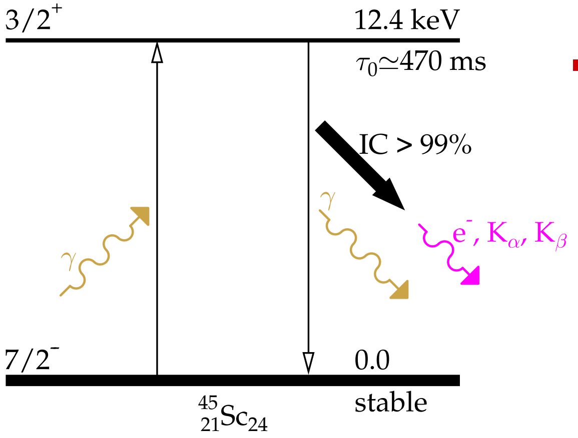
Nuclear Clock Isomer Scandium-45 driven by X-ray Free-Electron Lasers (XFEL)

Yuri Shvyd'ko

First Hard X-ray Self-seeding Workshop (HXRSS 2025)
Pohang, South Korea, October 15-17, 2025

Brief Introduction to Scandium-45

- ^{45}Sc nuclei exhibit an extraordinary narrowband
$$\Gamma_0 = \hbar/\tau_0 = 1.4 \times 10^{-15} \text{ eV} \ (0.33 \text{ Hz})$$
x-ray resonance transition with energy
$$E \simeq 12.4 \text{ keV}$$
and with exceptionally large quality factor
$$Q_0 = E/\Gamma_0 \simeq 10^{19}.$$



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- Despite its extremely sharp spectral profile, it has become possible to drive and study the x-ray quantum transition in ^{45}Sc using x-ray sources with the highest spectral brightness—x-ray free-electron lasers (XFELs).

Yu. Shvyd'ko, R. Röhlsberger, O. Kocharovskaya, J. Evers, et al. Nature 622 (2023) 471

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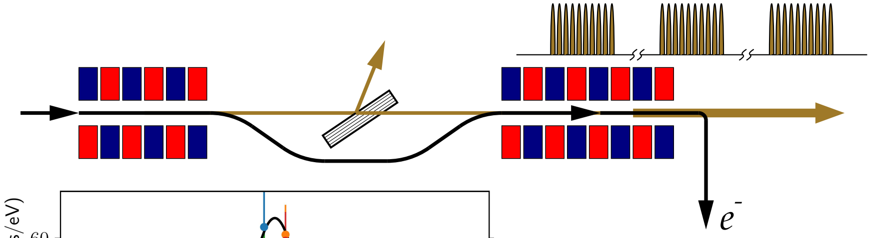

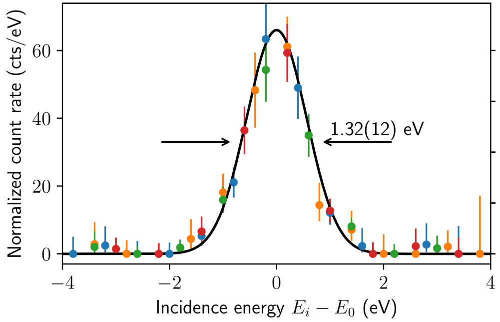

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- This advancement opens up new opportunities for exquisite applications of ^{45}Sc, such as: nuclear frequency standard, nuclear clocks, extreme x-ray metrology, ultra-high-resolution spectroscopy, etc.

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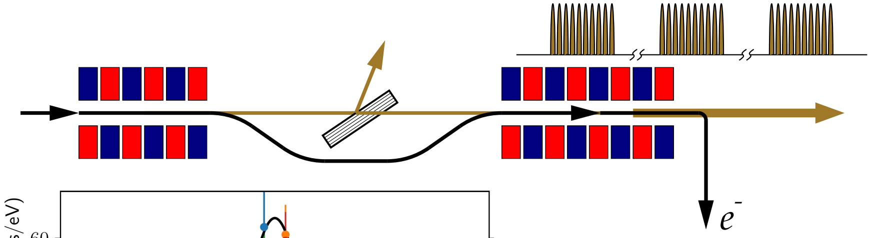

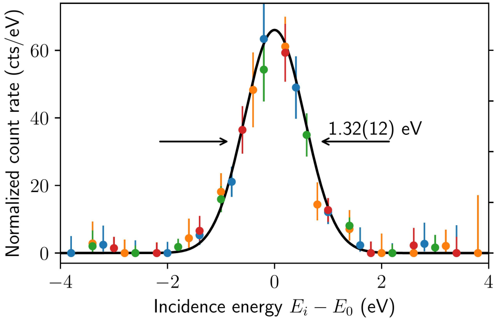

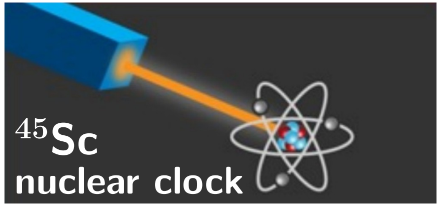

Content

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- X-ray free-electron lasers (XFELs).
- Resonant x-ray excitation and experimental studies of the ^{45}Sc resonance.
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Article

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Resonant X-ray excitation of the nuclear clock isomer ^{45}Sc

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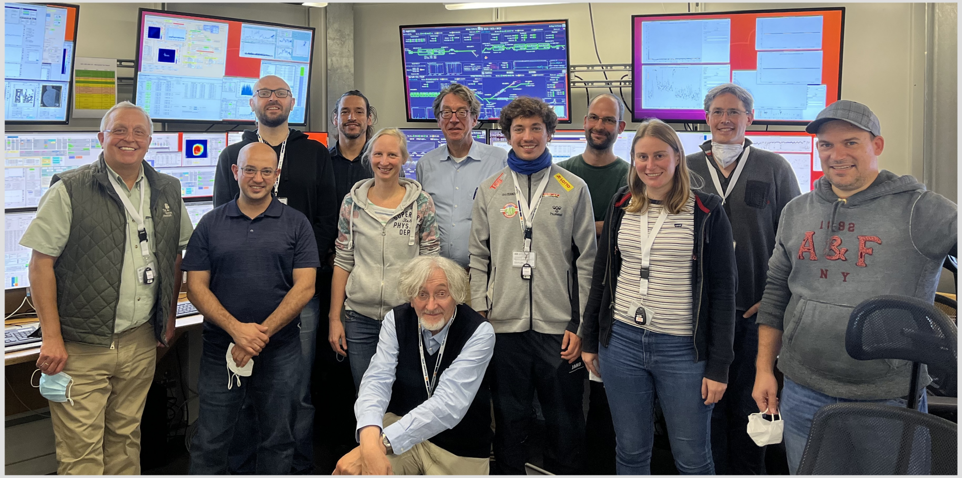

