EuXFEL Usermeeting,
25 January 2023

New regimes in nuclear resonant scattering at the European XFEL

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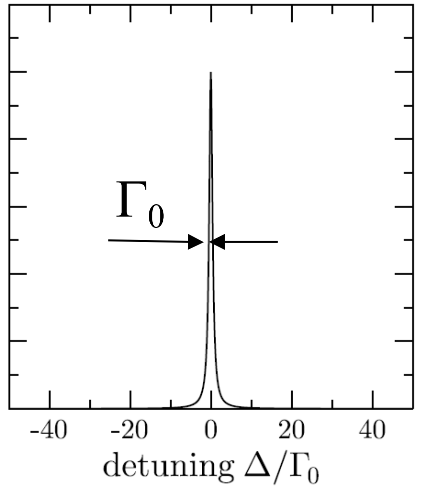
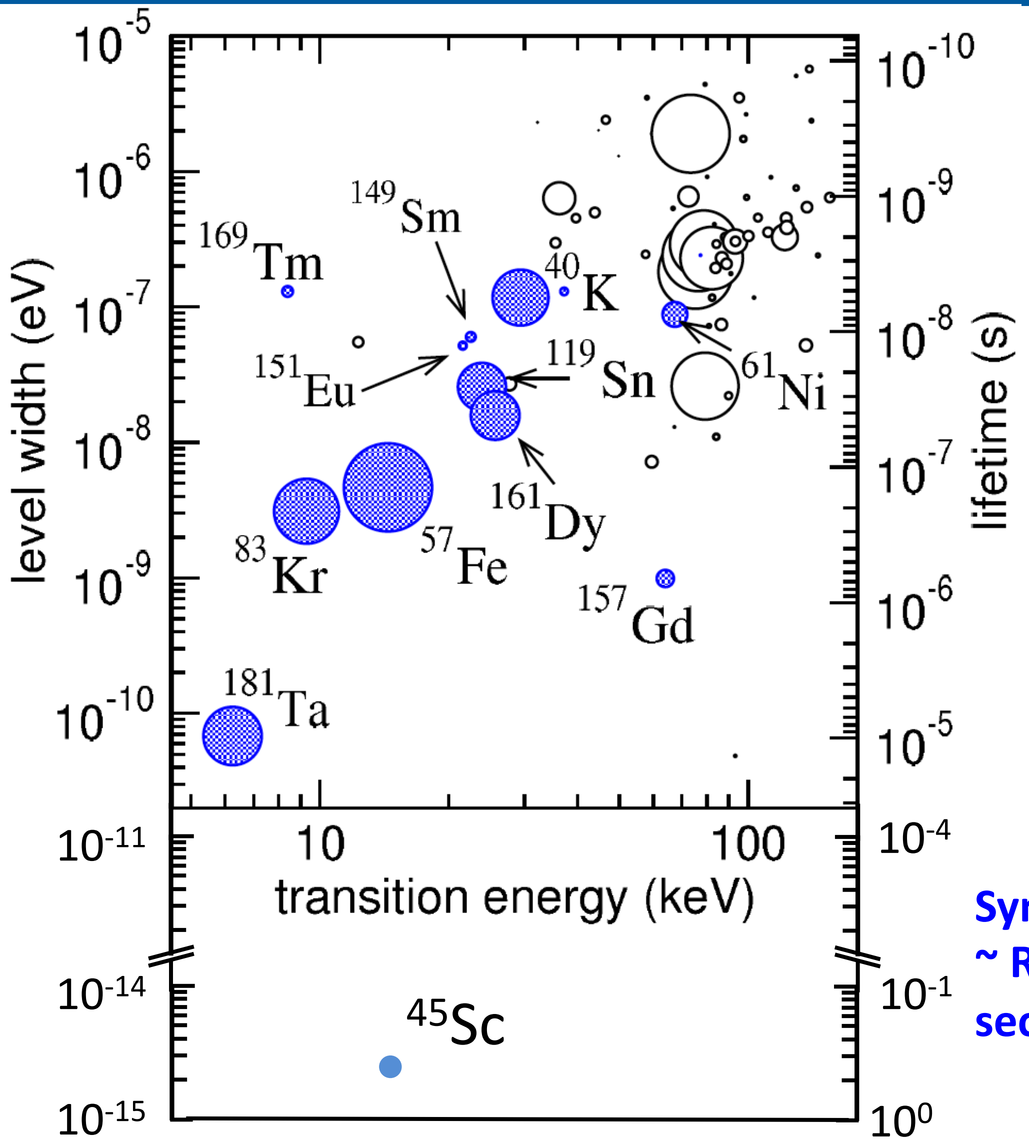
Friedrich-Schiller-Universität Jena

Deutsches Elektronen-Synchrotron DESY

Nuclear resonances for Mössbauer spectroscopy

Nuclear resonances as ideal two-level systems





Isotope	E_0 [keV]	Γ_0 [neV]	Γ_0/E_0
^{181}Ta	6.23	0.067	$1.1 \cdot 10^{-14}$
^{169}Tm	8.41	114	$1.4 \cdot 10^{-11}$
^{83}Kr	9.40	12.0	$1.3 \cdot 10^{-12}$
^{57}Fe	14.41	4.7	$3.3 \cdot 10^{-13}$
^{153}Eu	21.53	47.0	$2.2 \cdot 10^{-12}$
^{149}Sm	22.49	64.1	$2.9 \cdot 10^{-12}$
^{119}Sn	23.87	25.7	$1.1 \cdot 10^{-12}$
^{161}Dy	26.65	16.2	$6.1 \cdot 10^{-13}$
^{67}Zn	93.3	0.049	$5.3 \cdot 10^{-15}$
^{45}Sc	12.36	$1.4 \cdot 10^{-6}$	$1.1 \cdot 10^{-19}$
^{229}Th	0.0083	$1.8 \cdot 10^{-11}$	$2.6 \cdot 10^{-21}$

