Nuclear Resonance Spectroscopy: Opportunities with New Radiation Sources

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Argonne National Laboratory

Symposium

50 Years After - the Mössbauer Effect Today and in the Future

Technische Universität München, Physics Department,
Garching (Germany), 9-10 October, 2008

Acknowledgements: Some of the early pioneers at Argonne and to a few hundred collaborator and friends
DOE/BES for Funding the Research

Outline

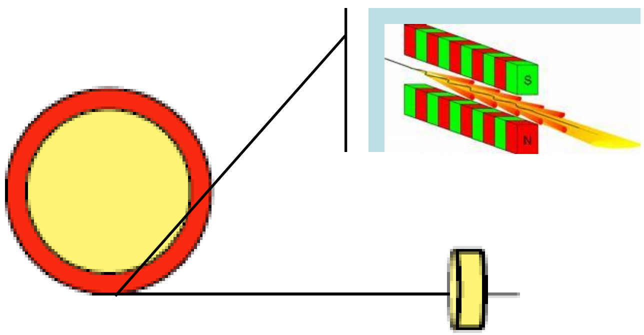
- *Recall Third Generation Sources*
- *Introduction to Next Generation Sources*
- *Opportunities in Materials Physics*
- *Dreams and Speculations*

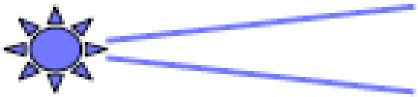
Why Synchrotron Radiation Sources?

Nuclear resonance and brightness of synchrotron radiation

Undulator based SR

10 mCi ^{57}Co source

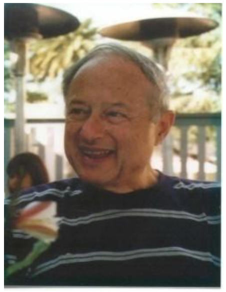




10^{23} photons / sec / eV / sr

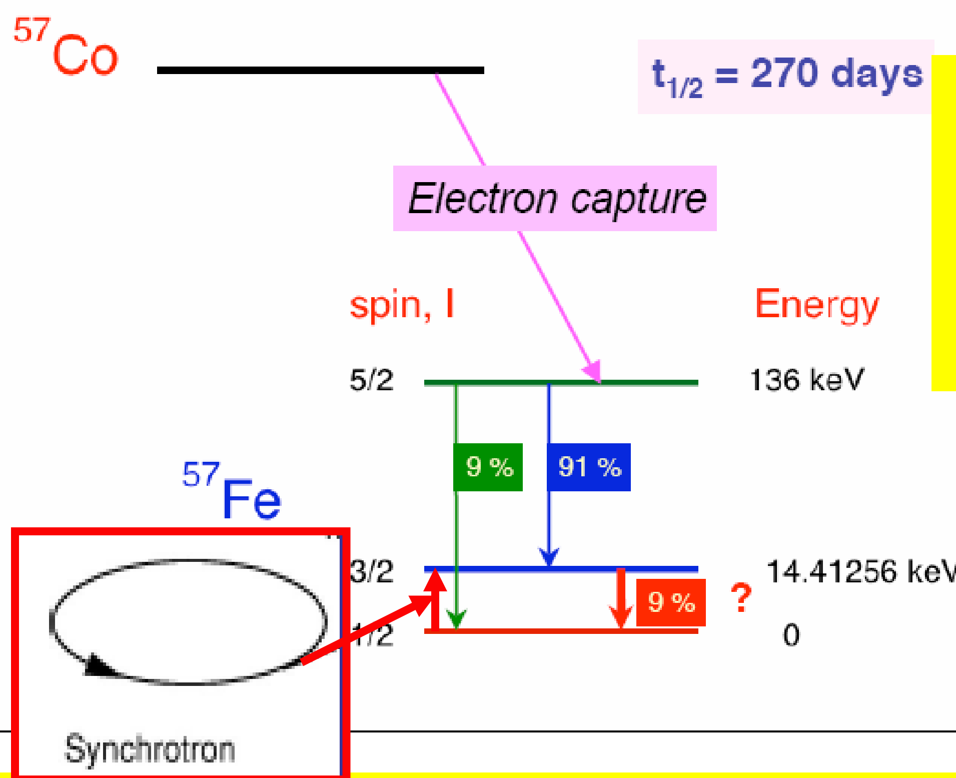
10^{11} photons /sec / eV / sr

Characteristics of nuclear excitation and decay



Stan Ruby,
J. Phys. 35 (1974)

Eric Gerdau et al.
PRL 54 (1985)



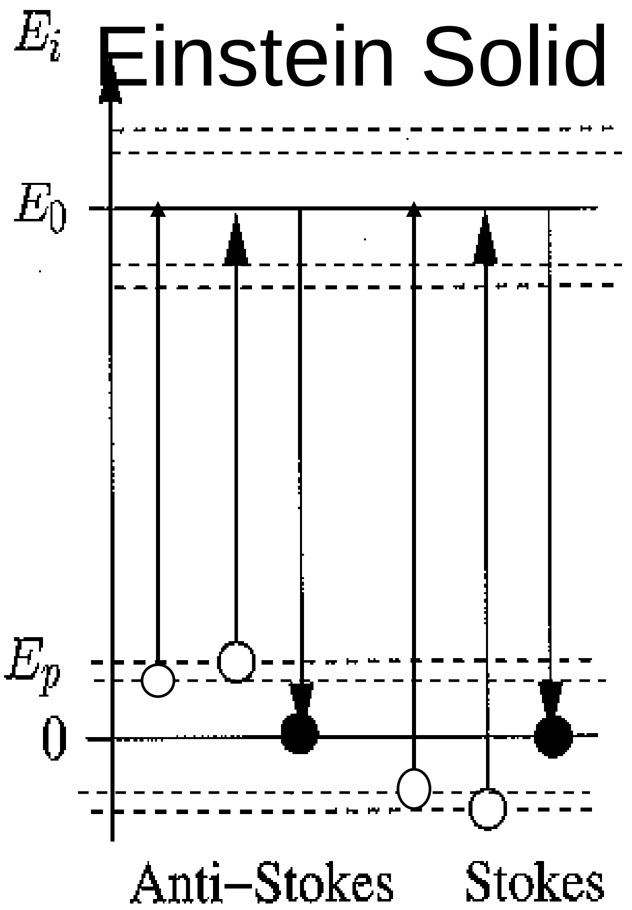
Seppi, E.J., Boehm, F.: Nuclear resonance excitation using a diffraction monochromator. Phys.Rev. 128, 2334–2339 (1962)

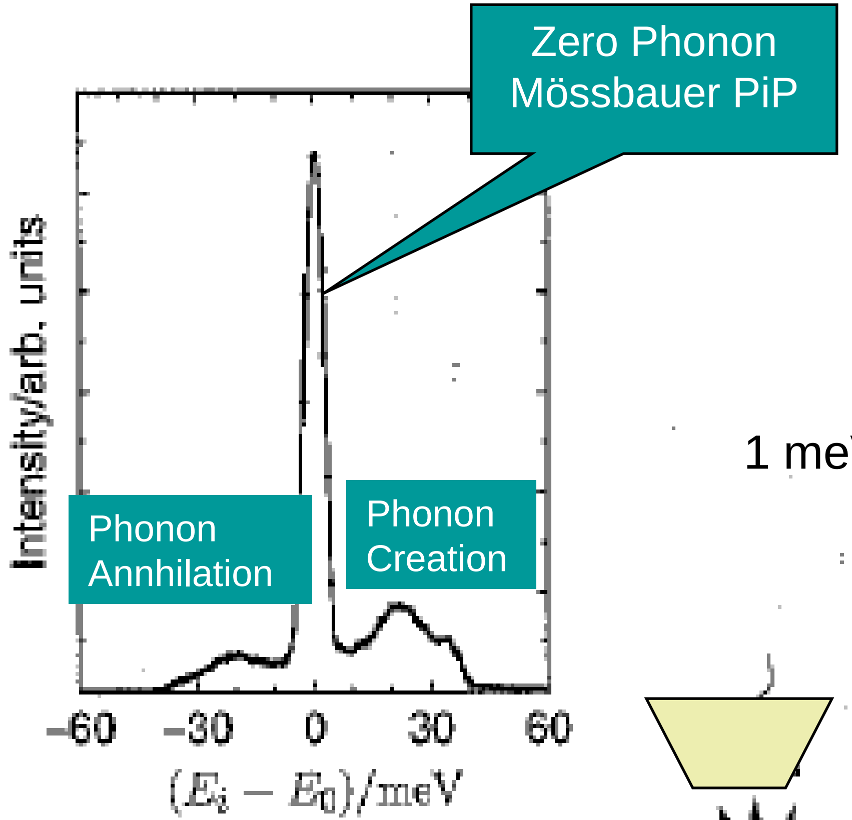
Z	Isotope	E_o (keV)	τ_o (ns)	Γ_o (neV)	σ_o (kbarn)	σ_o / σ_{el}
26	^{57}Fe	14.4125	141.1	4.7	2464	426

Incoherent Nuclear Resonant Inelastic X-ray Scattering (NRIXS)

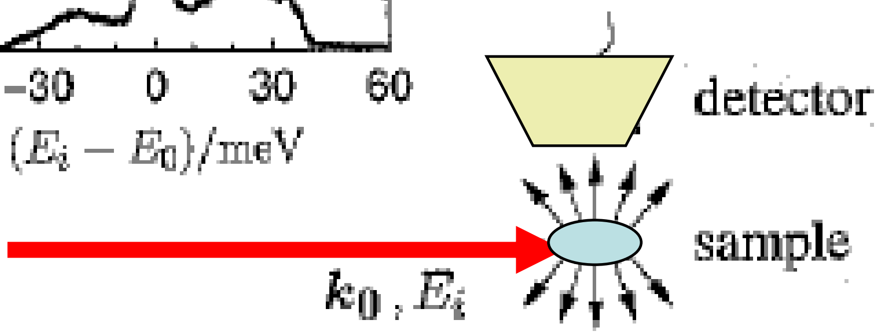
Seto et al. PRL 74, 3828, 1995
Sturhahn et al. PRL 74, 3832, 1995

