi Sato</td>
<td>KEK</td>
</tr>
<tr>
<td>Manabu Hoshino</td>
<td>TITECH</td>
<td>Ayana Tomita</td>
<td>KEK</td>
</tr>
<tr>
<td>Matthieu Chollet</td>
<td>→APS</td>
<td>Laurent Guérin</td>
<td>→Univ. Rennes 1</td>
</tr>
<tr>
<td>Hirohiko Ichikawa</td>
<td>JST</td>
<td>Shin-ya Koshihara</td>
<td>TITECH</td>
</tr>
</tbody>
</table>
Recovery from the earthquake...

Damages at KEK
March 11, 2011

Photon Factory now operating!
Top-up mode
June 20, 2011
Thank you for your attention