^{57}Fe

E_0 = 14.4 keV

λ = 0.086 nm

Γ_0 = 4.7 neV,

τ_0 = 141 ns

Symbol size ~ Resonant cross section σ_0

^{45}Sc

E_0 = 12.4 keV

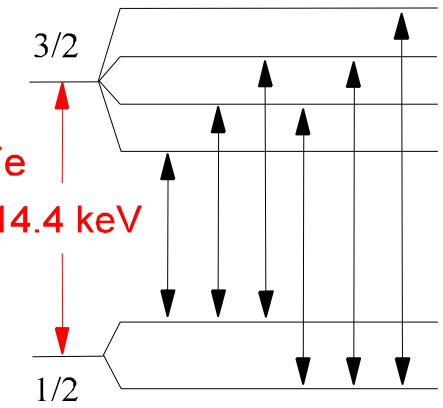
λ = 0.1 nm

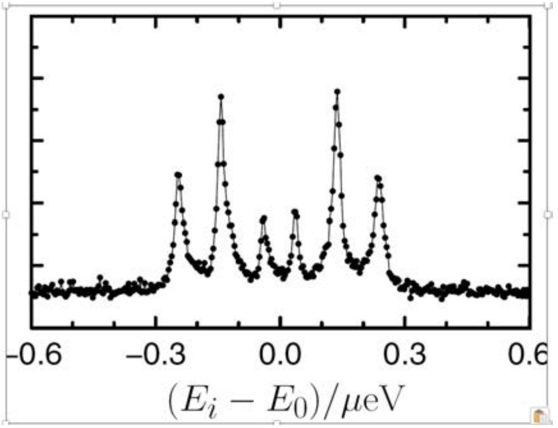
Γ_0 = 1.4 feV,

τ_0 = 460 ms

^{57}Fe in a ferromagnetic environment: The nuclear Zeeman effect

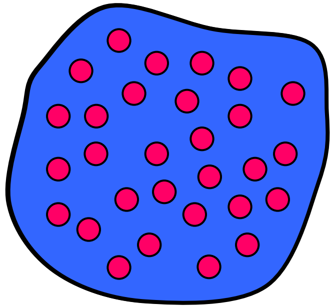
Broadband excitation of hyperfine-split nuclear levels

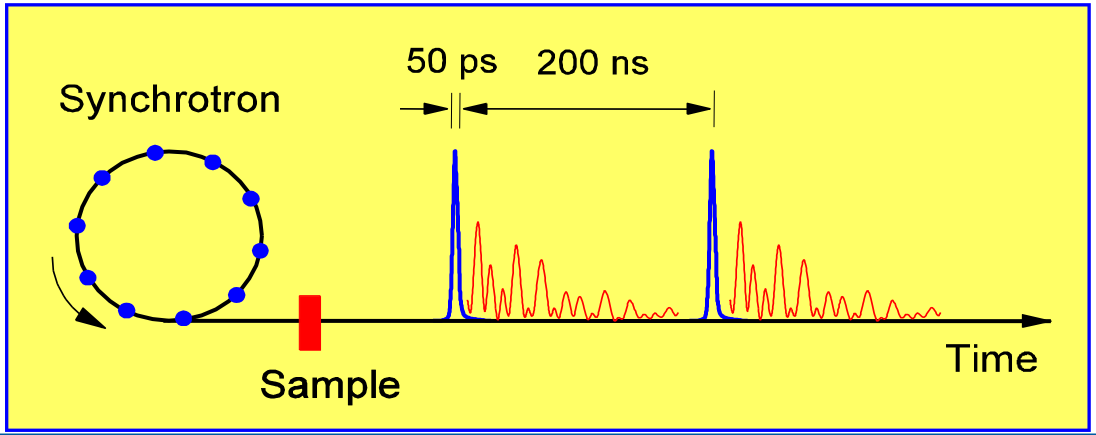




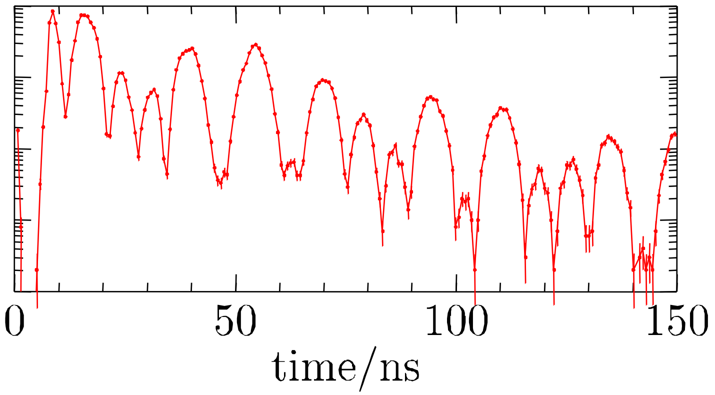
The temporal beat pattern is a fingerprint of the electronic and magnetic structure of the sample.