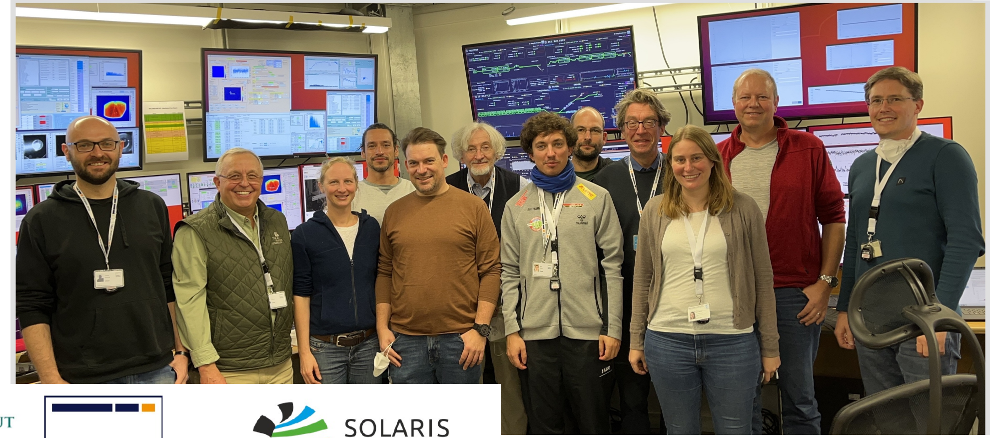

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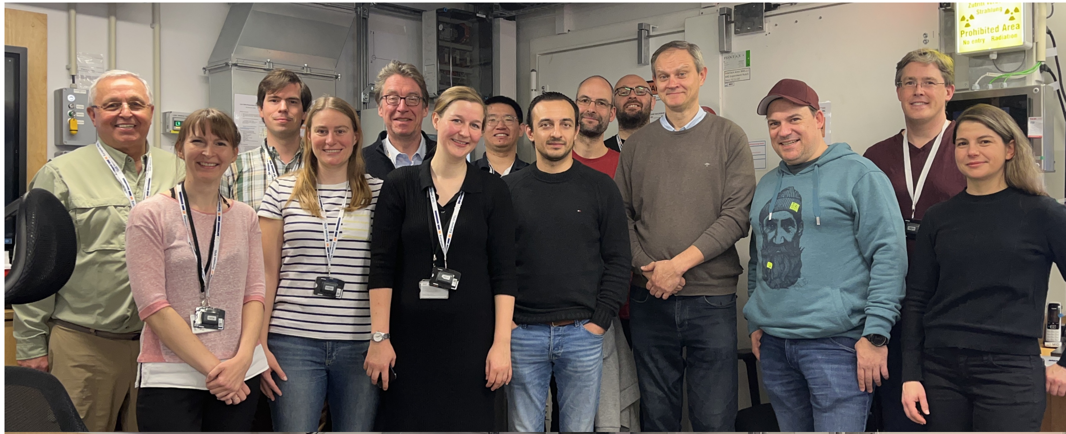

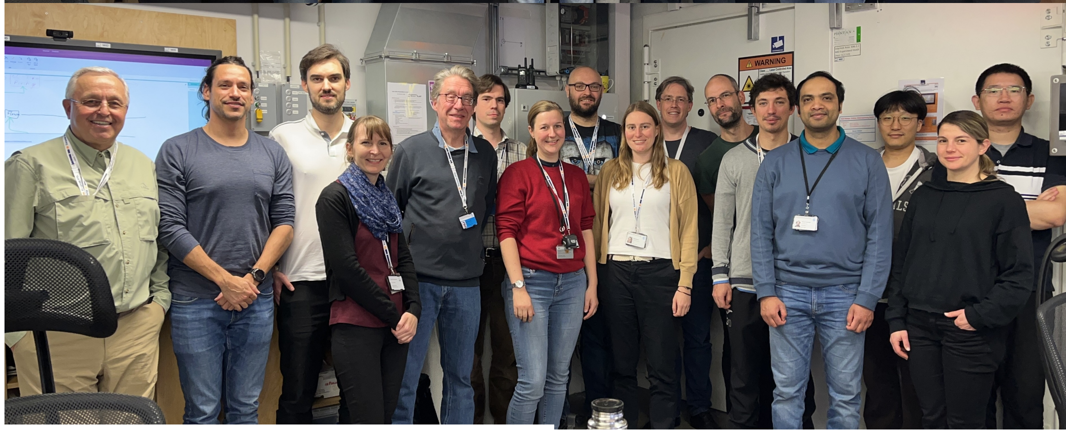

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Frequency Standards Enable Time Measurements

Oscillators with stable frequencies and large quality factors – frequency standards – determine our ability to keep track of time. Atomic clocks are presently our most precise measurement devices.

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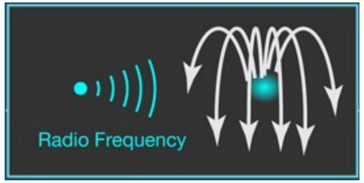
10^{-2}

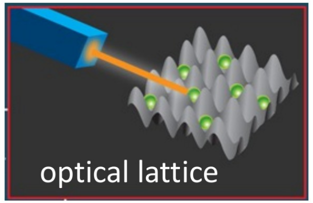
10^{-4}

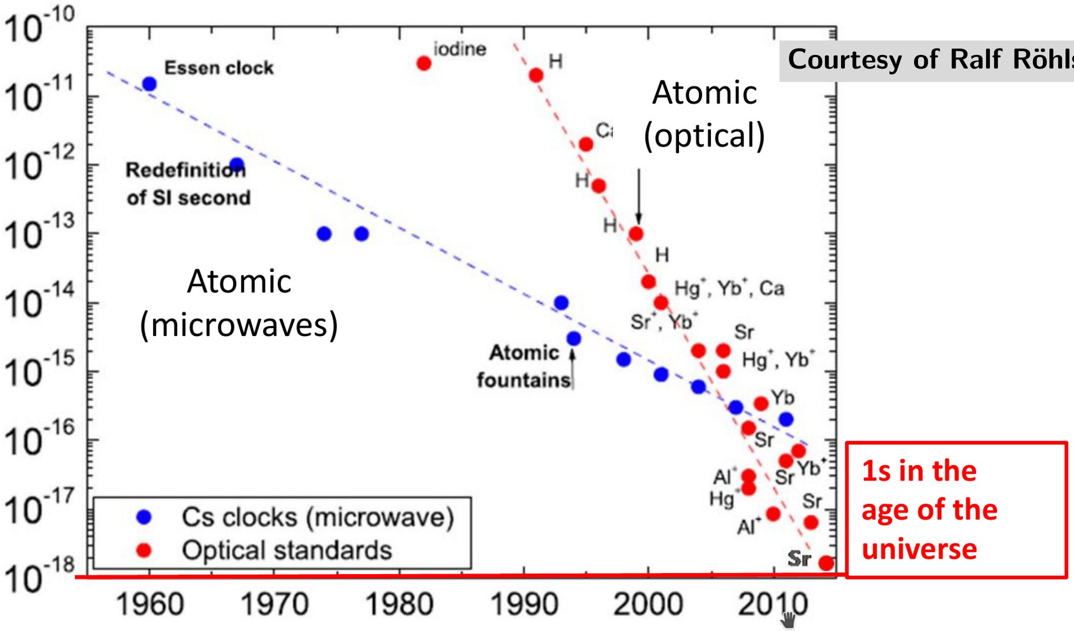
solid state

10^{-6}

fractional uncertainty







Courtesy of Ralf Röhlsberger

A. Ludlow, M. Boyd, J. Ye, E. Peik, and P.O. Schmidt "Optical atomic clocks" Rev. Mod. Phys 87 (2015) 637

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Search for more accurate, stable, and convenient reference oscillators is ongoing.

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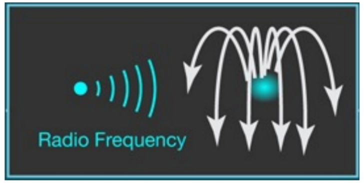