Einstein Solid

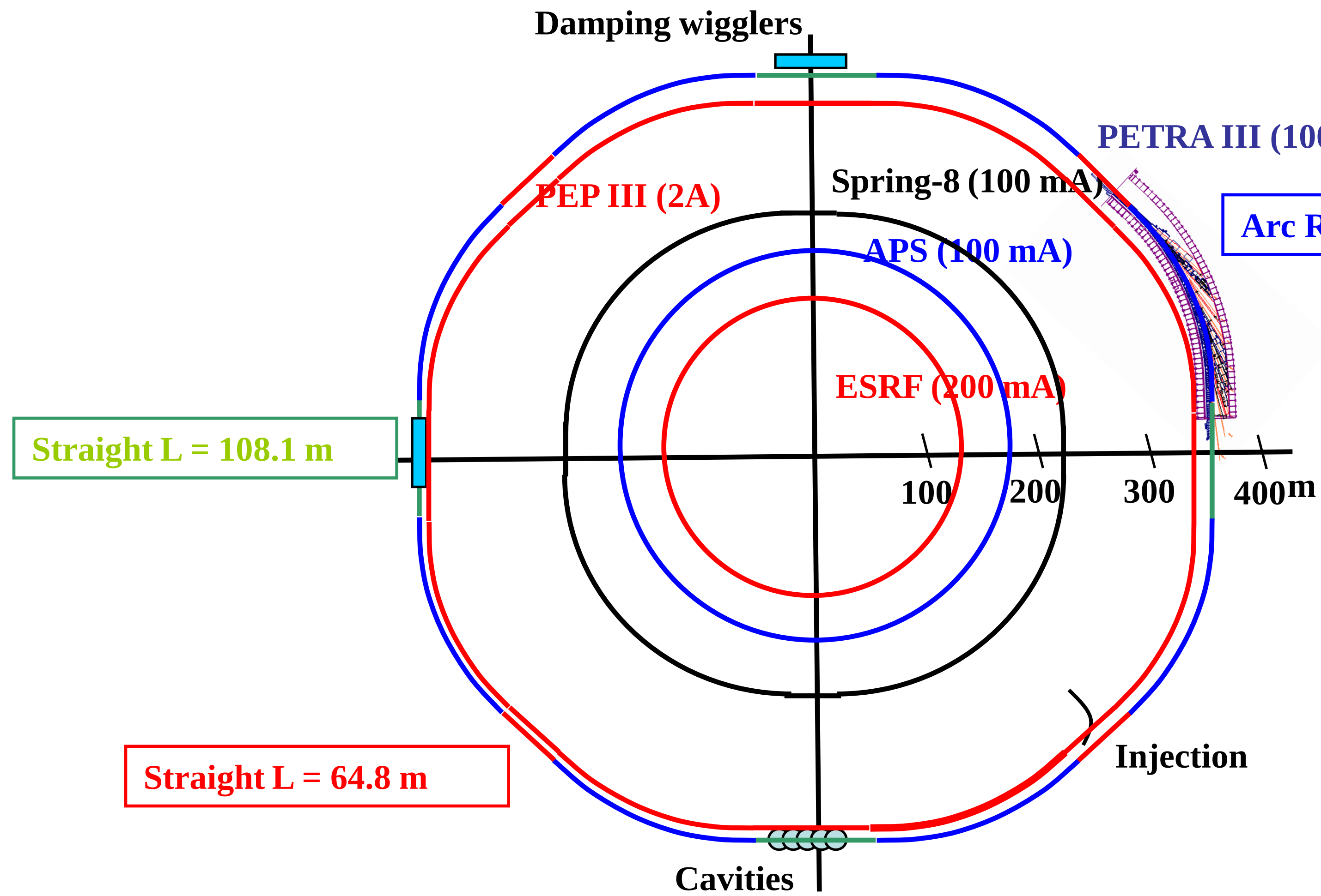




1 meV ~ 0.66 ps



Overview: 3rd generation sources

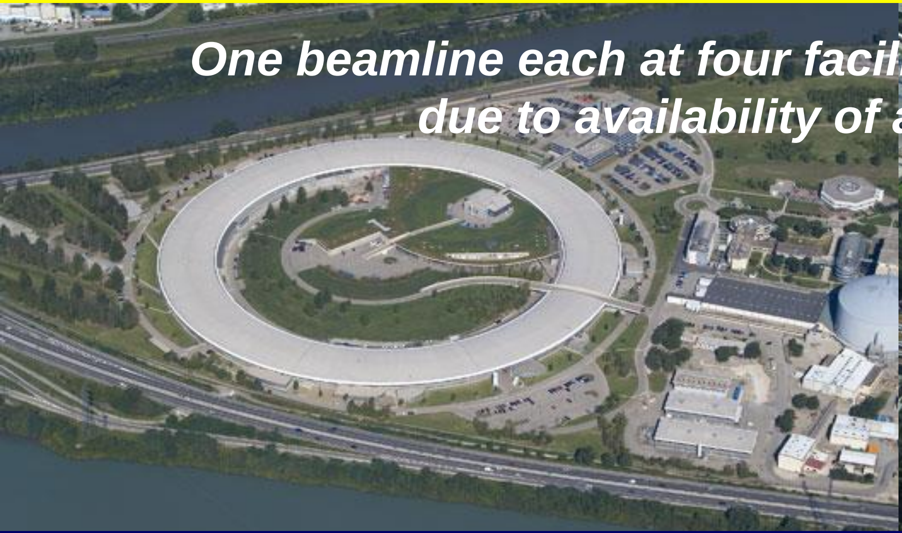


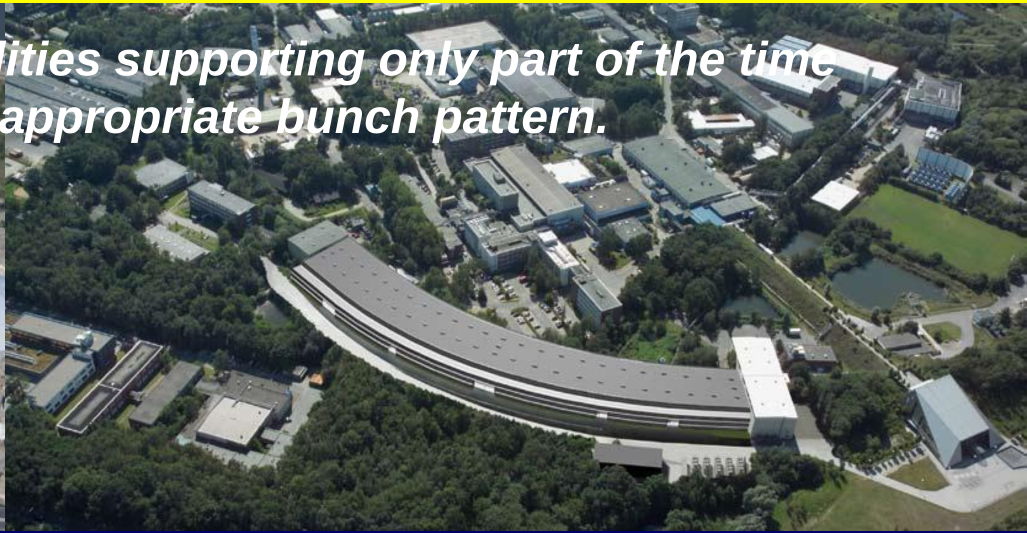
What Are the Requirements to Perform 14.4 keV ^{57}Fe Nuclear Resonance?

- **Low Emittance Lattice and Top-up Operation**
- **Undulator Source: High Average Brilliance and High Average Flux**
- **High Current per Bunch ~ 5-20 mA**
- **Bunch Width ~ 50-100 ps**
- **Bunch-to-Bunch Separation ~ 100 – 300 ns (> 150 ns for ^{57}Fe)**
- **Clean Bunches (Purity ~ 10^{-10})**
- **Energy Width of Excitation (14.41 keV) Beam ~ 1-3 meV**
- **A Detector with a Time Resolution < 1 ns**
  → Avalanche Photo Diodes (APDs)

Third-Generation Storage-Ring Facilities in the World where

One beamline each at four facilities supporting only part of the time due to availability of appropriate bunch pattern.





Over Subscription of Beamlines

Geoscience, High Pressure Research, Biophysics, Material Physics

Comparison of Characteristics of the X-ray Beam from Undulator Sources Used for NRS (around 14 keV) at Third-generation Storage-ring Facilities.

Properties	Operating Third-Generation Facilities (APS, ESRF, SPring-8)	PETRA-III *
Energy (GeV)	6.0 - 8.0	6.0
Maximum Current (mA)	100 - 250	100
Horizontal Beam Emittance (nm.rad)	3.0-4.0	1.0
Undulator Length (m)	4.8 - 5.0	20
Brilliance ph/s/0.1%BW/mrad^2	1 – 2 x 10^{20}	2 x 10^{21}
Vertical divergence (mrad)	3 - 5	3
Average Flux ph/s/meV	4 x 10^9	2-4 x 10^{10}
Bunch length (ps)	70-100	40
Rep Rate (MHz)	3.8-6.5	5.2
Photons/meV/bunch	0.6-1.0 x10^3	5-8x10^3

Mössbauer Resonant Nuclei

Radioactive Source: A

SR Source: A

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Ti	Pb	Bi	Po	At	Rn
Fr	Ra	Ac															
			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

List of Resonances Using the Third-Generation SR Facilities

Z	Isotope	E_o (keV)	t_o (ns)	Γ_o (neV)	σ_o (kbarn)	σ_o / σ_{el}
73	^{181}Ta	6.214	8730	0.075	1999	12
69	^{169}Tm	8.410	5.89	110	242	7
36	^{83}Kr	9.404	212.1	3.1	1226	152
26	^{57}Fe	14.4125	141.1	4.7	2564	426
63	^{151}Eu	21.514	14.0	47	243	29
62	^{149}Sm	22.496	10.3	64	120	17
50	^{119}Sn	23.871	35.6	26	1381	563
66	^{161}Dy	25.651	42	16	1110	176
19	^{40}K	29.23	6	110	281	1337
51	^{121}Sb	37.133	5	130	195	40
28	^{61}Ni	67.41	7.5	88	721	8100
93	^{237}Np	59.5	98	7	310	115

What future sources are suitable for NRS?