Applications
in all natural sciences





Temporal beats



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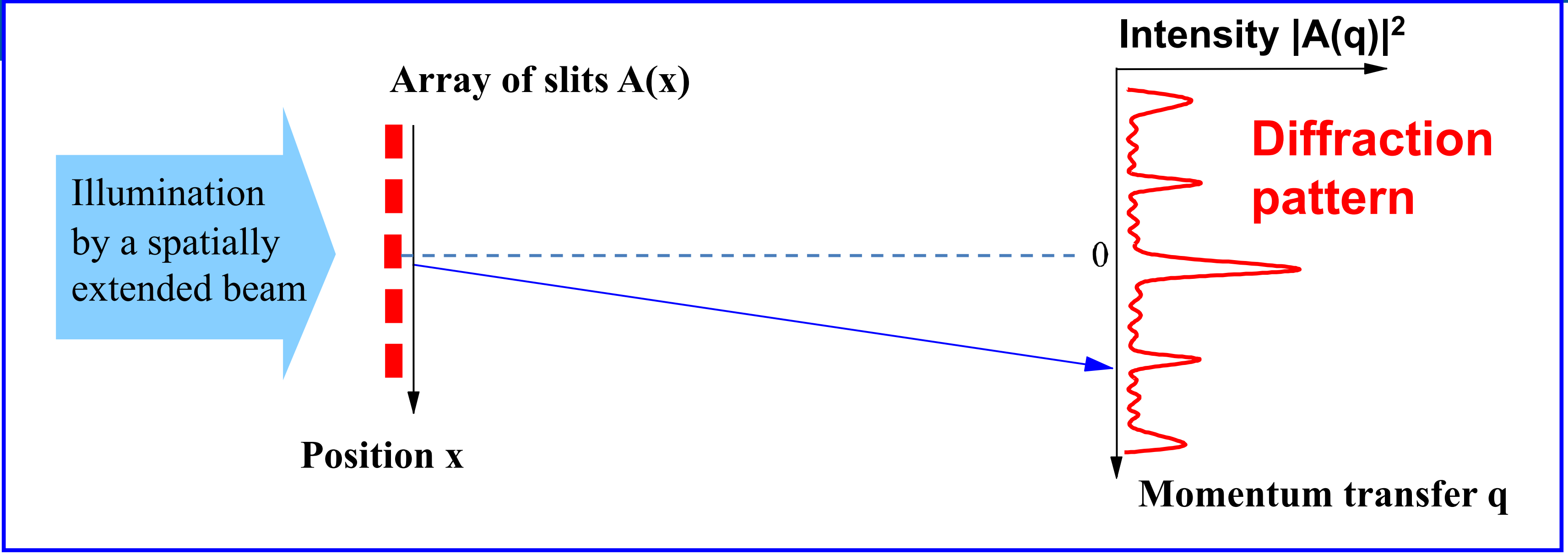
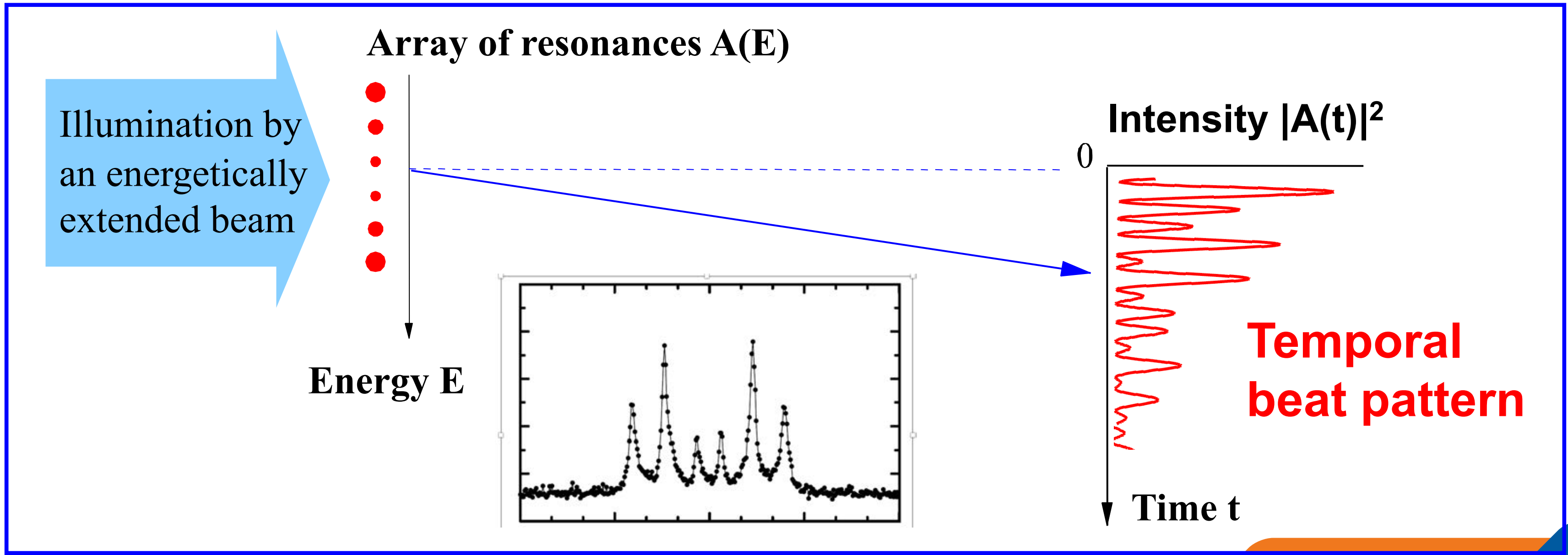
Structure Determination in Space and Time: Diffraction





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Milestones in the greater Hamburg research area

1985 @ DORIS by Erich Gerdau, Rudolf Rüffer et al.

2010 @ PETRA III

2022 @ European XFEL

2027 @ XFELO (?)