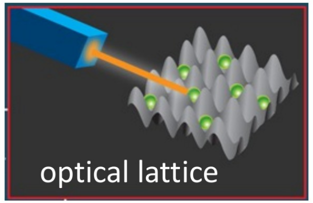

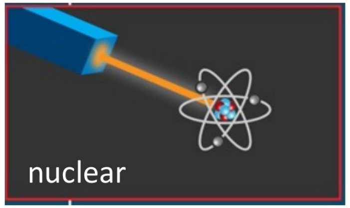

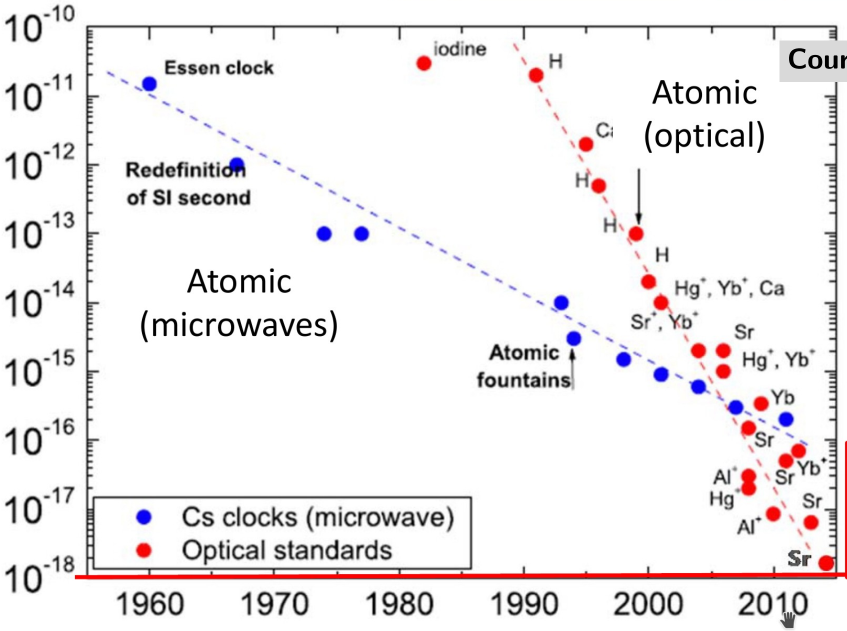

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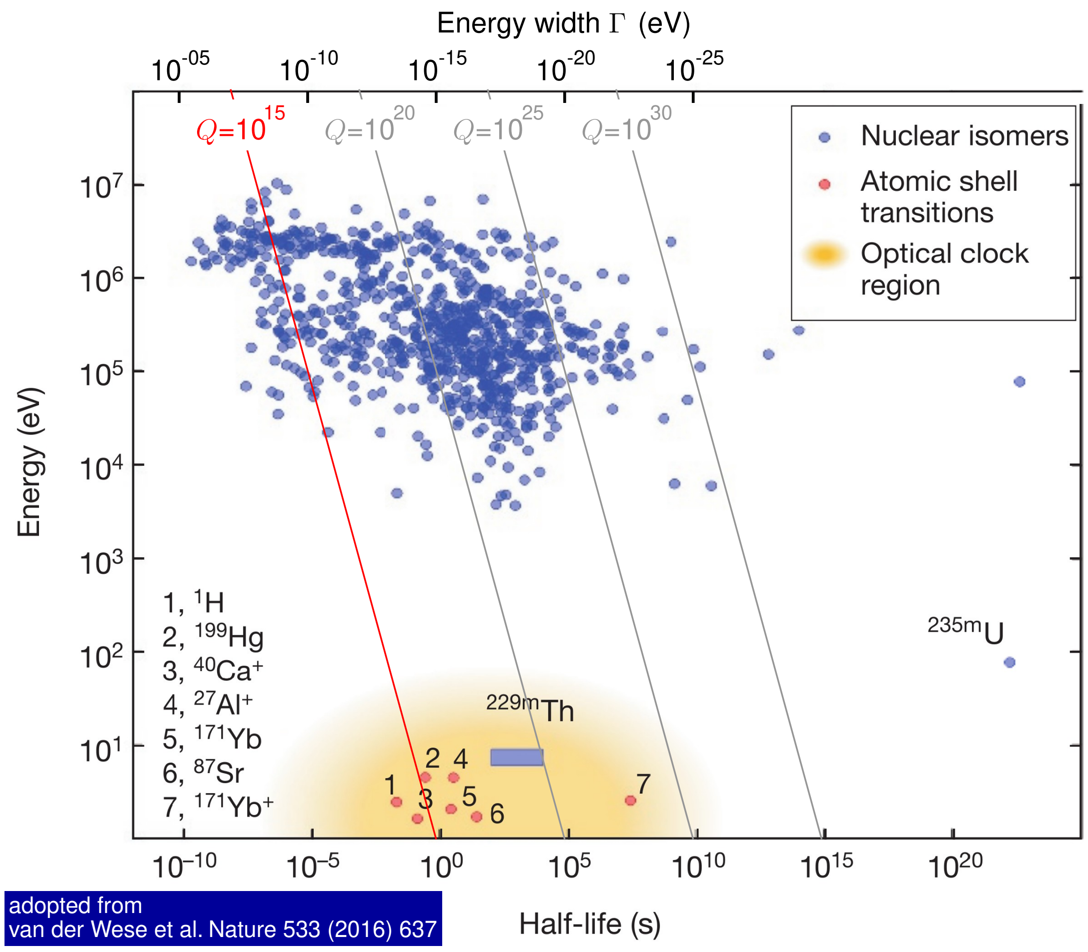
Nuclear vs. Atomic Resonances

1. **Higher Q-factors.** $Q = E/\Gamma$
2. **Higher frequency**: a higher-frequency transition offers greater stability for simple statistical reasons (fluctuations are averaged over more cycles).
3. **Insensitivity to environmental effects.** Due to its small size, small magnetic moments, and the shielding effect of the surrounding electrons, an atomic nucleus is much less sensitive to ambient electromagnetic fields than is an electron in an orbital.
4. **Macroscopic number of atoms can be used,** because of the insensitivity to ambient fields, it is not necessary to have the clock atoms well-separated in a dilute gas or in ion traps a very low temperatures.
5. **Mössbauer effect with nuclei in solids** would allow solid-state amount of nuclei to exhibit narrow spectral resonance line at moderately-low temperatures.

E Peik, et al."Nuclear clocks for testing fundamental physics" Quantum Sci. Technol. 6 (2021) 034002



adopted from
van der Wese et al. Nature 533 (2016) 637

Nuclear Clock Isomer ^{229m}Th

$\Gamma_o^{(^{229m}\mathrm{Th})} \sim 10^{-18}$ eV (0.24 mHz)

A lot of effort was dedicated to studies of the ^{229m}Th isomer and measurements of the nuclear transition energy: 8.338(24) eV [*].

L. van der Wese et al. Nature 533 (2016) 637
"Direct detection of the ^{229m}Th nuclear clock transition"

Seiferle, B. et al. Nature 573, 243 (2019)
Energy of the ^{229m}Th nuclear clock transition.

[*] Kraemer, S. et al. Nature 617, 706 (2023)
Observation of the radiative decay of the ^{229m}Th nuclear clock isomer.

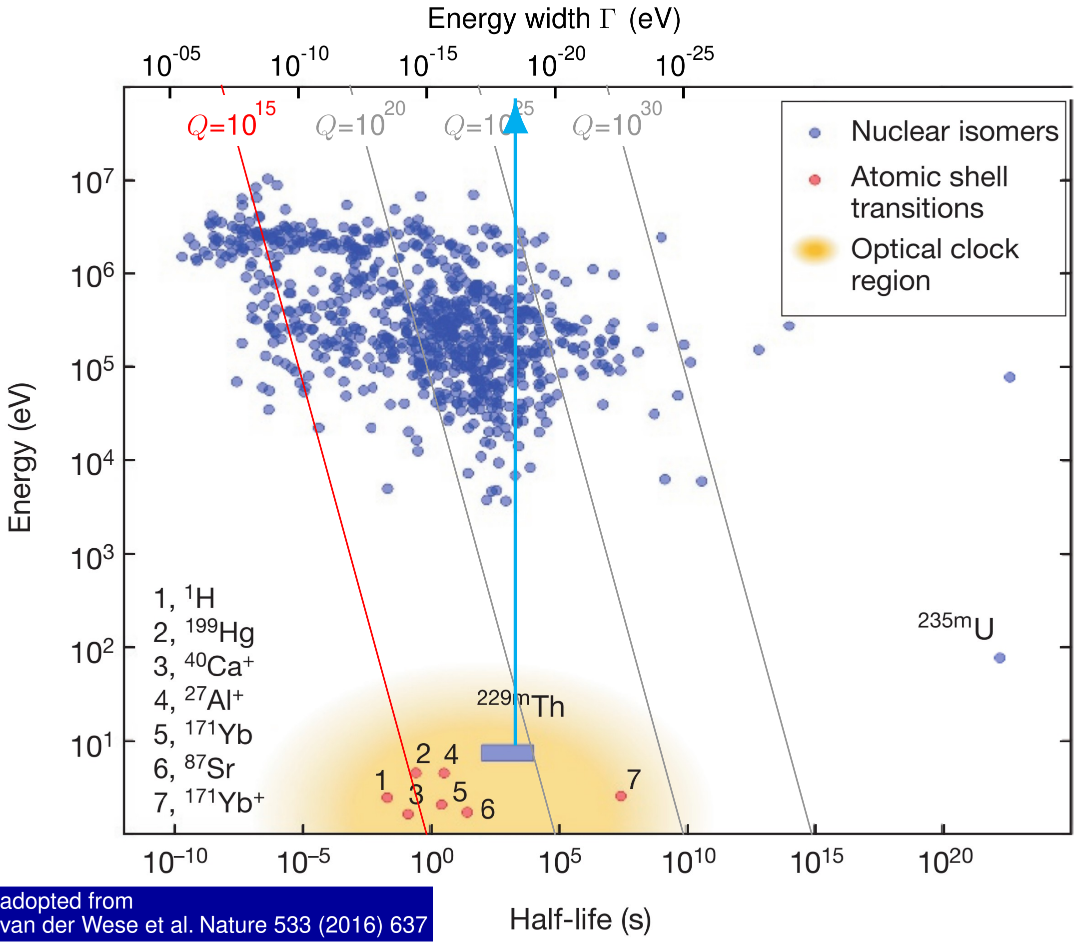
Resonant laser excitation of ^{229m}Th in a solid-state host was recently achieved:

J. Tiedau, E. Peik et al. Phys. Rev. Lett. 132 (2024) 182501

R. Elwell, E. Hudson et al. Phys. Rev. Lett. 133 (2024) 013201

A frequency ratio of the ^{229m}Th nuclear isomeric transition and the ^{87}Sr atomic clock transitions was established using a VUV comb with a 10^{-13} relative uncertainty. C. Zhang, J. Ye et al. Nature, 633 (2024)
Resonance width: $\Gamma \simeq 25$ kHz.

T. Ooi, J Ye (2025) arXiv:2507.01180



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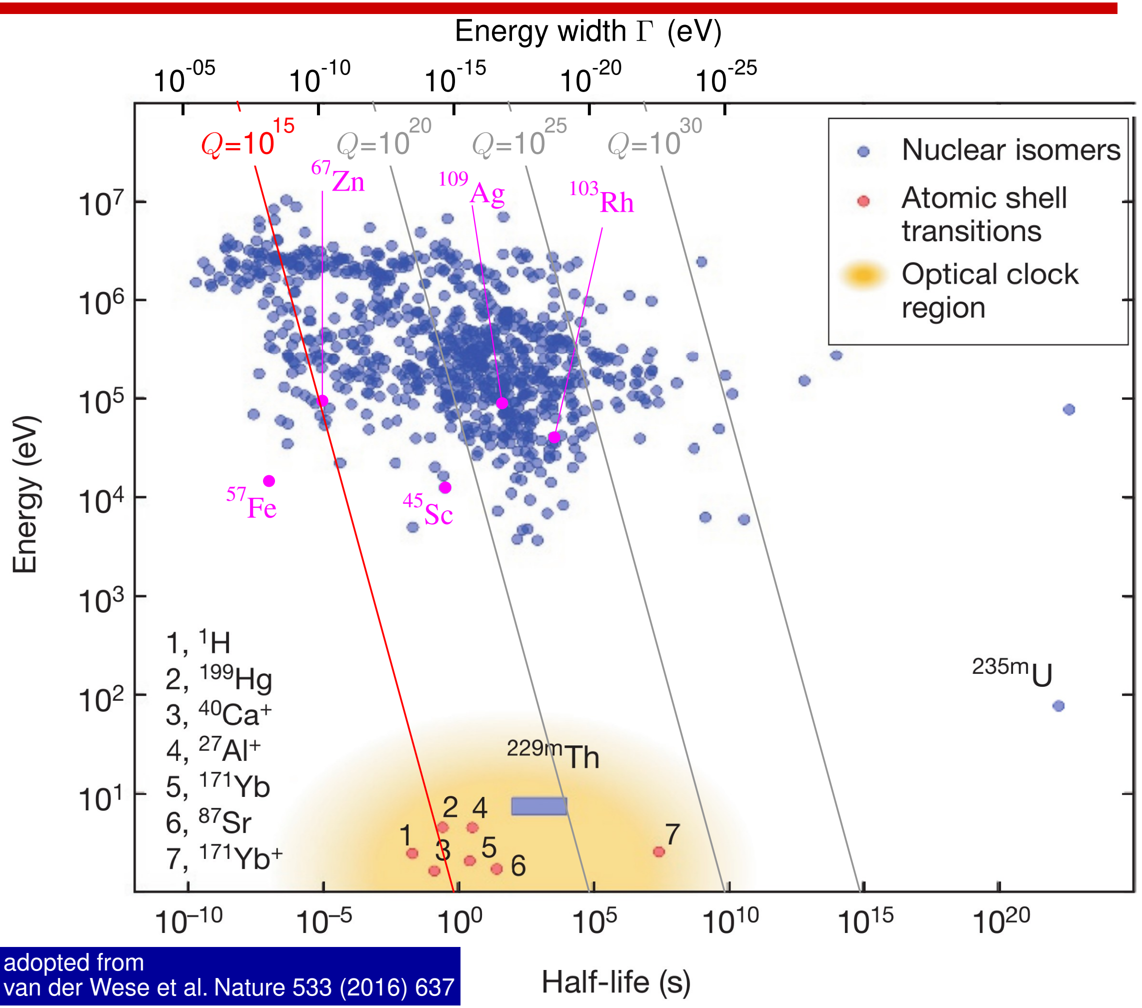
$\tau_o^{(^{229m}\mathrm{Th})} = 640$ s

Other Candidates for Nuclear Clocks

Most of the nuclear isomers (with stable ground states) have the excitation energy above 30 keV (^{67}Zn, ^{109}Ag, ^{119}Rh) and therefore cannot be driven by the modern coherent sources of electromagnetic radiation.

Fortunately, along with ^{229m}Th there is another notable exception – ^{45}Sc.



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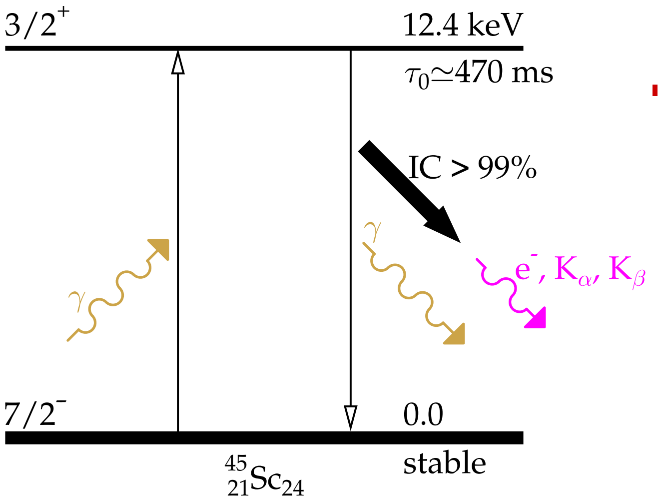
12.4-keV Nuclear Resonance of ^{45}Sc Basic Features

Excitation energy: $E = 12.40 \pm 0.05$ keV, [1,2,3,6]
excited state lifetime: $\tau = 0.47$ s, [1,5,6]
natural energy width: $\Gamma_0 = \hbar/\tau = 1.4 \times 10^{-15}$ eV=1.4 feV (0.33 Hz),
resonance quality: $Q = E/\Gamma_0 \simeq 10^{19}$,
internal conversion coefficient: $\alpha \simeq 630$, [4,5,6]
^{45}Sc natural abundance: 100%,
gravitational red shift: 1 mm/Γ_0,

room temperature Lamb-Mössbauer factor: $f_{LM} \simeq 0.8$.