- PETRA III Storage Ring , DESY, Hamburg
- PEP III Storage Ring, SLAC, Stanford

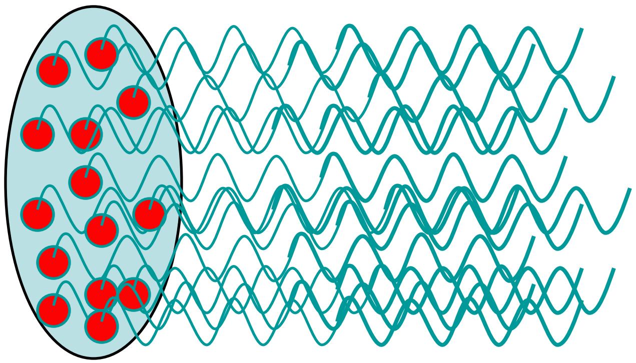
- LCLS SASE-XFEL, SLAC, Stanford
- European SASE-FEL, DESY, Hamburg
- SASE SCSS–SPring8, Japan
- PSI-XFEL, Switzerland

- Seeded XFEL - Future

What are the science directions using NRS at the future facilities?

Essence of a Free-Electron Laser

Incoherent Emission

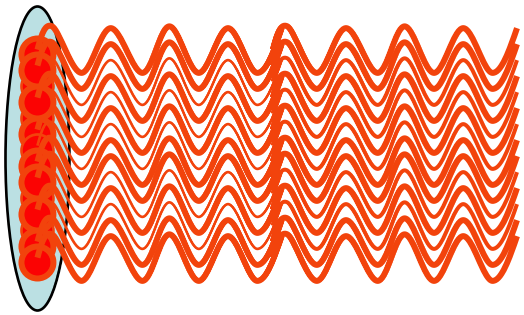


Each electron is independently radiating and the phases of the electric fields are random

$$E \propto \sqrt{N_e}$$

$$Intensity \propto N_e$$

Coherent Emission



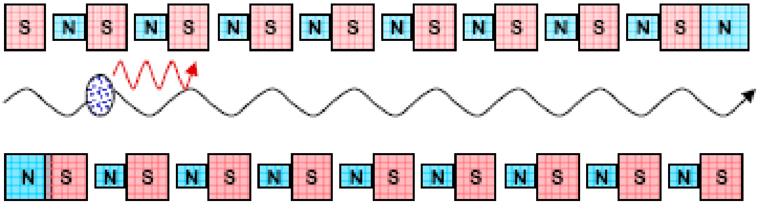
If the electrons are in lock synch and radiate coherently, electric field grows linear with number of electrons

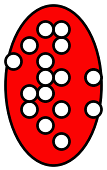
$$E \propto N_e$$

$$Intensity \propto N_e^{\ 2}$$

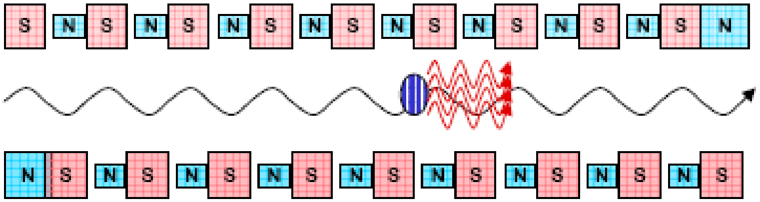
Self-Amplified Spontaneous Emission (SASE)

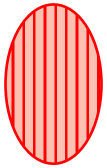
Spontaneous Emission from
***High-Density** Short Electron Bunch*



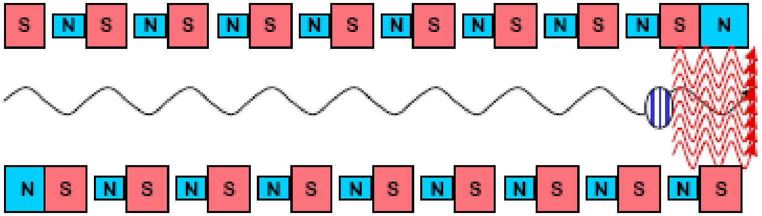


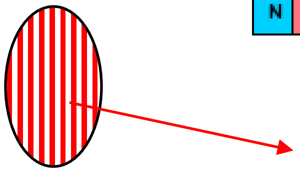
Phase Separation in Electron Bunches with a Period of the Wavelength of Radiation





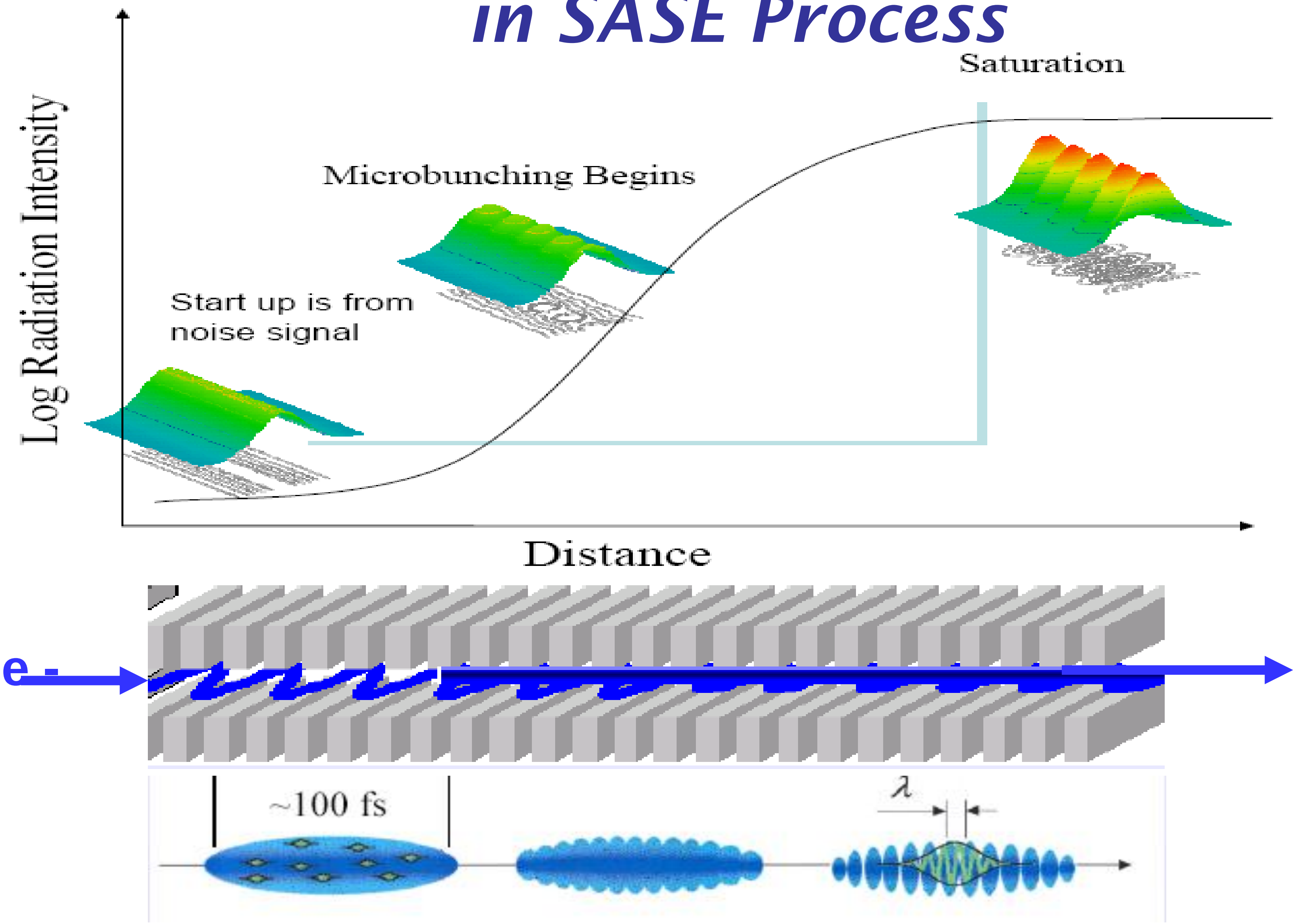
Microbunches Behave Like Super Charge and Emit Coherent Radiation with Intensity Enhancement





N_{mb} = number of electrons in coherent volume

Exponential Growth of Intensity in SASE Process

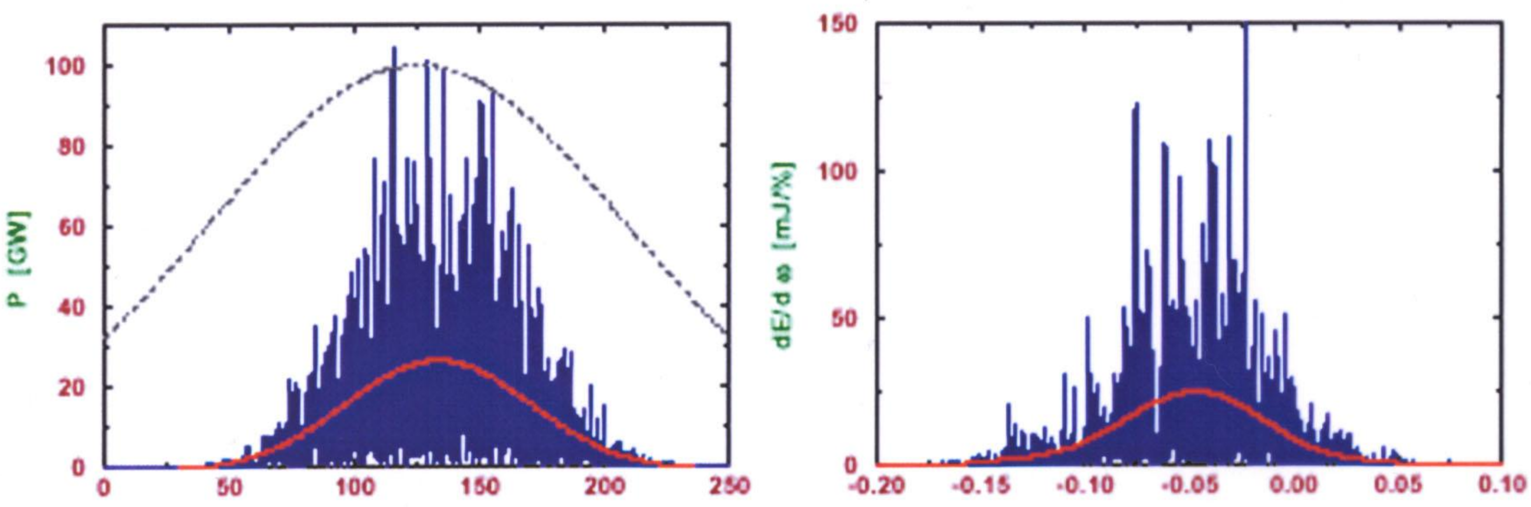


Does a SASE Laser Work?