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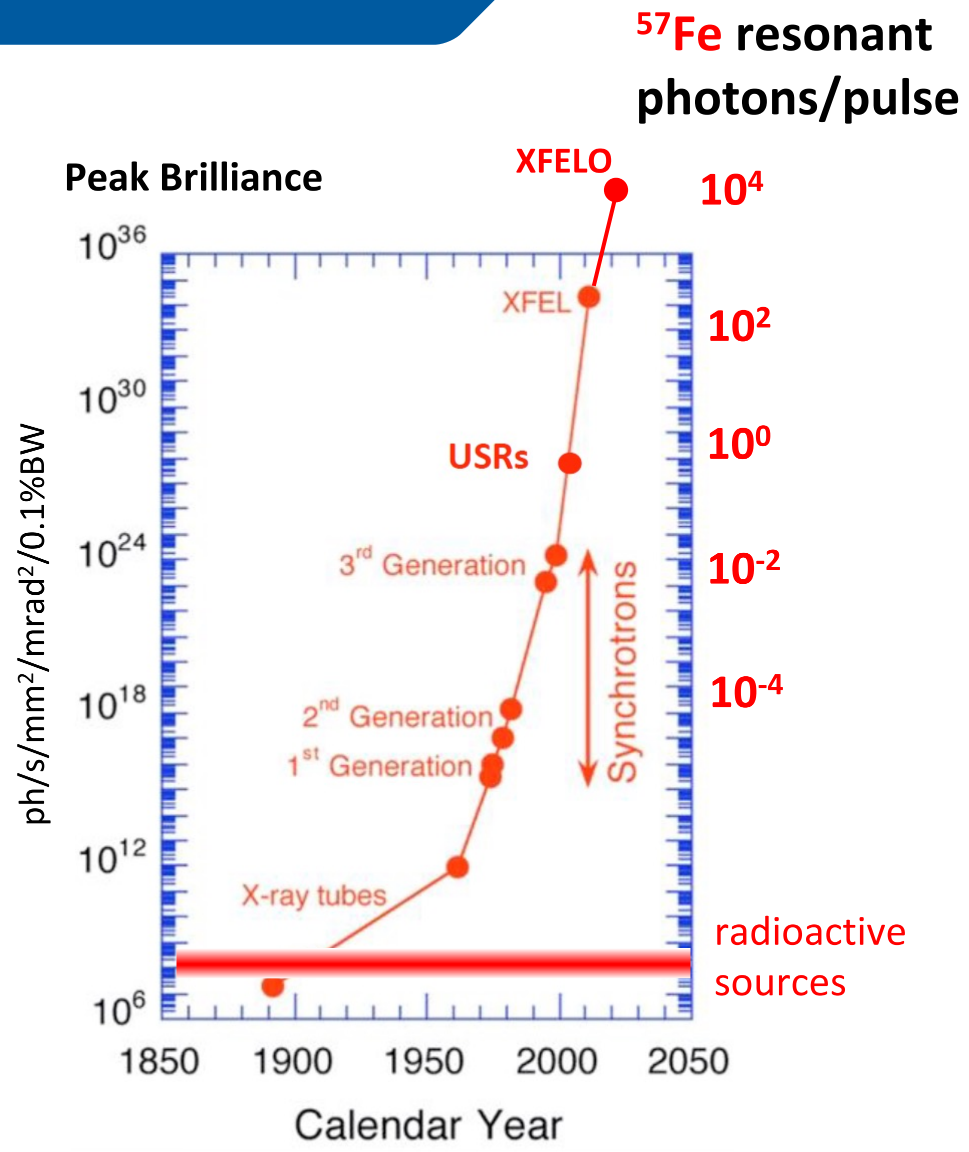
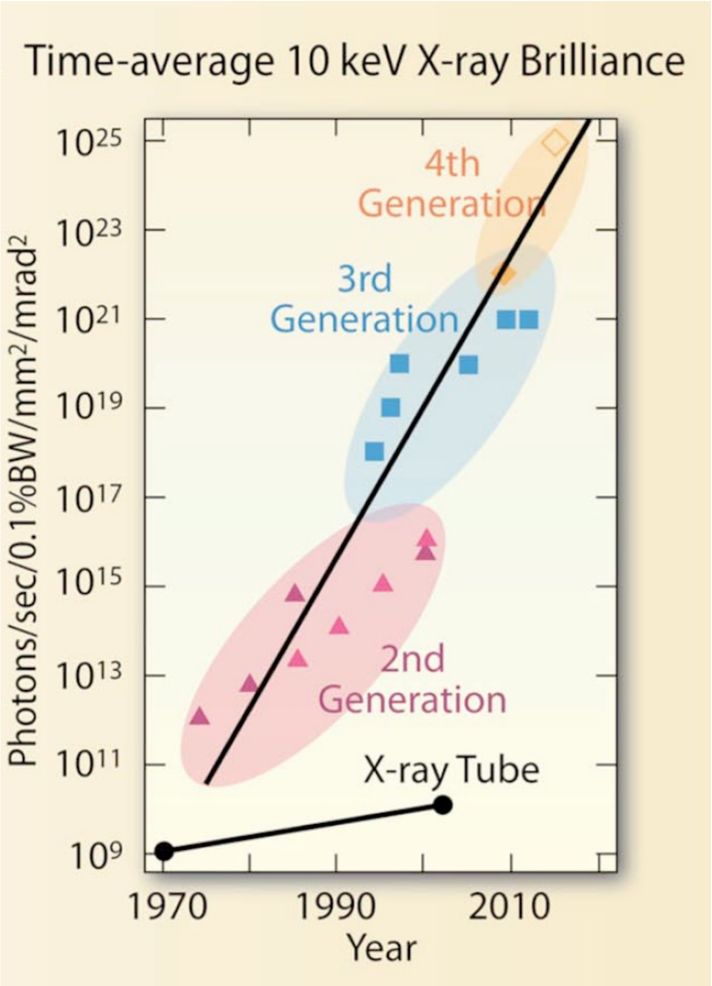
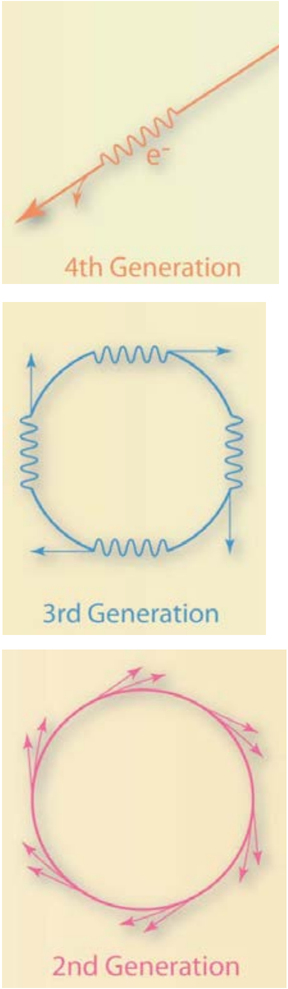
X-ray sources for excitation of Mössbauer nuclei: Evolution of peak brilliance



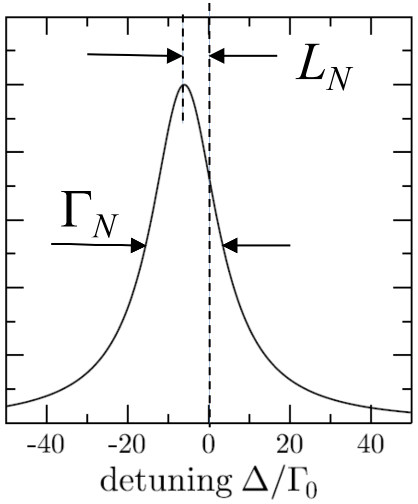




From: G. E. Ice, J. D. Budai, J. W. L. Pang, Science 334, 1238 (2011)

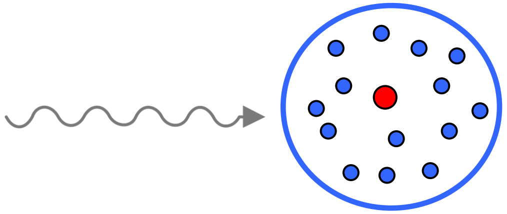
How does NRS depend on the number of excitations in the system ?