[1] Holland R.E., Lynch F.J., Nystén K.E., PRL 13 (1964) 241-243.
[2] Freedman, M. S., Porter, F. T. & Wagner F.Jr, Phys. Rev. 140, B563-B565 (1965).
[3] Porter, F. T., Freedman, M. S., Wagner F., & Orlandini, K. A., Phys. Rev. 146, 774-780 (1966).
[4] Jones, K. W. and Schwarzschild, Phys. Rev. 148, 1148–1150 (1966).
[5] Blaugrund, A. E., Holland, R. E. & Lynch, F. J., Phys. Rev. 159, 926–930 (1967).

[6] Burrows, T. W., Nuclear Data Sheets 109, 171-296 (2008).

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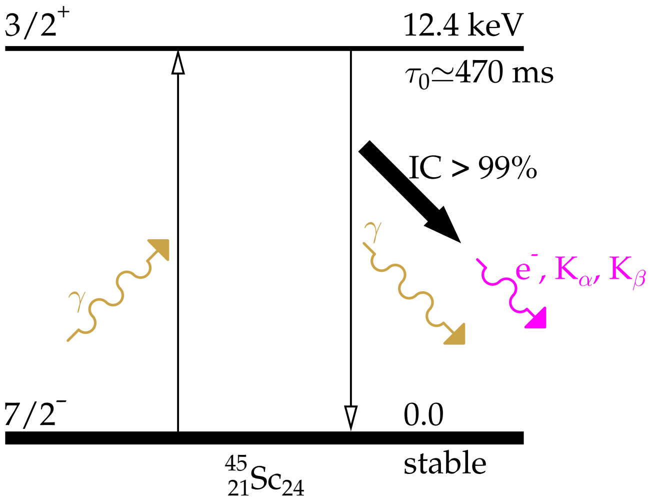

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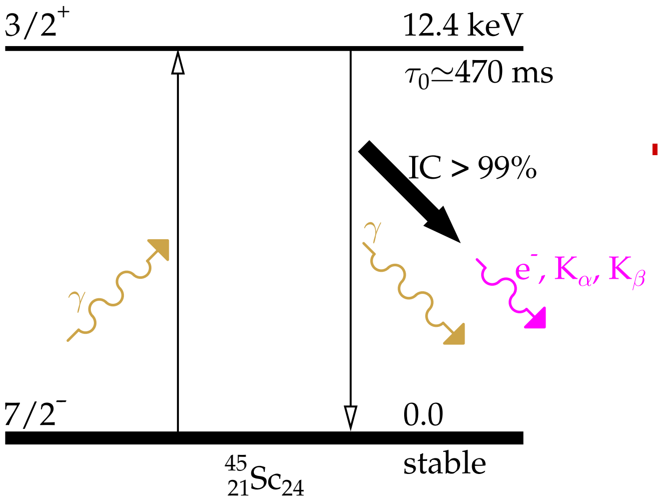

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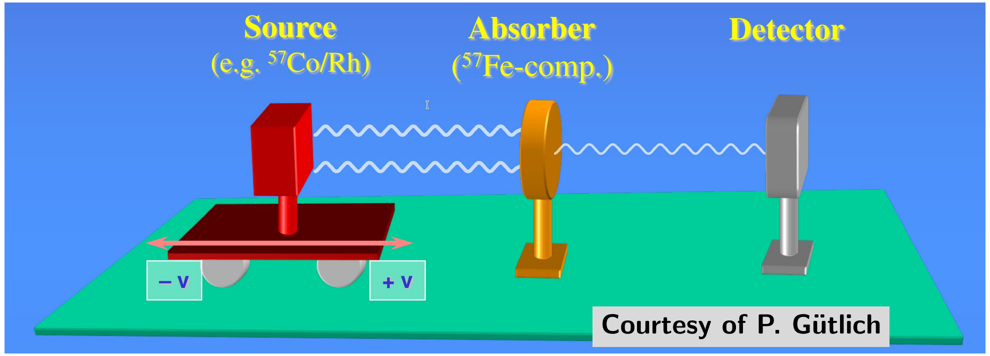
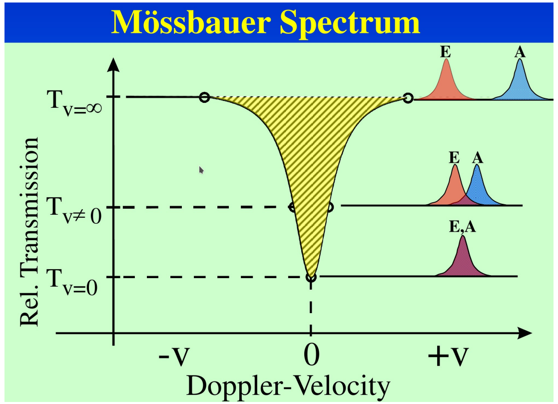
This prevented performing classical Mössbauer-type nuclear-resonance measurements.





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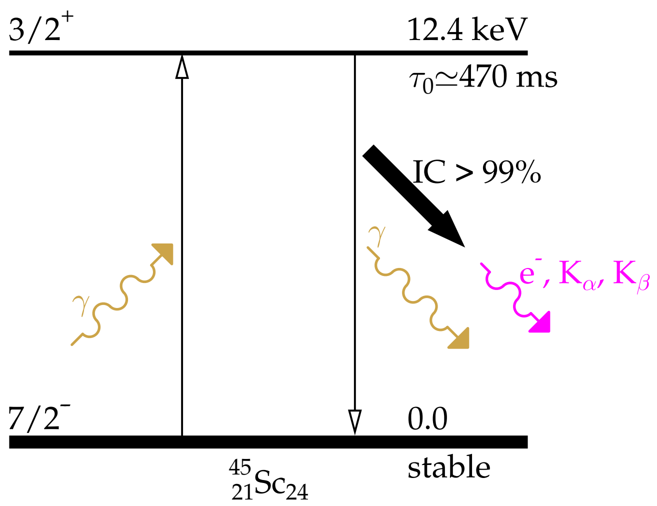

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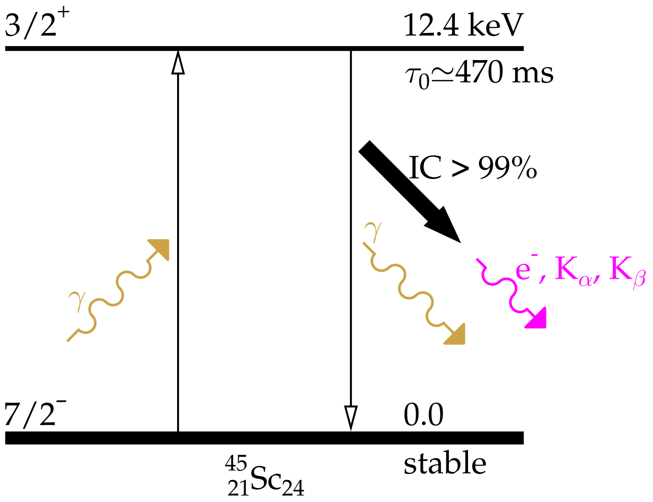