1980	*First proposal*
1996	*First experimental observation*
	UCLA: λ = 16 μm
Dec. 1999	*Argonne: lasing at λ = 523 nm*
Sep. 2000	*Argonne: saturation at λ = 523 nm*
Feb. 2001	*VISA/BNL: saturation at λ = 800 nm*
Apr. 2001	*Argonne: saturation at λ = 265 nm*
Sep. 2001	*Hamburg -DESY: saturation at λ = 100 nm*
Oct. 2001	*First user experiments @ DESY- FLASH(TTF)*

These early developments led to VUV and X-ray FELs designs and construction.

A SASE FEL amplifies random electron density modulations



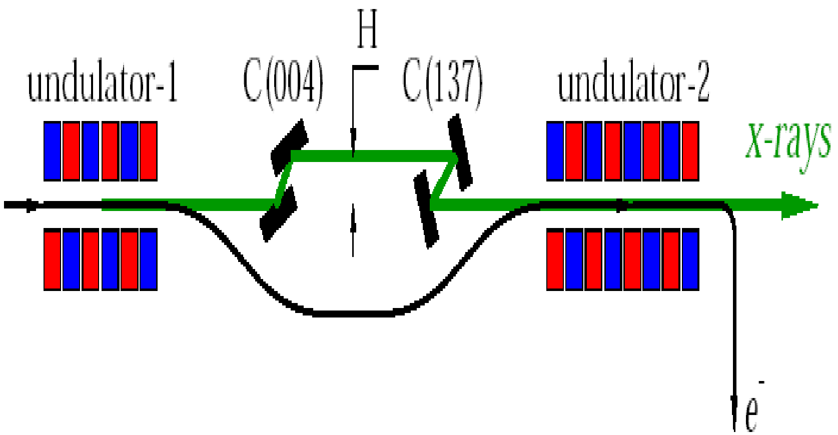
τ (fs) Δω/ω (%)

The SASE radiation is powerful, but noisy!

One solution: Impose a strong coherent modulation with an external laser source

Two of the Many Seeding Schemes
Seeded XFEL (SXFEL)

Two Stage Seeding

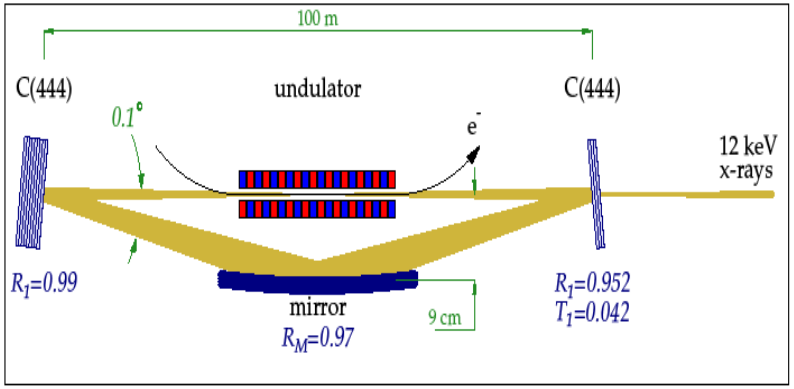


X-ray Monochromator and Debuncher

Saldin, et al. Nucl. Instrum. Methods in Physics A 475 (2001) 357–362

FEL **Oscillator**

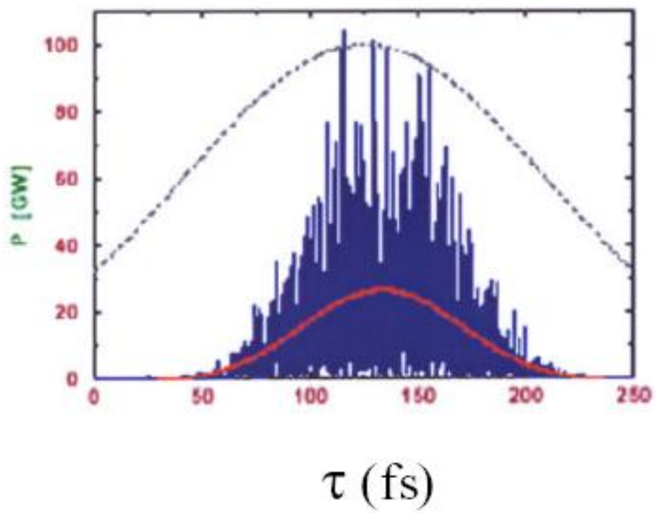
Diamond cavity for the X-FEL Oscillator

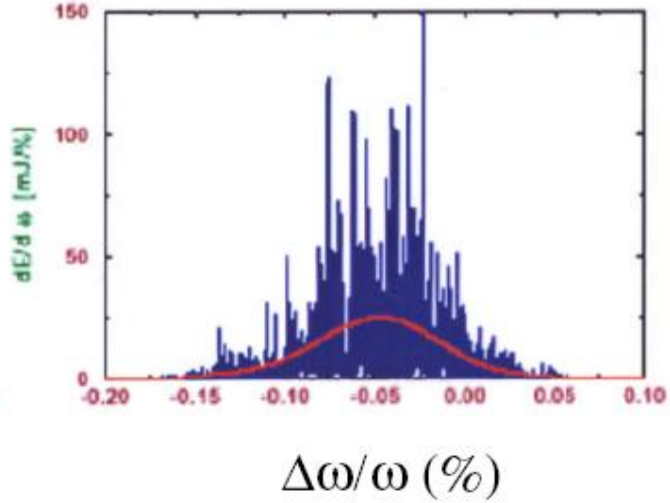


$R_1 \times R_2 \times R_M = 0.91$ $T_1 \simeq 0.042$

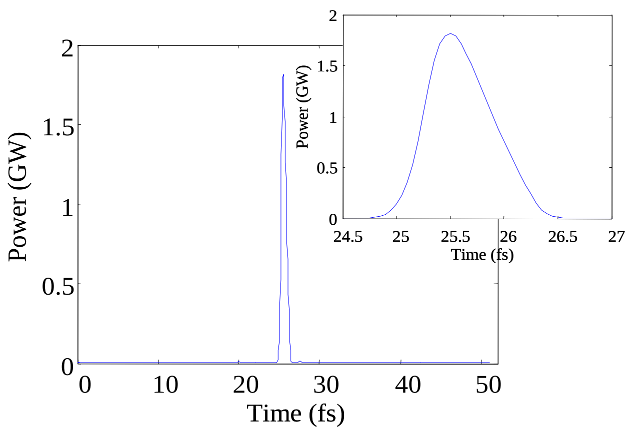
K.J. Kim and Y. Shvydko

SASE FEL Output in Time and Energy Domain

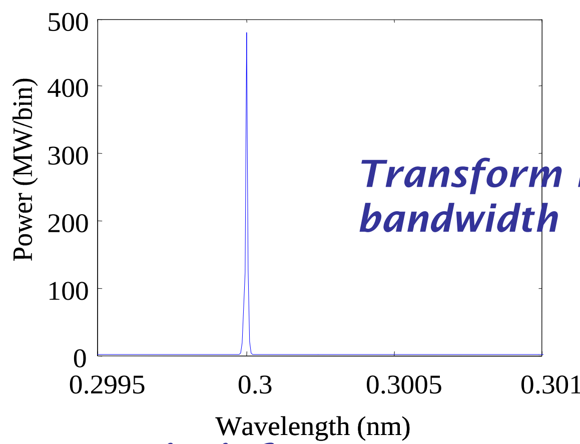




Seeded FEL Output in Time and Energy Domain



Seeded for fs time domain



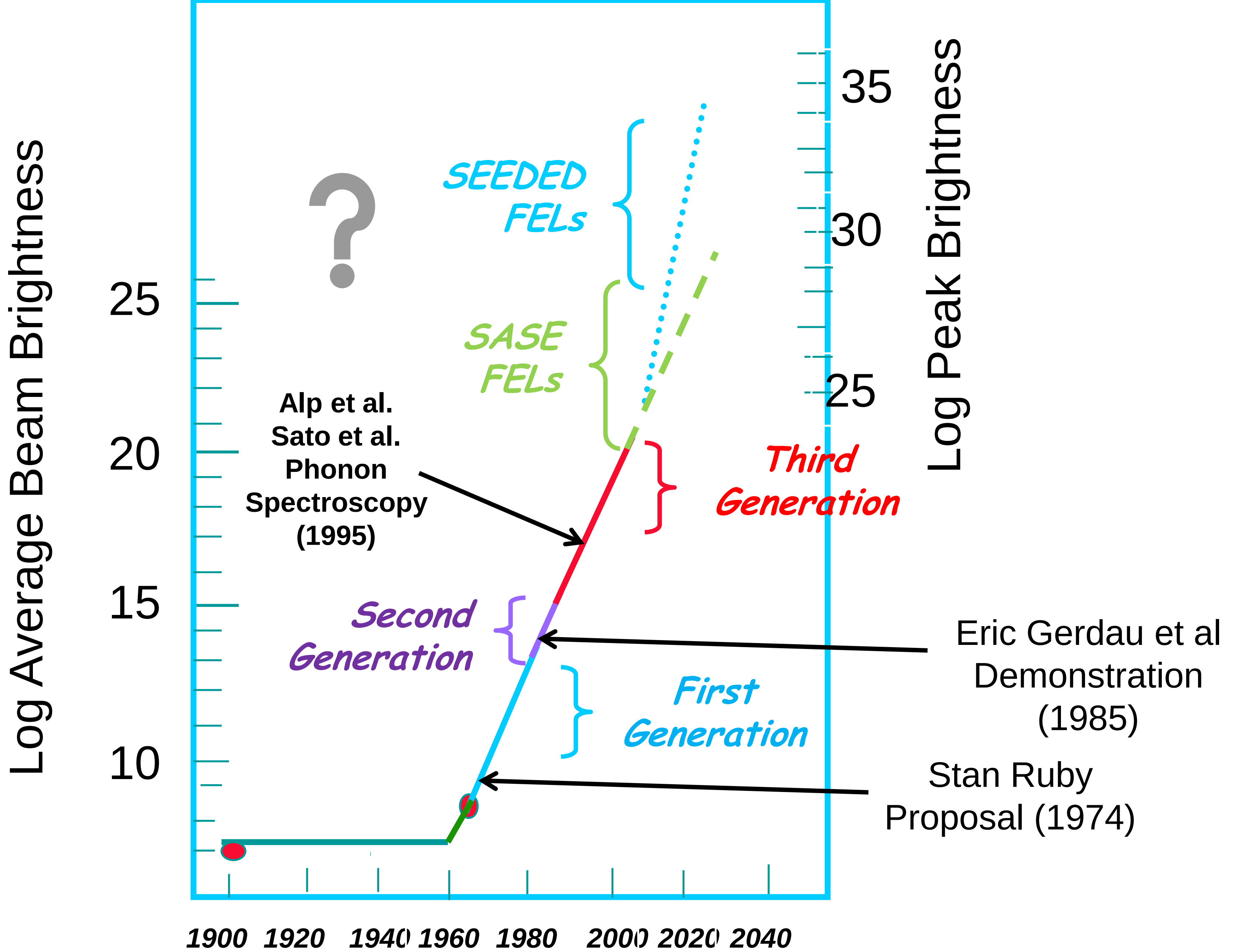
Transform Limited 20 meV bandwidth 10-15 keV beam