Single-photon, multi-atom



… Synchrotron radiation

~ 0.01 resonant photons/pulse

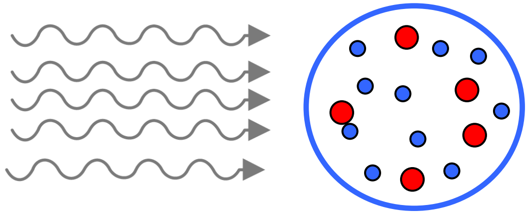


Single-photon collective effects

- Single photon superradiance
- Collective Lamb shift

… XFEL SASE radiation

Several 100 resonant photons/pulse



Multi-photon, multi-atom scattering

New phenomena !

- **Photon correlations**
- **Nonlinear effects**
- **Entanglement, nonclassical states**
- **Collective effects (e.g. resonance energy shifts)**

Multiphoton Collective Lambshift

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Cooperative emission from nuclei excited by an x-ray laser

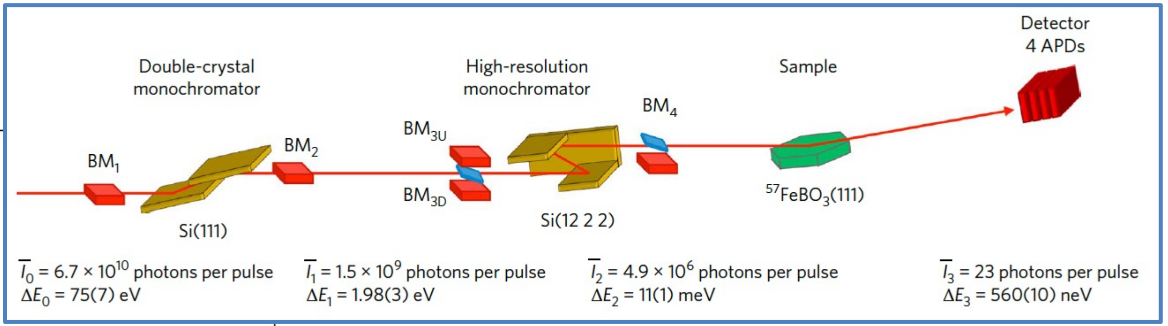
Entering the regime of multiphoton collective excitations

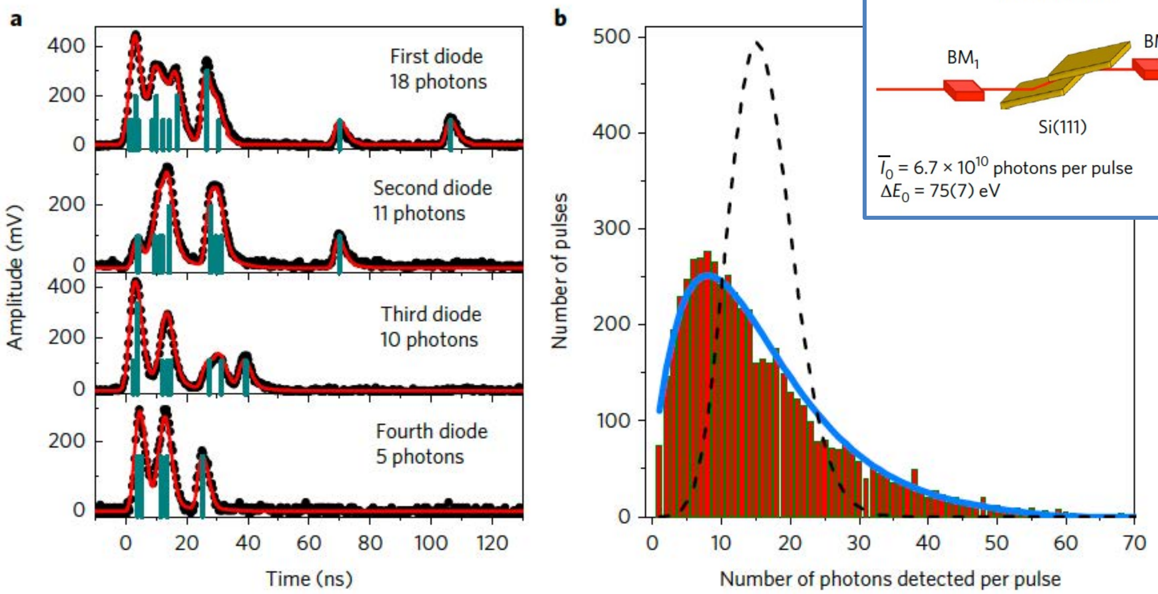
Superradiance of an ensemble of nuclei excited by a free electron laser

Aleksandr I. Chumakov[1,2*], Alfred Q. R. Baron[3*], Ilya Sergueev[4], Cornelius Strohm[4], Olaf Leupold[4], Yuri Shvyd'ko[5], Gennadi V. Smirnov[2], Rudolf Rüffer[1], Yuichi Inubushi[6], Makina Yabashi[3], Kensuke Tono[6], Togo Kudo[3] and Tetsuya Ishikawa[3]

Nature Physics 14, 261 (2018)

First time-resolved measurement of multi-photon NRS





Experiment performed at SACLA, Japan

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Multiphoton Collective Lambshift in Nuclear Resonant Scattering

Goal: Study the spectral dependence of multiphoton NRS

European XFEL Proposal #2778 **17 – 23 May 2022**

The Team

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Freiburg

Dominik Lentrodt

Theory, data analysis, evaluation

European XFEL

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Jörg Hallmann et al.

The Data analysis group
Luca Gelisio
James Wrigley et al.