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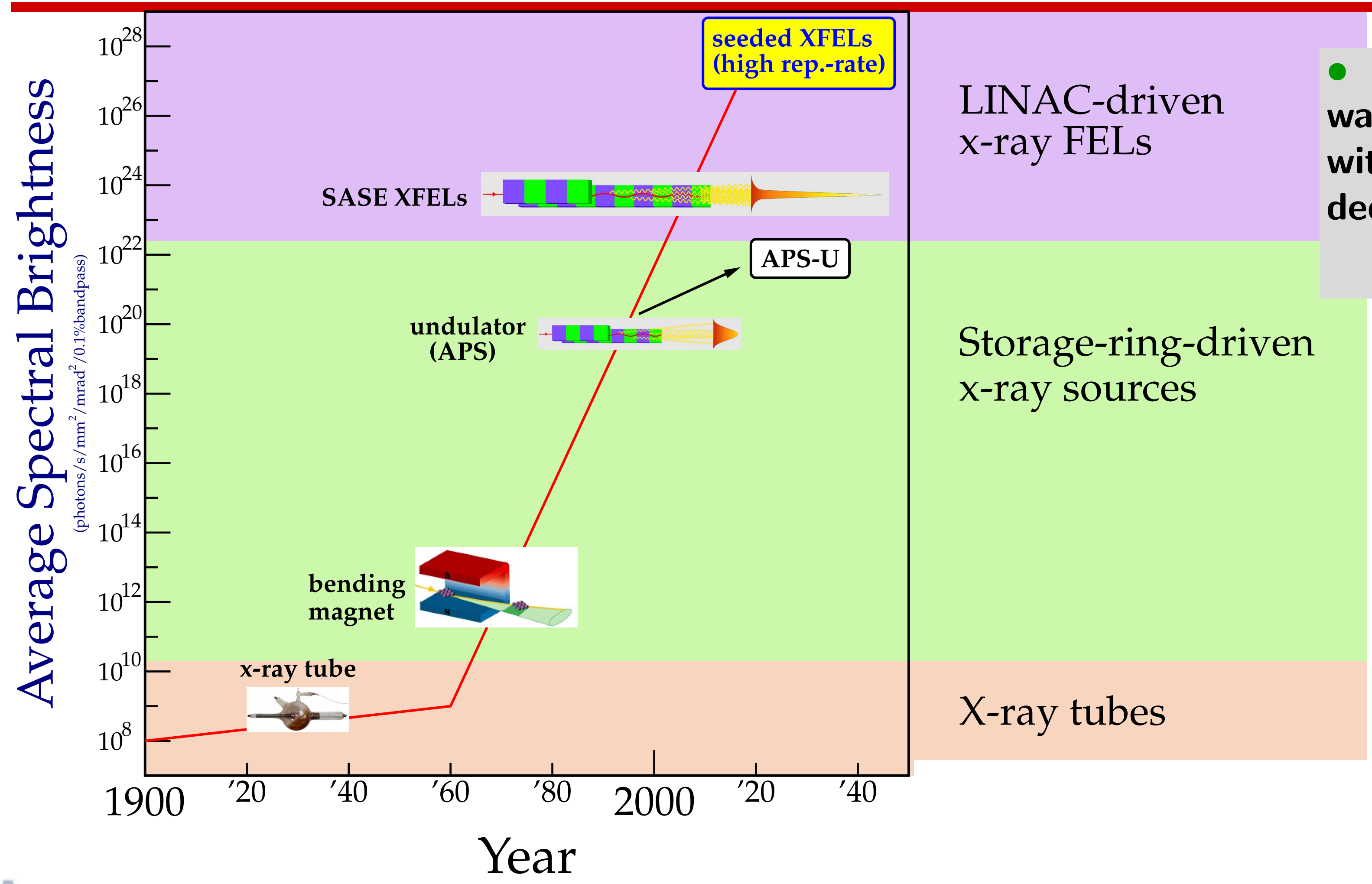
- Attempts to detect the ^{45}Sc resonance at 3rd-generation synchrotron radiation sources (ESRF, APS, SPring-8) were so far unsuccessful. Spectral flux was still low.

History of Hard X-ray Spectral Brightness



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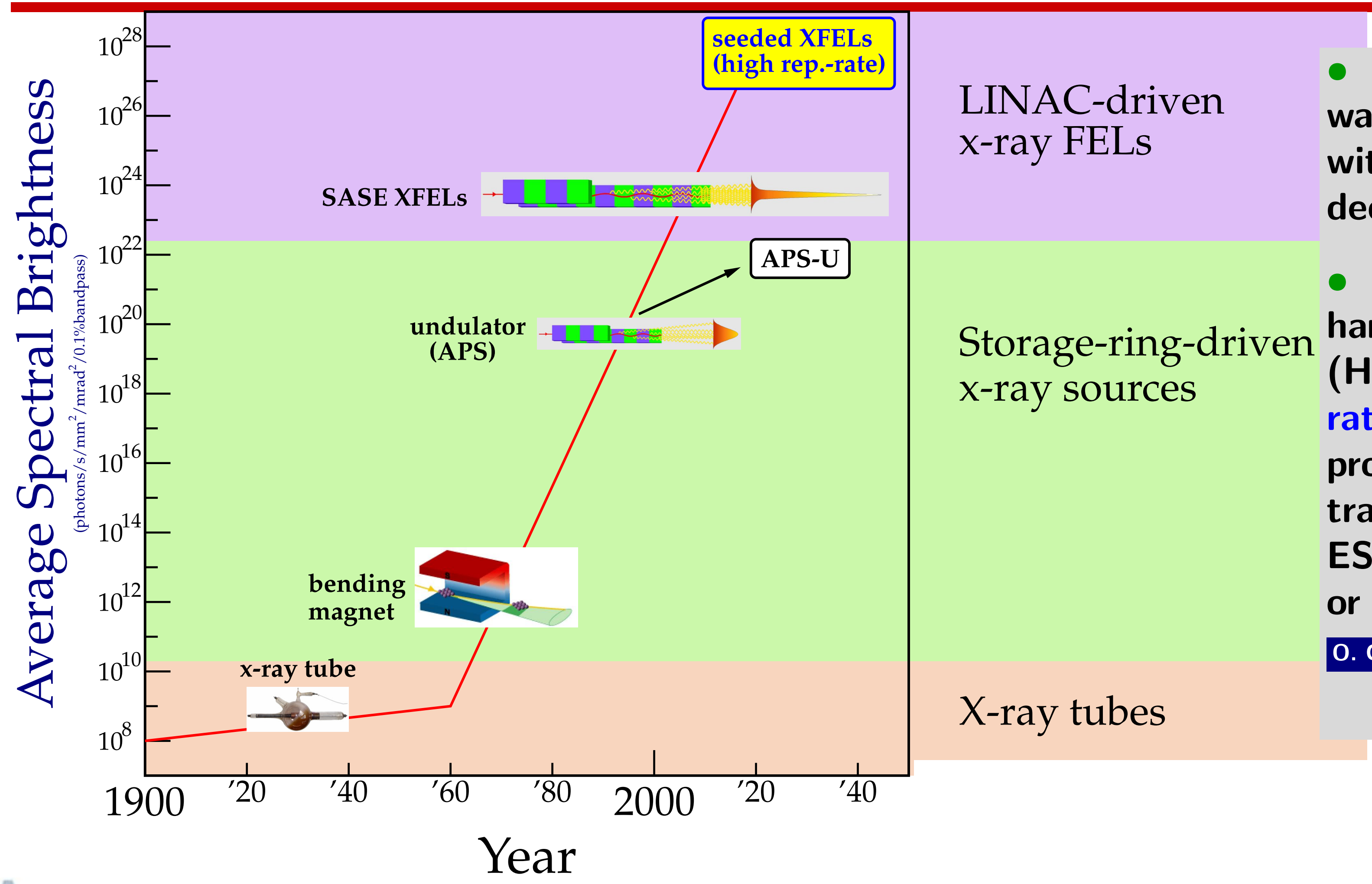

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O. Chubar et al., JSR 23 (2016) 410