Seeded for meV energy domain

Courtesy: Bill Graves, David Moncton MIT

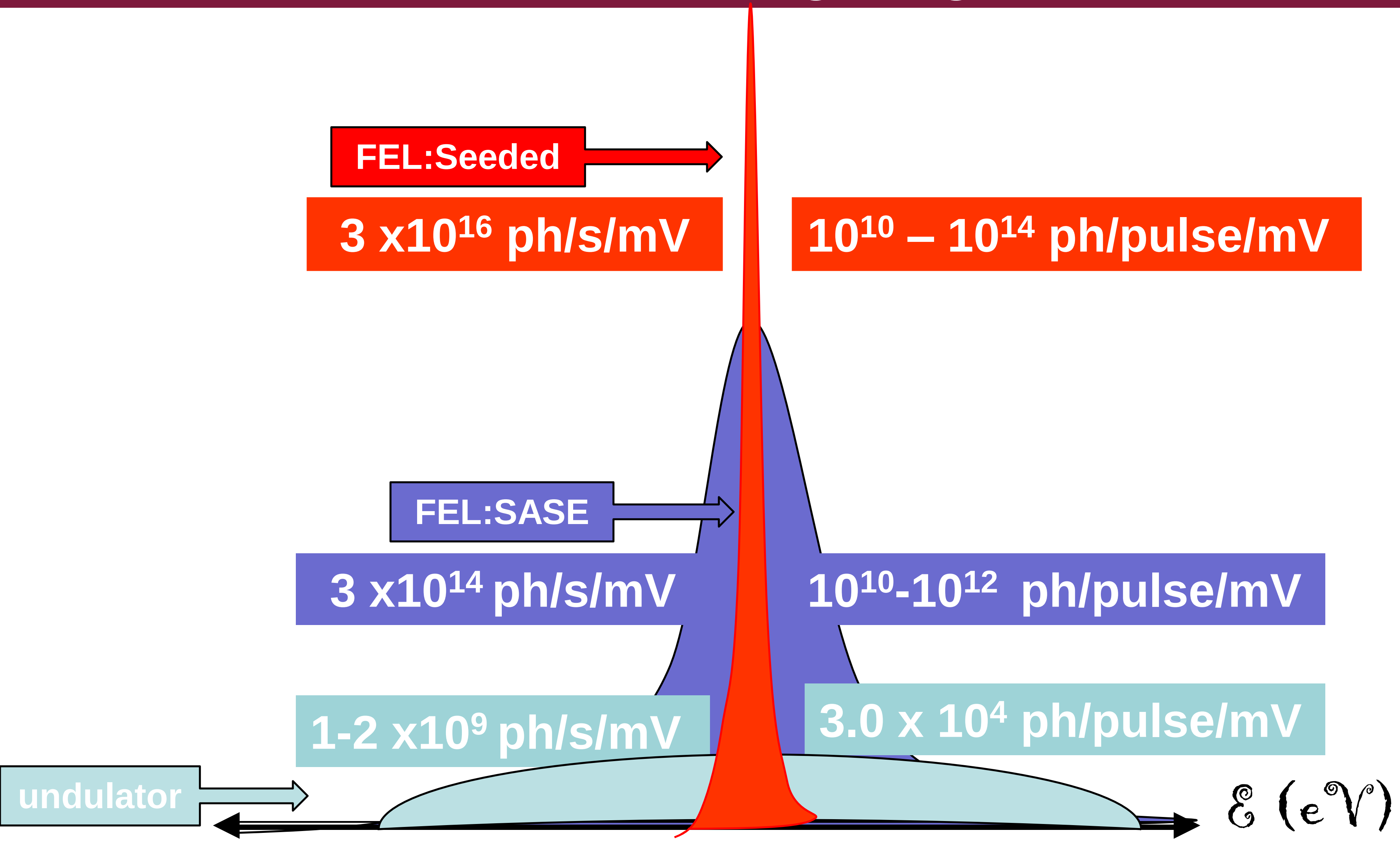
Brightness of Synchrotron Radiation Sources

Generation	Source	Electrons	U-Periods	Enhancement
2nd	Bend Magnet	$\sim N_e$	1	1
2nd	Wiggler	$\sim N_e$	$\sim N_p$	10
3rd	Undulator (APS, ESRF, SPring-8, ERLs)	$\sim N_e$	$\sim N_p^{2-x}$	10^4
Next	SASE FEL	$\sim N_{mb}^2$	$\sim N_p^2$	10^9
Next	Seeded FEL	$\sim N_e^2$	$\sim N_p^2$	10^{12}

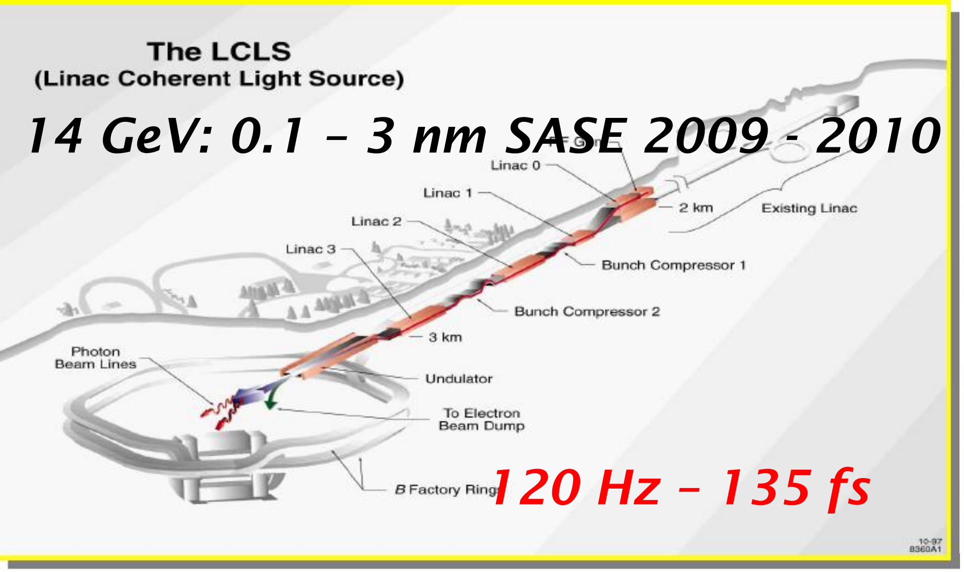


Log Average Beam Brightness
25
20
15
10
35
30
25
Log Peak Brightness
SEEDED FELs
SASE FELs
Third Generation
Second Generation
First Generation
Alp et al.
Sato et al.
Phonon
Spectroscopy
(1995)
Eric Gerdau et al
Demonstration
(1985)
Stan Ruby
Proposal (1974)
1900 1920 1940 1960 1980 2000 2020 2040

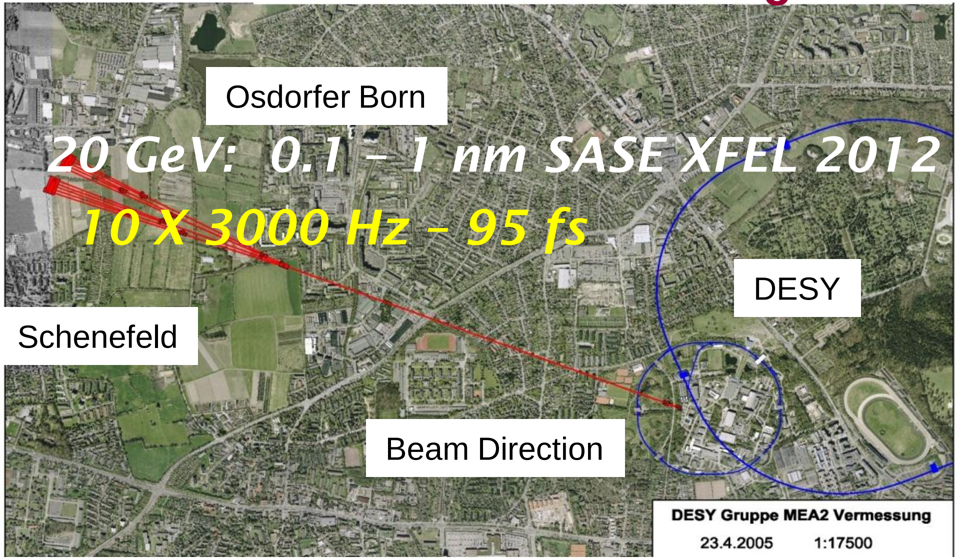
Gains to be made by SASE FEL and SXFEL over a Third Generation Storage Ring Source