The HXRSS group
Gianluca Geloni et al.
Shan Liu (DESY)

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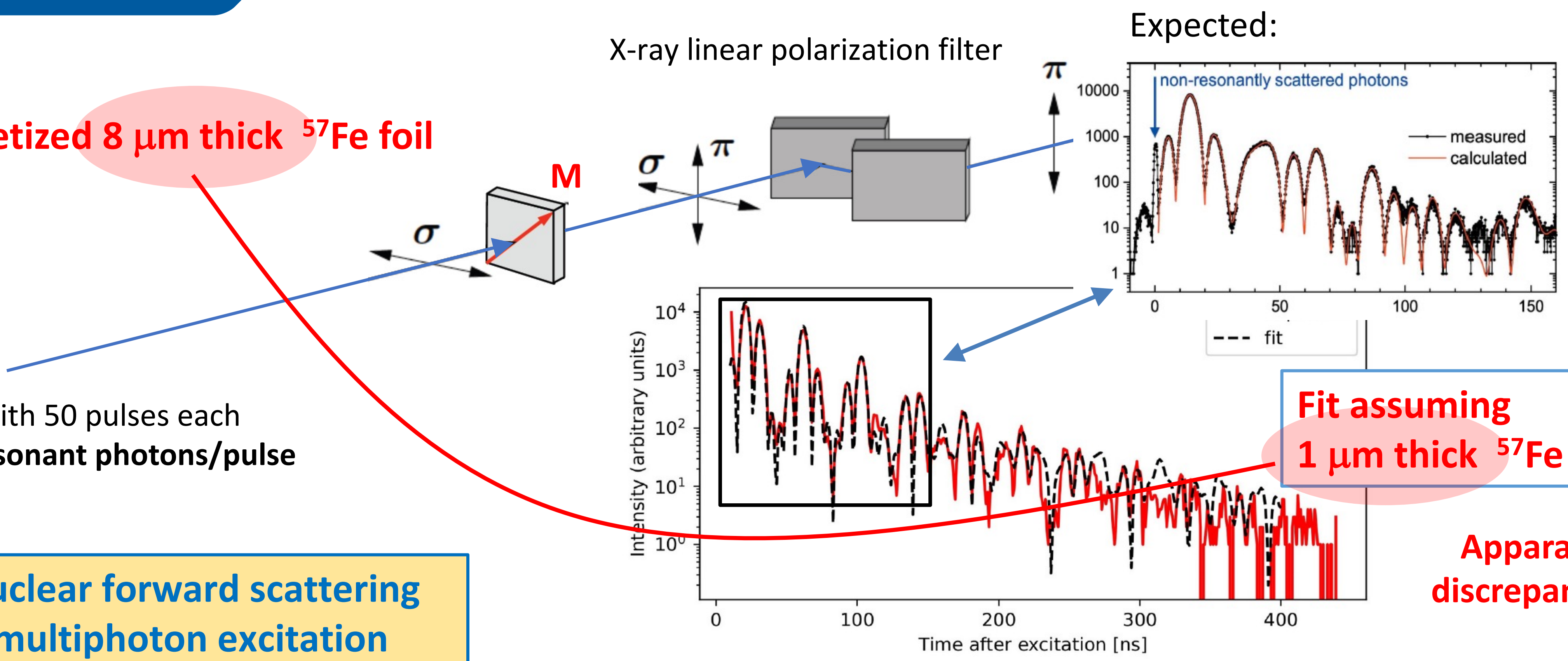
First nuclear forward scattering experiment at the European XFEL

Magnetized 8 μm thick ^{57}Fe foil



Fit assuming
1 μm thick ^{57}Fe foil

Apparant discrepancy !

10 pulse trains with 50 pulses each
~ 150 nuclear resonant photons/pulse

First nuclear forward scattering under multiphoton excitation conditions

Time evolution of nuclear resonant scattering under multi-photon excitation conditions

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UNI FREIBURG
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Mössbauer Isotopes with $\Gamma_0/E_0 < 3 \times 10^{-13}$

Isotope	Energy E_0 (eV)	Linewidth Γ_0 (eV)	Γ_0/E	natural lifetime	natural abundance	conversion coefficient
^{229}Th	8.3	$\sim 10^{-19}$	$\sim 10^{-20}$	$10^2 \dots 10^3$ s	N/A	>2000 (?)
^{57}Fe	14413	4.7×10^{-9}	3×10^{-13}	141 ns	2.4 %	8.6
^{73}Ge	13260	1.5×10^{-10}	1×10^{-14}	2.0 μs	7.8 %	1100
^{181}Ta	6230	6.7×10^{-11}	1×10^{-14}	4.7 μs	100 %	46
^{67}Zn	93319	5.1×10^{-11}	5×10^{-16}	6.4 μs	4.1%	0.9
^{45}Sc	12400	1.4×10^{-15}	1.1×10^{-19}	0.46 s	100 %	>400
^{109}Ag	88000	1.1×10^{-17}	1.2×10^{-22}	27.6 s	48 %	26

^{45}Sc: Main problem for conventional Mössbauer measurements:
No convenient radioactive source available.

Besides that, ^{45}Sc appears to be very attractive:

- High Lamb-Mössbauer factor (~0.7) due to low photon energy (12.4 keV)
- Isotopic abundance 100%
- Very high Q factor: $E_0/\Gamma_0 \sim 10^{19}$

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History

Resonant excitation of ^{45}Sc using 12.4-keV x-rays from accelerator-based x-ray sources of high spectral brightness was predicted to be feasible:

Yu. Shvyd'ko and G.V. Smirnov
On the direct measurement of nuclear γ-resonance parameters of long-lived (> 1s) isomers
Nucl. Instrum. Meth. B 51, 452-457 (1990)

Attempts to detect the ^{45}Sc resonance at 3rd generation SR sources were so far unsuccessful (spectral brightness too low).