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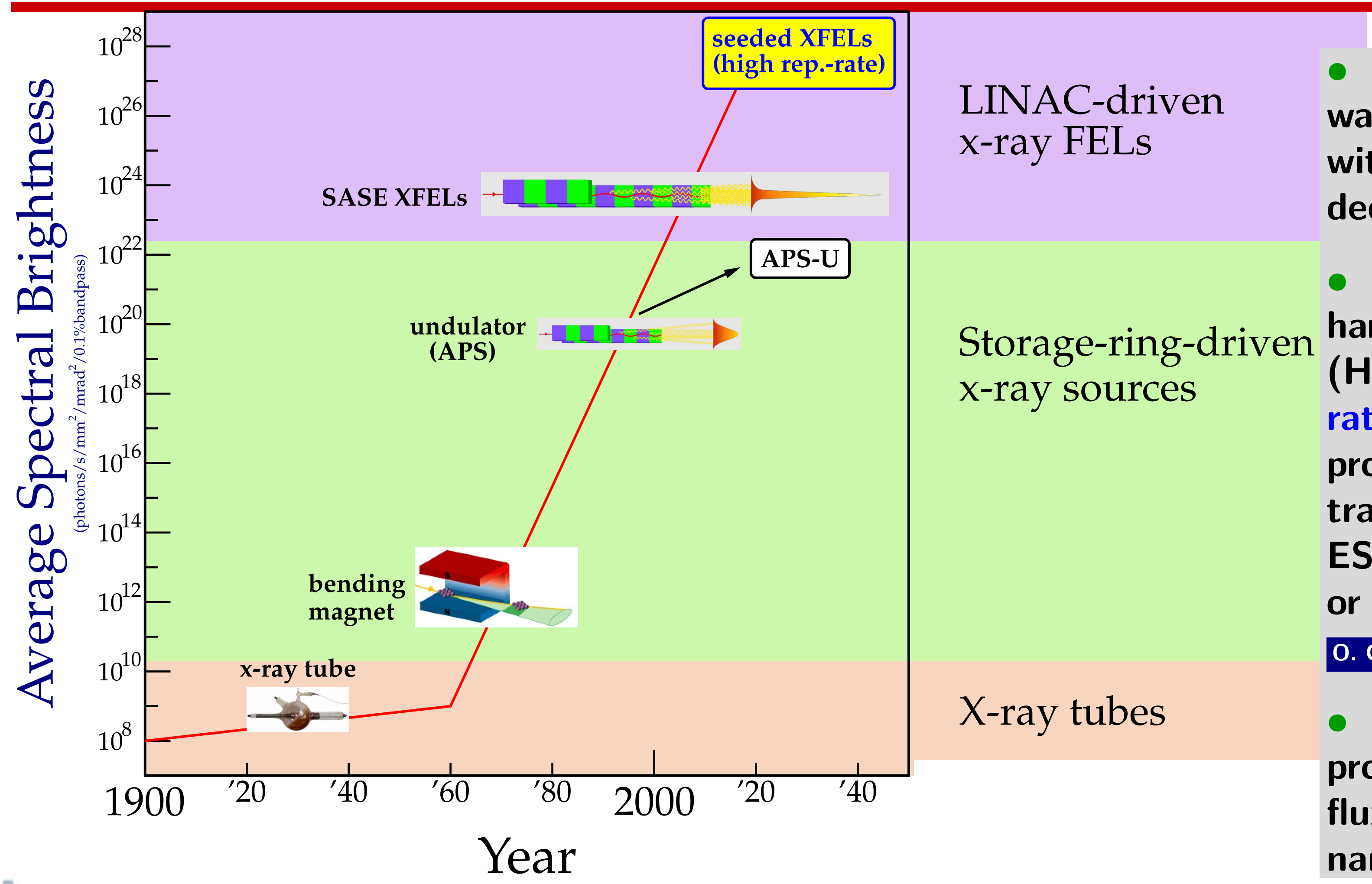

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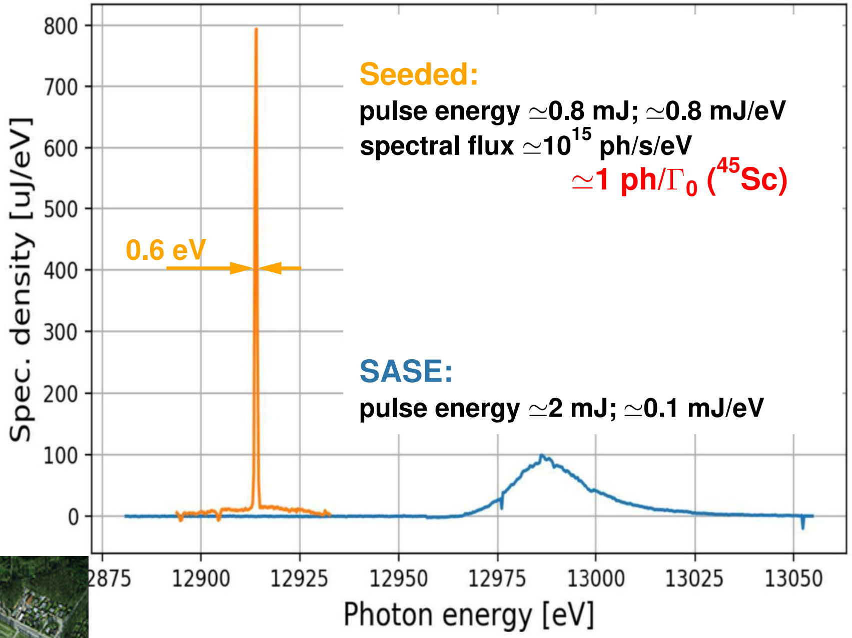
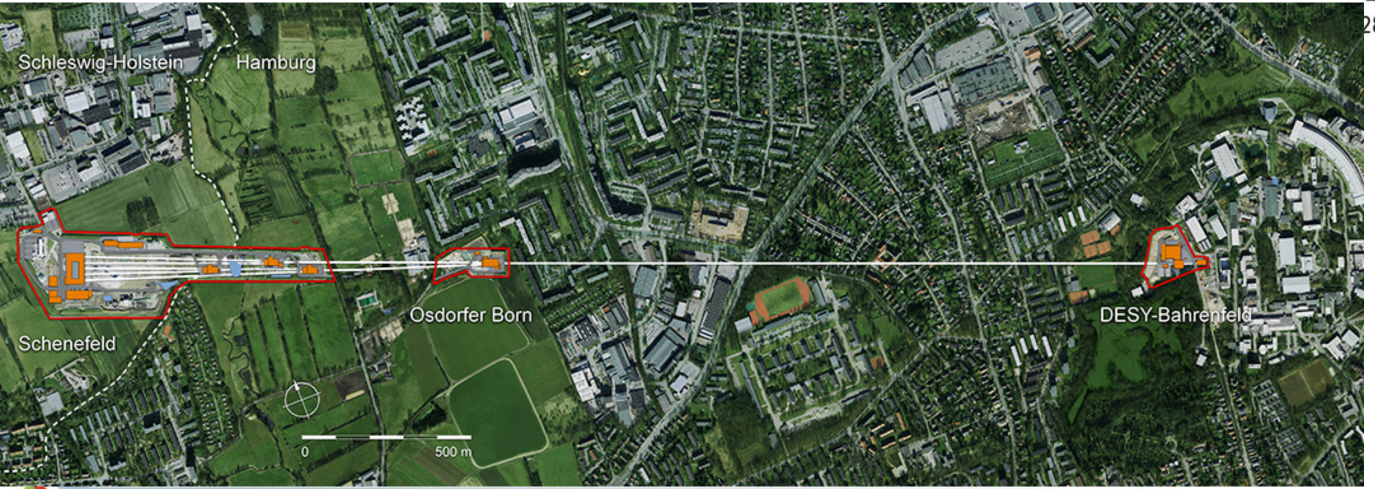
- HXRSS HRR XFELs may provide sufficient spectral flux to drive ^{45}Sc resonance.

^{45}Sc Experiment at European XFEL (Hamburg, Germany)

- European XFEL is the 1st high rep.-rate XFEL W. Decking, et al. Nat. Photonics, 14 (2020) 391.

- An average spectral flux at 13 keV of up to $\simeq 10^{15}$ ph/s/eV $\simeq 1$ ph/Γ_0(^{45}Sc) was demonstrated in self-seeded mode S. Liu et al. Nature Photonics 17 (2023) 984.