LCLS- SLAC - Stanford

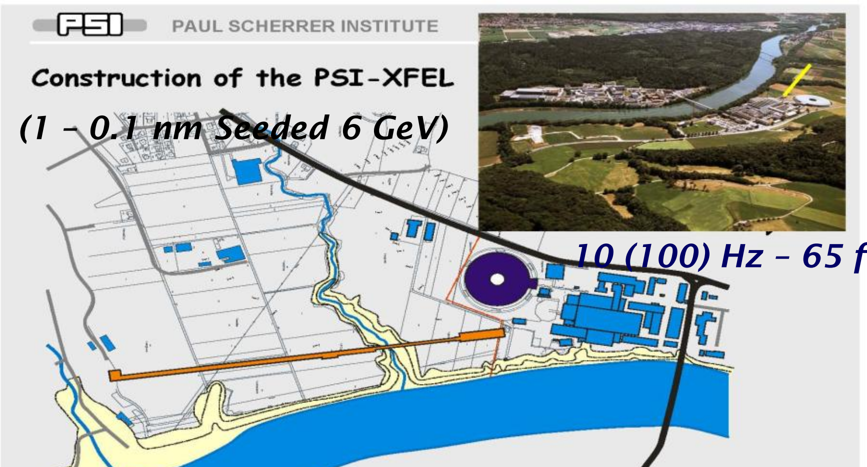


Euro-XFEL SASE DESY Hamburg



6 GeV: SASE SCSS–SPring8, Japan 2010

PSI- XFEL, Switzerland



- o Test of the acceleration concept: 2008 – 2011
- o Construction of an X-FEL: 2011-2016

^{57}Fe NRS Dream Source: Seeded XFEL In next 20 years

Energy (keV – 3rd Harmonic)	~ 14.4
Pulse length (fs)	~ 30-40
Energy Resolution (meV)	~ 20
Rep Rate (kHz)	~ 10 – 100
Photons/meV/pulse	~ 10^{10} - 10^{14}
Photons/meV/pulse/(1-20 nm)^2	~ 10^{8} - 10^{12}

Revealing the Interactions in Condensed Systems

10 eV

100 meV

1 meV

10 μeV

100 neV

1 neV

peV, feV, aeV, zeV,

Multiphonons & Multimagnons ~ 50 – 500 meV

Pseudogap ~ 30 meV – 300 meV

Optical phonons ~ 40 – 70 meV

Magnons ~ 10 meV – 40 meV

Superconducting gap ~ 1 – 100 meV

Dynamics of disordered and nano systems Boson peak in Amorphous Solids
Phasons in quasicrystals
Rotational excitations in liquids
Soft phonons

} ~ 10 -1000 μeV

Relaxation dynamics of fluids ~ 1 – 400 neV

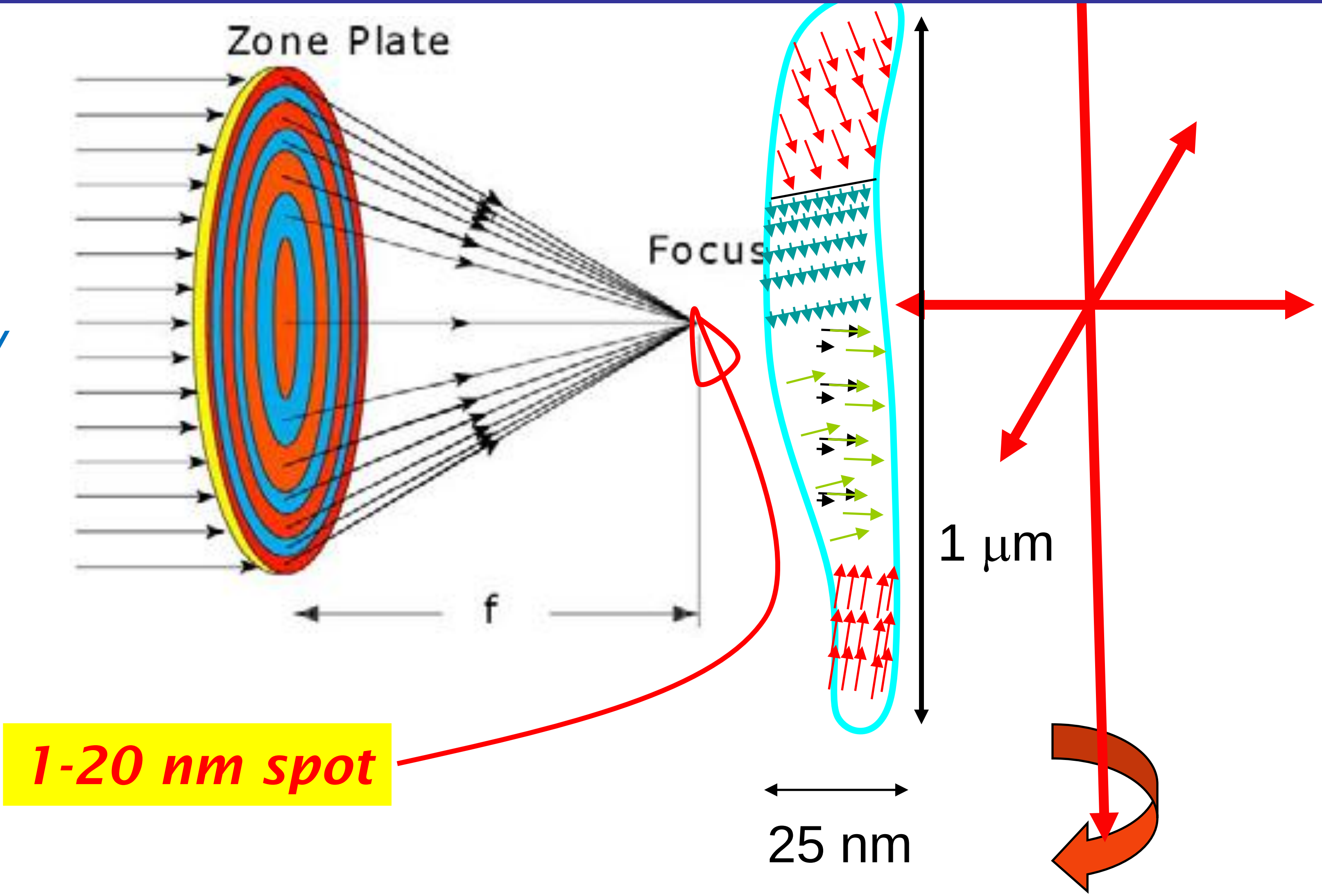
Hyperfine Interactions ~ 1 – 100 neV

?

Fundamental Physics

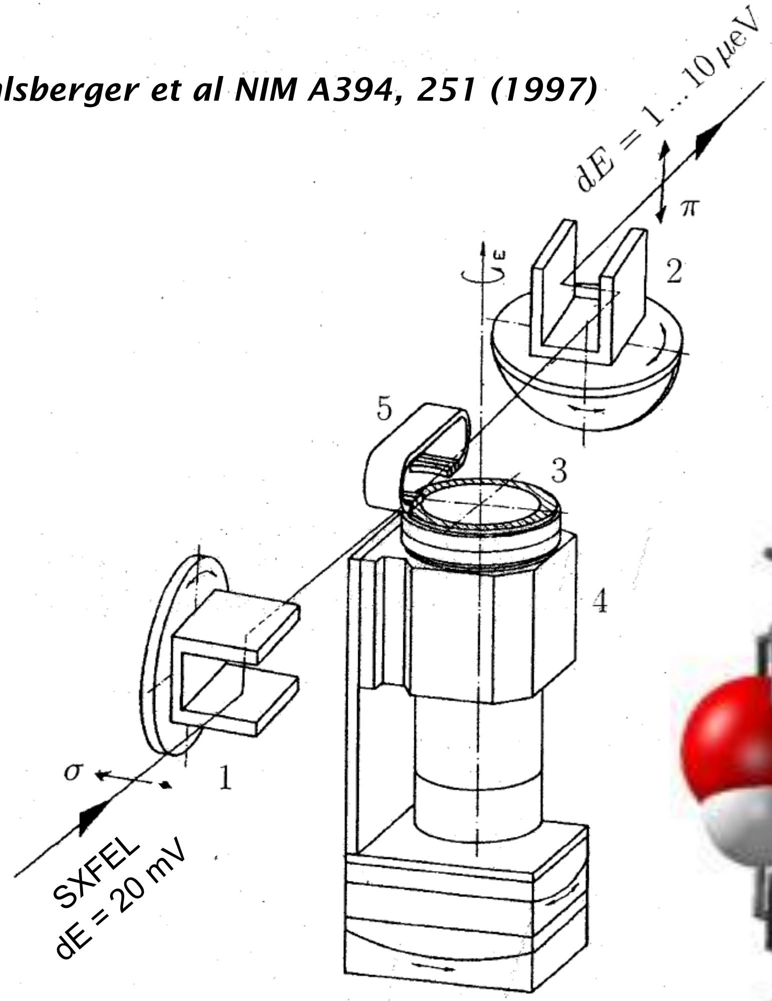
Probing Inside a Nanorod with 1-20 nm Beam

SXFEL
100 μV



Measuring the *spatially-resolved* diffusion dynamics in confined systems (nanofluidics)

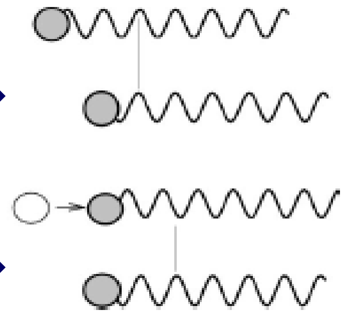
Röhlsberger et al NIM A394, 251 (1997)