4. Numerical estimates for ^{109}Ag and ^{45}Sc

As a second example we shall consider the γ-resonance of ^{45}Sc nuclei ($E_0 = 12.4$ keV, $\tau_0 = 0.46$ s) [17]. In this case the observation of the γ-resonance is difficult, not only due to its small energy width, but also due to the absence of a suitable parent radioactive nucleus that could be used as a resonant γ-radiation source. Therefore we shall assume the ^{45}Sc nuclei to be irradiated by synchrotron radiation with a white spectrum. According to ref. [18], by using undulators on electron–positron storage rings of the new generation, it will be possible to achieve a γ-quanta flux with a spectral density of about 10^{13} quanta/(s eV) (in the energy range of 12 keV). This means that in this case $N_{\Gamma_0} \approx 0.01$ quanta/s will fall in the band of natural width of γ-resonance in ^{45}Sc. The

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The ^{45}Sc collaboration

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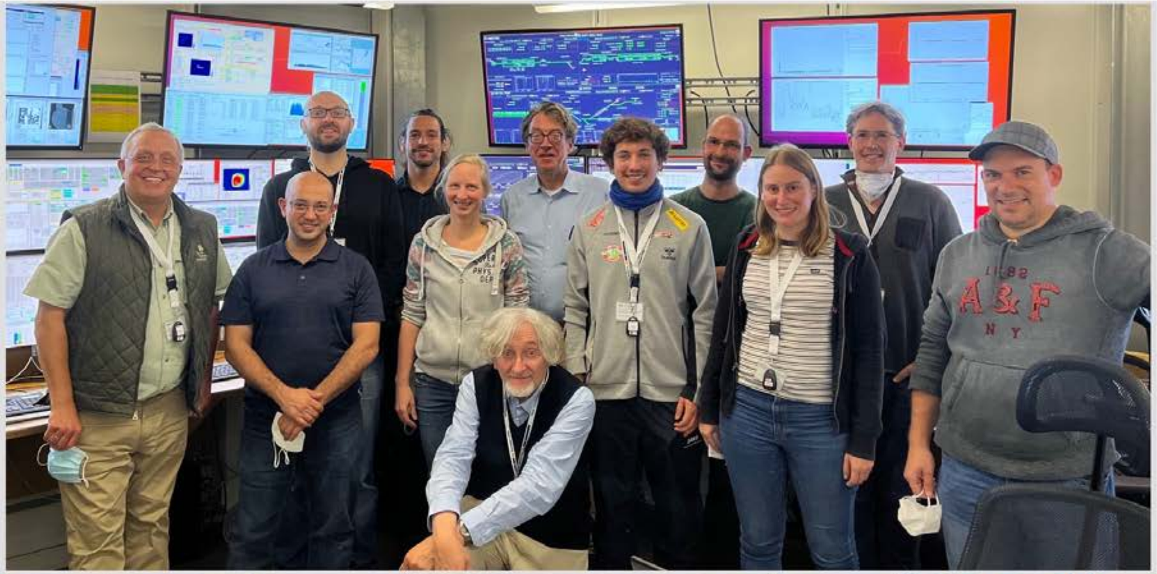
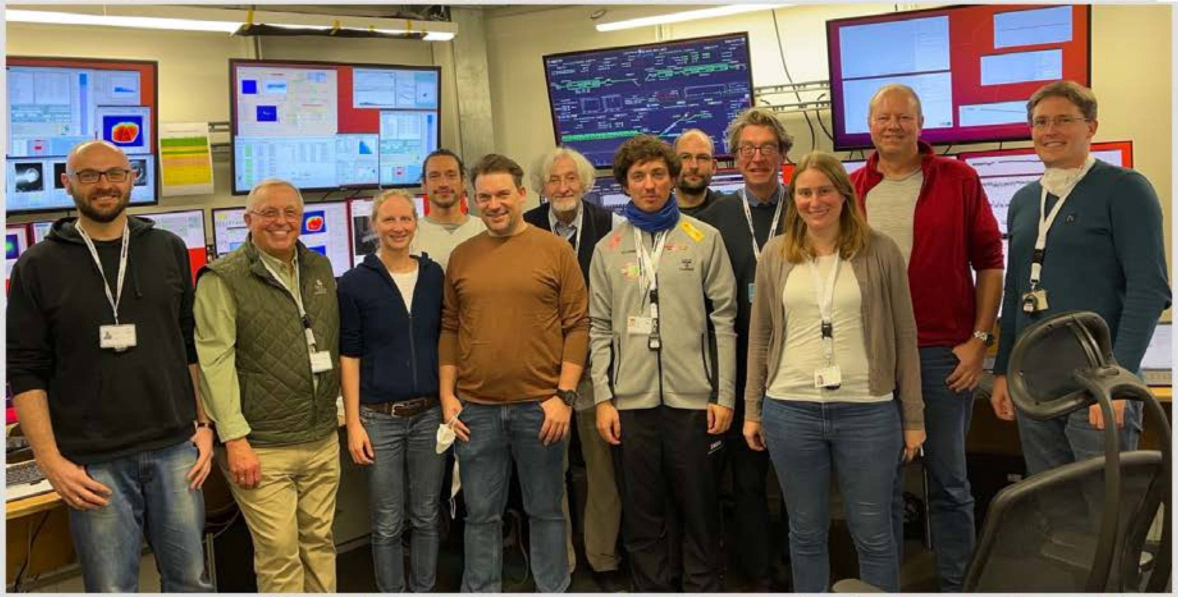