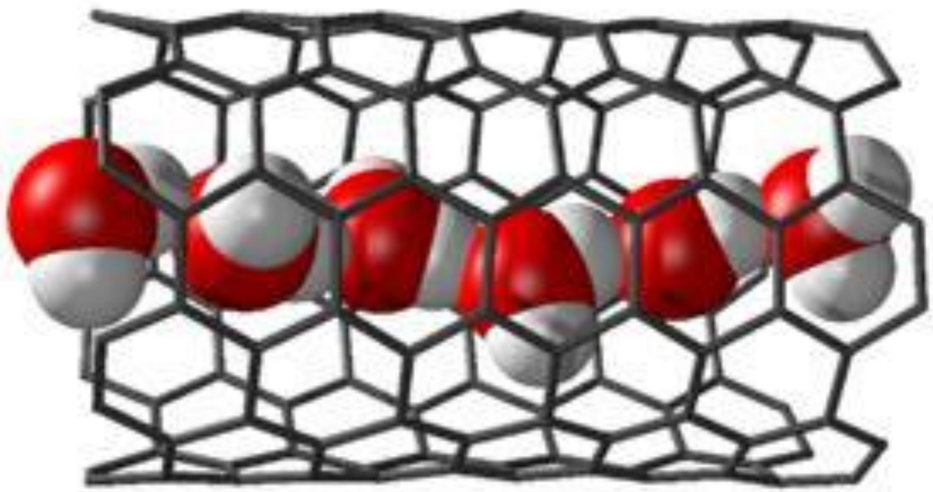


Coherent forward scattering

Before atomic diffusion
Scattered radiation in phase →

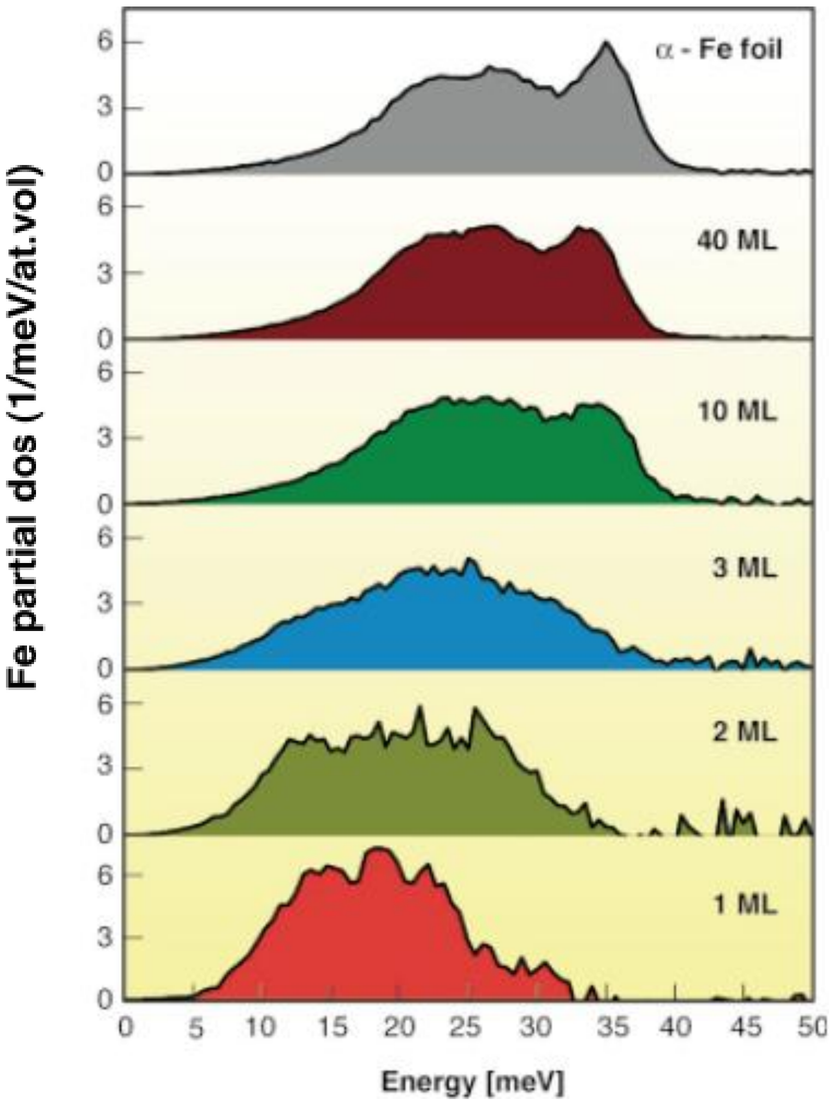
Atomic jump during nuclear life-time leads to dephasing scattered radiation →





Molecular diffusion through Carbon Nanotube

Decorated Nanodot Interfaces, Surfaces….



Partial Phonon Density Of States of Iron Films

Fe films deposited on W(110)

Transition from the bulk to a single iron monolayer

S. Stankov, R. Röhlsberger, T. Slezak, M. Sladecek, B. Sepiol, G. Vogl, A. I. Chumakov, R. Rüffer, N. Spiridis, J. Lazewski, K. Parlinski, and J. Korecki,

ESRF Highlights 2006

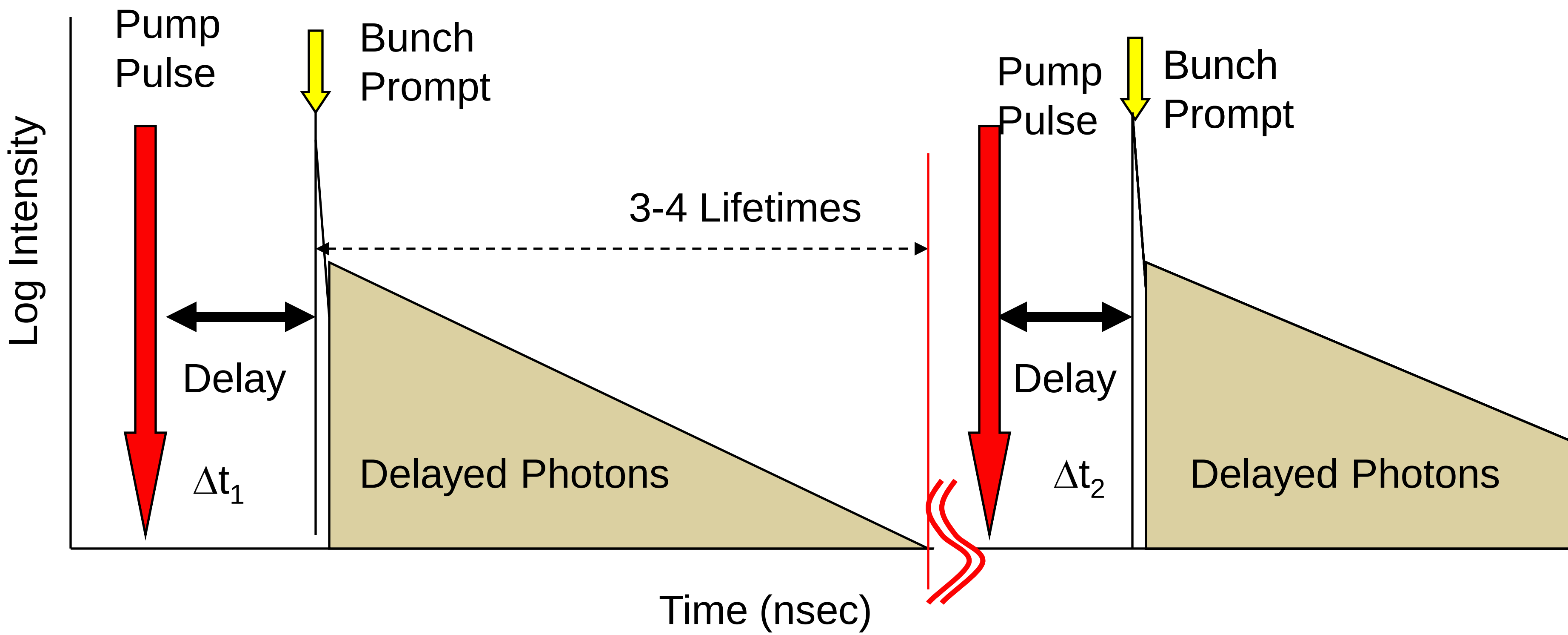
Phys. Rev. Lett. **99**, (2007) 185501.

Can One Probe Far-From-Equilibrium Dynamics?

Processes in the ps to μs time domain

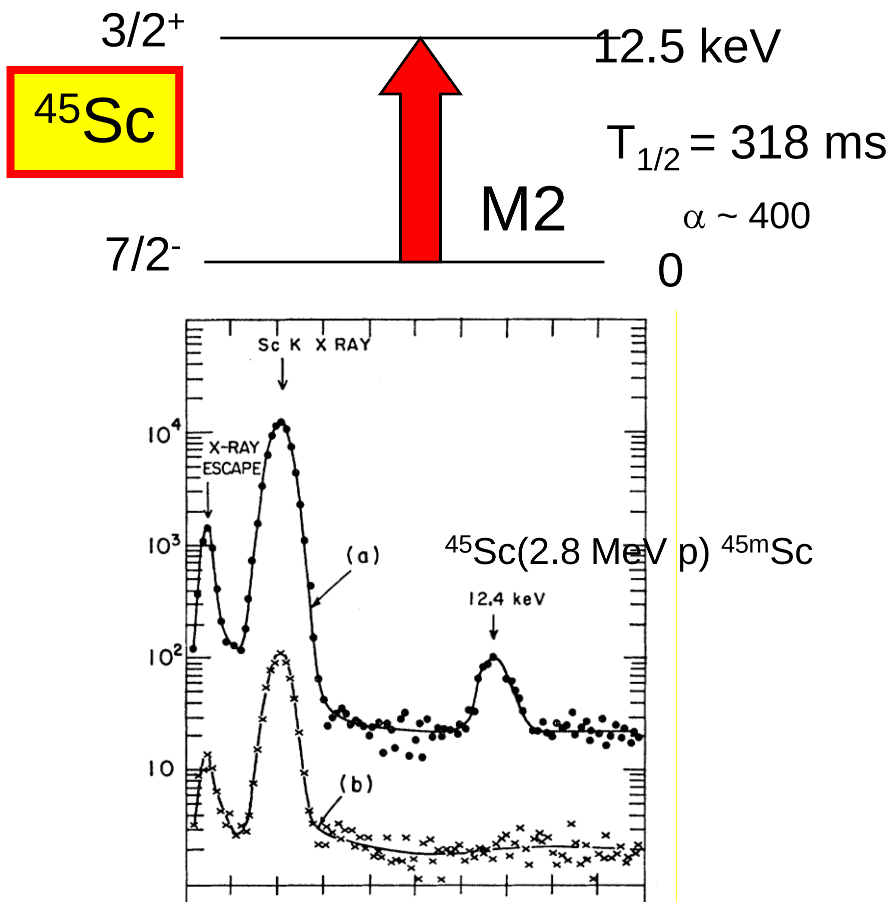
Pump: Electric, Magnetic, Laser, THz, Shock Wave, etc

Probe: NRS (both coherent & incoherent elastic and incoherent inelastic)