The 12.4 keV nuclear resonance of ^{45}Sc : Perspective

^{45}Sc

E_0 = 12.4 keV

Γ_0 = 1.4 feV

$Q = E_0/\Gamma_0 = 10^{19}$

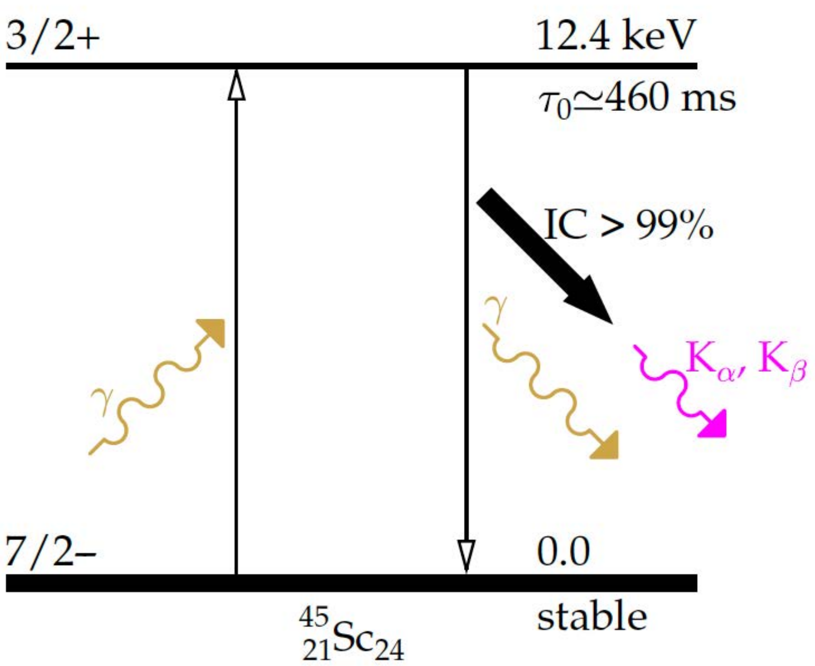
λ = 0.1 nm

τ_0 = 460 ms

(1) Exceeding the Q-factors of
   (a) the majority of optical transitions in modern atomic clocks
   (b) all measurable Mössbauer resonances by orders of magnitude.

(2) Applications in extreme metrology, e.g.,
   (a) development of a nuclear clock superior to the state-of-art atomic optical clocks
   (b) testing of fundamental principles of physics.

^{45}Sc level diagram



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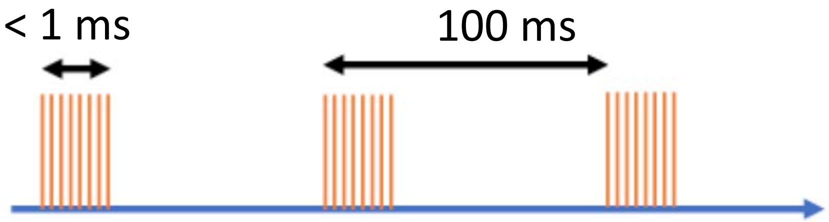
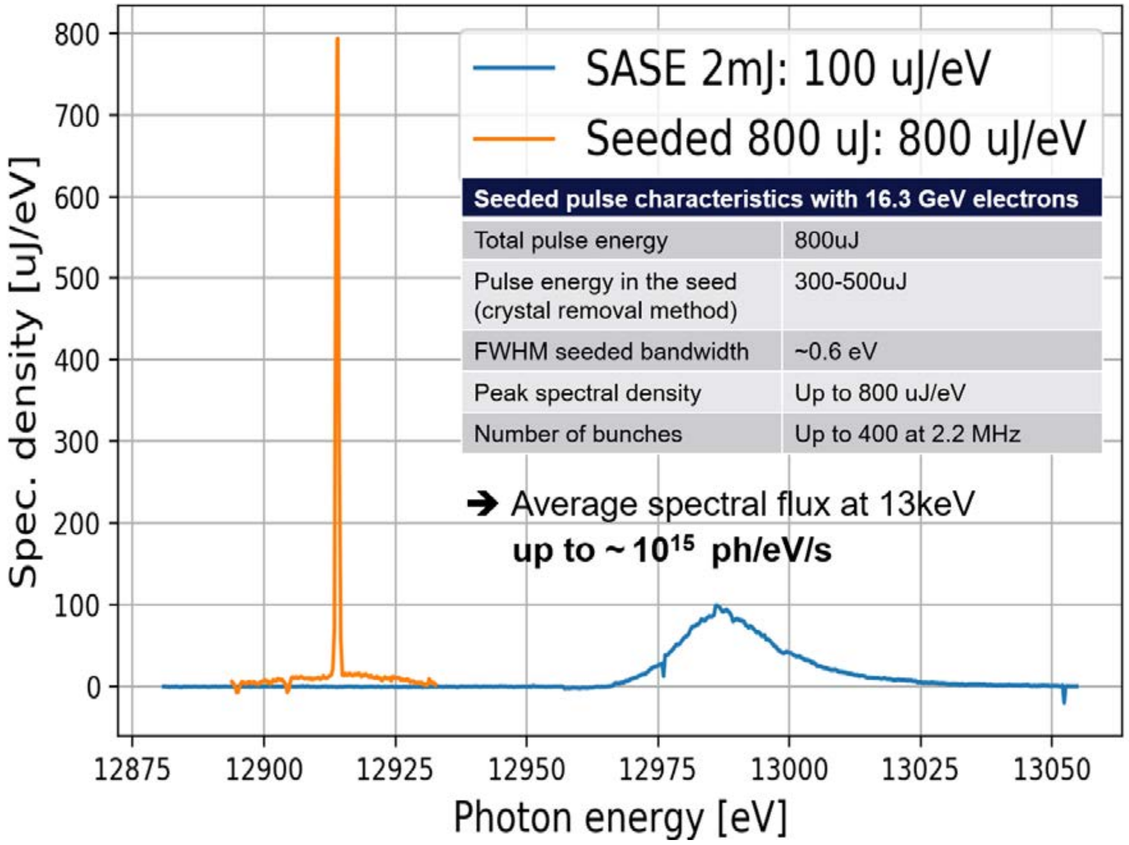
Why European XFEL ?

(1) Hard x-ray self-seeded (HXRSS), high rep rate (HRR) XFELs may provide average spectral flux > 1000 than ESRF, APS, SPring-8, or PETRA-III
O. Chubar et al., JSR 23 410 (2016)

(2) The European XFEL is the 1st HXRSS HRR XFEL
W. Decking et al., Nat. Photonics 14, 391 (2020)

(3) The European XFEL time structure is ideally suited for the detection of the ^{45}Sc-resonance: sub-ms pulse trains with a 100-ms dark time.





Data and graphics courtesy Gianluca Geloni

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Details about the ^{45}Sc experiment to be inserted here after publication

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Poster #68

Miriam Gerharz et al.

Direct x-ray-excitation of the ultra-narrow nuclear resonance of ^{45}Sc

Thank you very much for your attention !

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