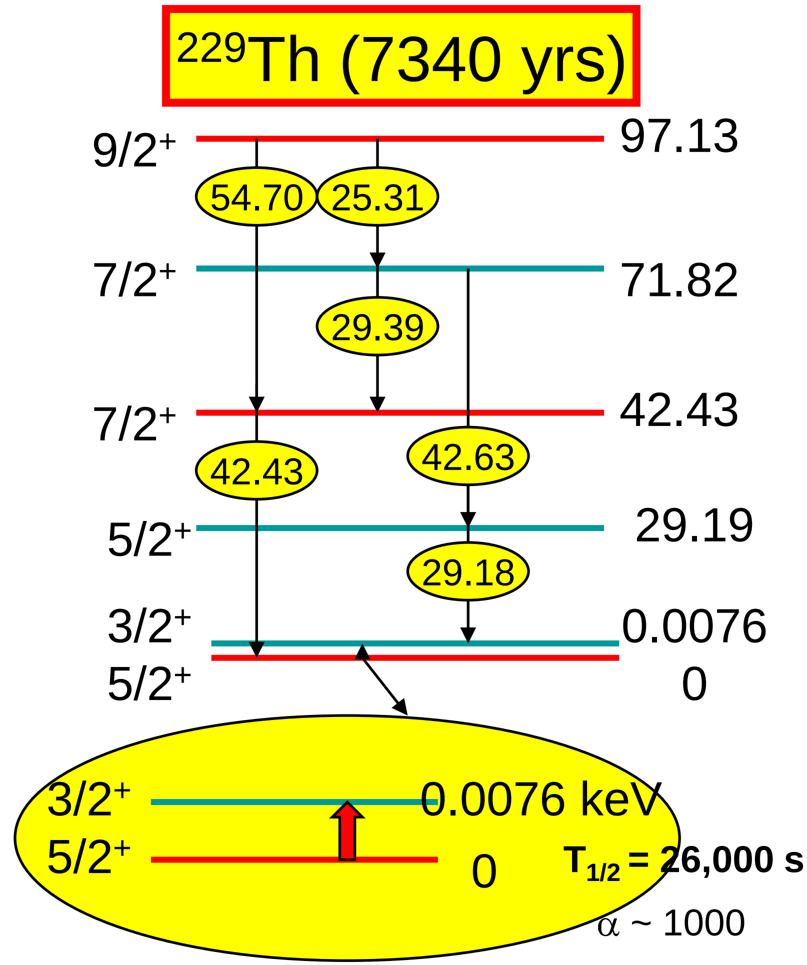


Can a single shot characterize the far-from-equilibrium processes?

Future of Ultimate Metrology



K W Jones et al PR 148, 1148 (1966)



Beck et al. PRL 2007

Nuclide	^{57}Fe	^{45}Sc	^{229}Th
Abundance, a (%)	Stable (2.14%)	Stable (100%)	7340 yrs (100%)
E_o	14.4 keV	12.4 keV	7.6 eV
I_g - I_e	$1/2^-$ - $3/2^-$	$7/2^+$ - $3/2^-$	$5/2^+$ - $3/2^+$
Multipolarity	M1	M2	M1
$T_{1/2}$	99 ns	318 ms	26,000 s
Γ	**4.6 neV (0.96 mm/s)**	**1.46 feV (35 nm/s)**	**17.5zeV (0.7 pm/s)**
$Q = \Gamma/E_o$	3.2×10^{-13}	1.18×10^{-16}	2.30×10^{-21}
α	8.21	~ 413	~1000
σ_o cm^2	2.56×10^{-18}	1.98×10^{-20}	2.11×10^{-14}
σ_o / σ_{el}	426	4	< 20
Unit Resonant Absorption Thickness (f=1, a = 100%)	4.6 (μm) (3.8×10^{19}/cm^2)	12.0 (μm) (4.8×10^{19}/cm^2)	1.6 (nm) (4.9×10^{15}/cm^2)

milli, micro, nano, pico, femto, atto, zepto, yocto

Coherent Nuclear Excitation of ^{229} Th

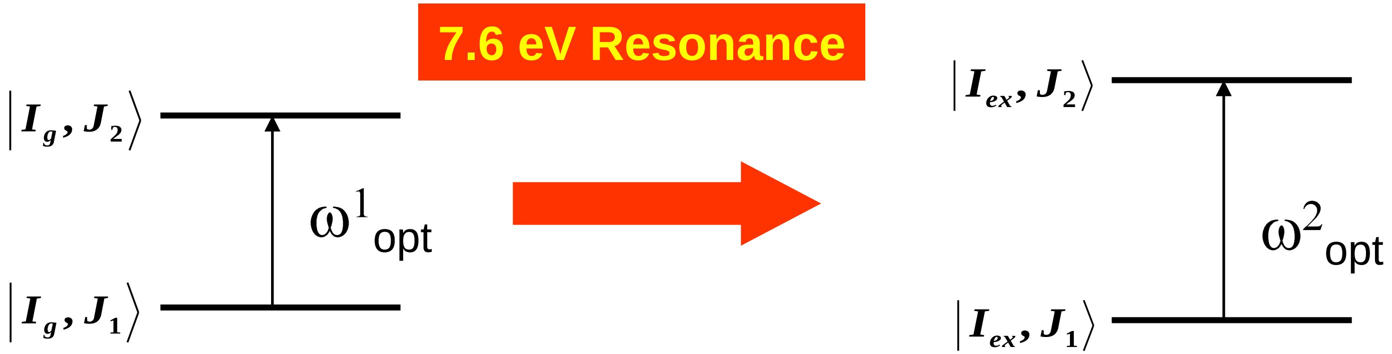
Issues:

- Hyperfine interaction of ground and excited moments with surrounding electronic and nuclear moments
- Elimination of all sources of line broadening
- Detection of 7.6 eV radiation

Approaches:

- *Intense laser excitation and detection through laser spectroscopy of atomic transitions (Habs: this symposium)*
- *Excitation of 29.19 keV state with x-ray SR/XFEL to populate 7.6 eV state (Argonne)*
- *Preparation of stripped nuclei in storage-cooler rings or in ion-traps (Suggestion: P. Kienle, TU Munich)*

Optical Detection Scheme: Test of Fundamental Physics



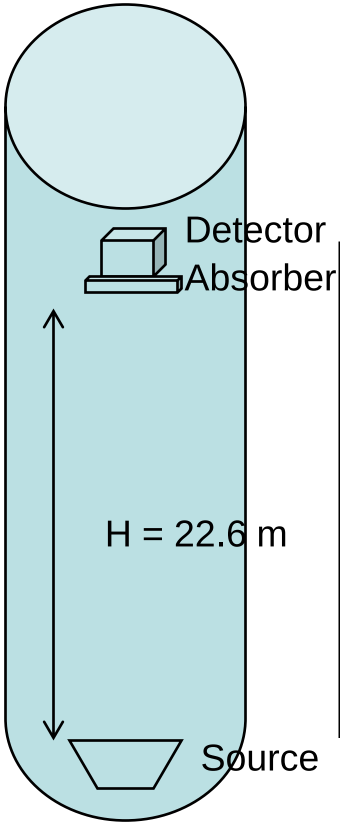
$$H^{hf}_{I_g, I_e, J_{1,2}, L_{1,2}, S_{1,2}} = \overline{A}\cdot(\overline{I}_{g,e}\cdot\overline{J_{1,2}})$$

E. Peik, Chr. Tamm, Europhys. Lett. 61, 181 (2003)

Dietrich Habs , The Lowest Nuclear Transition in ^{229}Th: This Symposium Presentation

Gravitational Red Shift Scheme

$$\Delta E_g = E_o gH/c^2 \text{ (eV)}$$



Jefferson Tower
Harvard

Nuclide	^{57}Fe	^{45}Sc	^{229}Th
ΔE_g (eV)	3.54×10^{-11}	1.35×10^{-15}	8.3×10^{-21}
H	22.6 m	1 mm	10 μm
$\Delta E_g/\Gamma$(%)	0.8	92	47

Th^{4+} ($6d^0$)

Summary:

1. Metrology, Fundamental Physics, Cosmology, Gravitation

Nobel Prize	Laureate (s)	Technique	Resolution	Applications
1960	Mössbauer	NRS	1: 10^{13}	Numerous
2005	Hall and Hänch	Frequency Comb	1: 10^{15}	Atomic Clock Metrology
????	?????	NRS Hyperfine Comb	1: 10^{16} (^{45}Sc) 1: 10^{21} (^{229}Th)	Fundamental Science Cosmology Gravitation

Flambaum, PRL 97, 092502(2006); Burvenich et al, PRL 96, 142501 (2006)

2. Nonlinear Quantum Optics

Non-linear Nuclear Quantum Optics

Incident Process	Generated Frequencies	Non-linear Frequencies	Susceptibility
Parametric conversion	ω_π	$\omega_\sigma, \omega_\iota$ ($\omega_\pi = \omega_\sigma + \omega_\iota$)	$\chi^{(2)}(\omega_2; -\omega_3, \omega_1)$
2nd (and Higher) Harmonic Generation	ω_1	ω_2 ($\omega_2 = 2\,\omega_1$)	$\chi^{(2)}(\omega_2; \omega_1, \omega_1)$
Mixing	ω_1, ω_2	ω_3 ($\omega_3 = \omega_1 + \omega_2$)	$\chi^{(2)}(\omega_3; \omega_2, \omega_1)$
Intensity dependent index of refraction	ω_1	ω_1	$\chi^{(3)}(-\omega_1; \omega_1, -\omega_1, \omega_1)$
2 photon absorption	ω_1	–	$\chi^{(3)}(\omega_1; \omega_1, -\omega_1, \omega_1)$

$$[c\kappa_1 + c\kappa_2] = \omega_M = 3 \text{ X } 7.2 \text{ keV} = 14.4 \text{ keV}$$

XFEL field strength is ~ 1 $V.A^{-1}$ ⟹ 10 keV

(Seb Doniach JSR 7, 216,2000)

Extra Reading

TDR XFEL workshop series
Nuclear Resonant Scattering at the TESLA XFEL

Edited by H. Franz and U. van Burck
February 25, 2001

Scientific opportunities in nuclear resonance spectroscopy from source-driven revolution
G. K. Shenoy · R. Röhlsberger
(in press)

Thank You