^{45}Sc Experiment at European XFEL (Hamburg, Germany)

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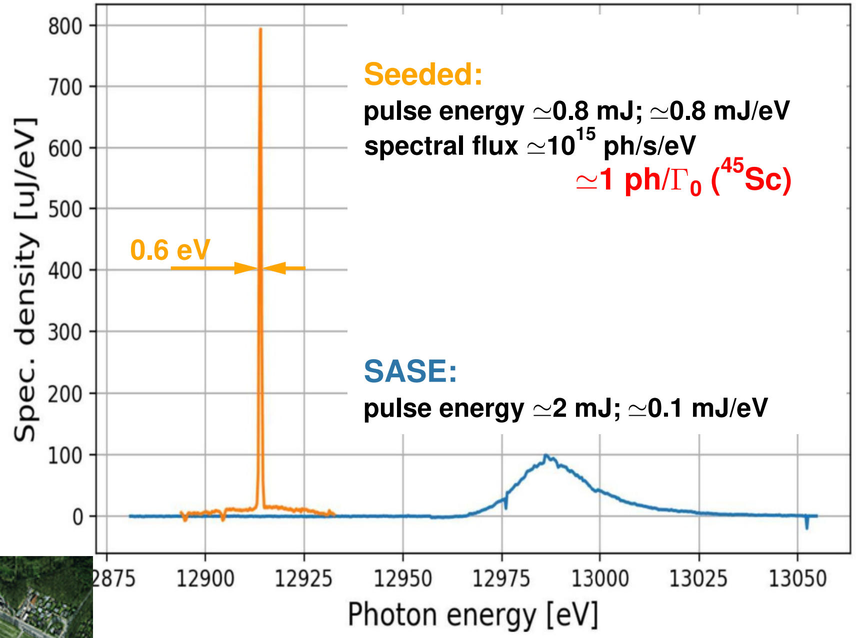
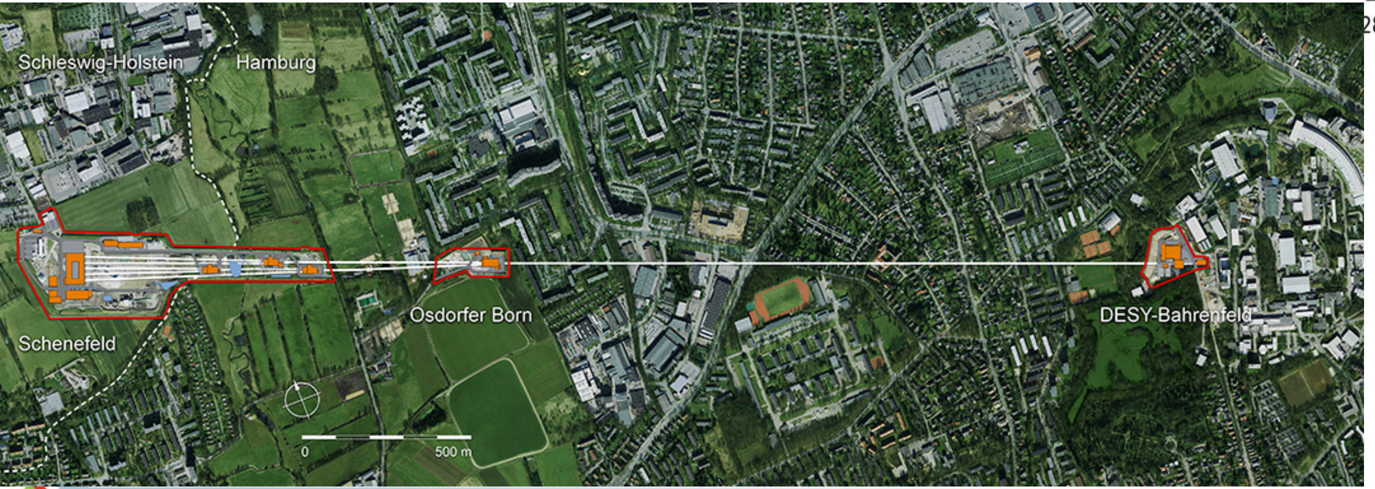
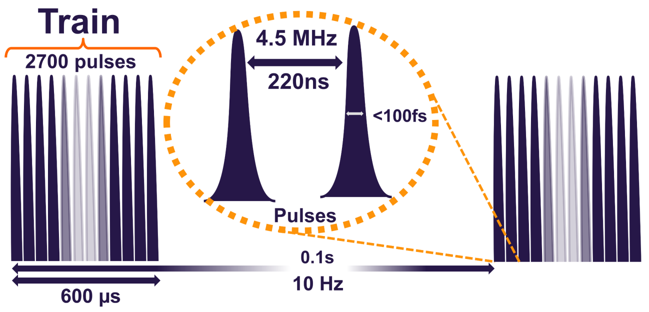
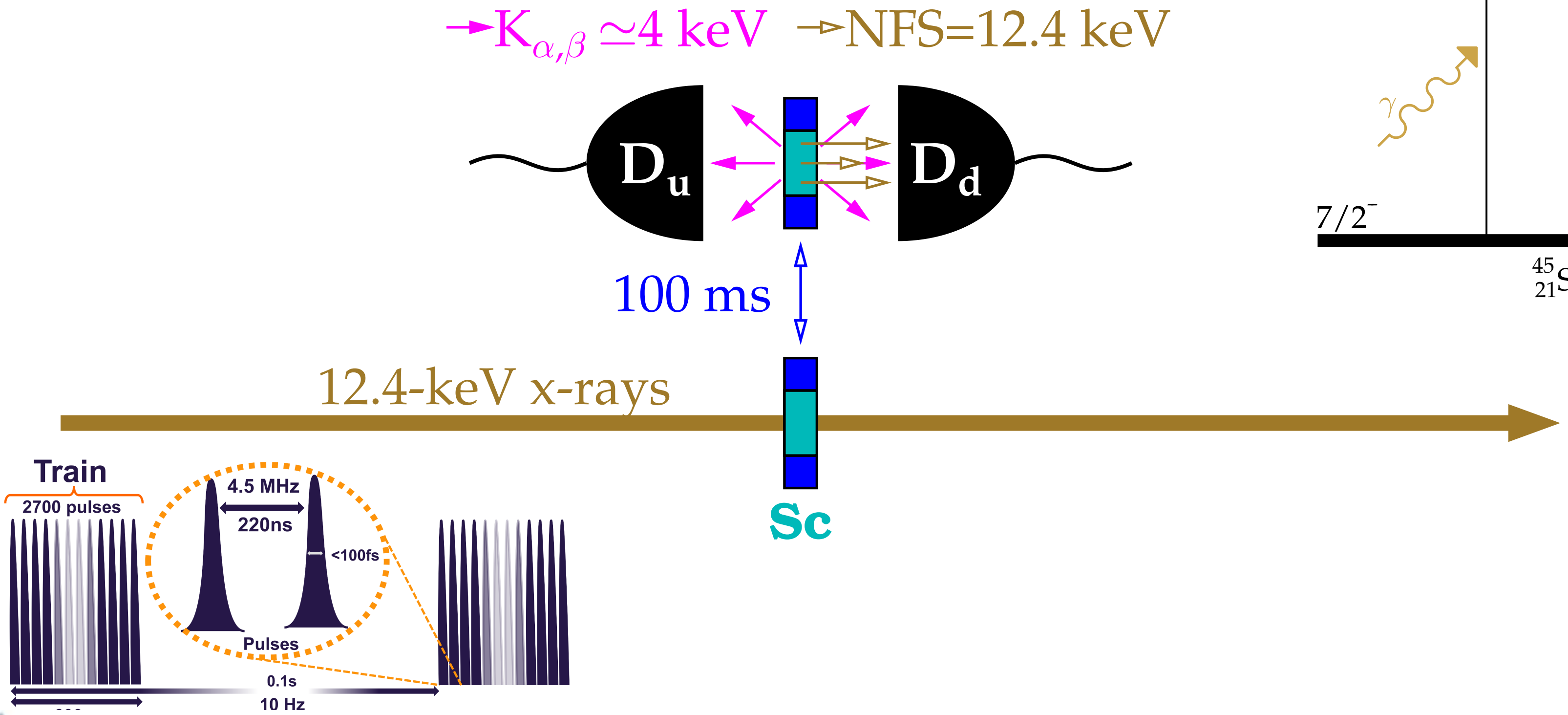
- European XFEL has a unique time structure suitable perfectly for the detection of the ^{45}Sc resonance: sub-ms pulse trains with a 100-ms dark time.

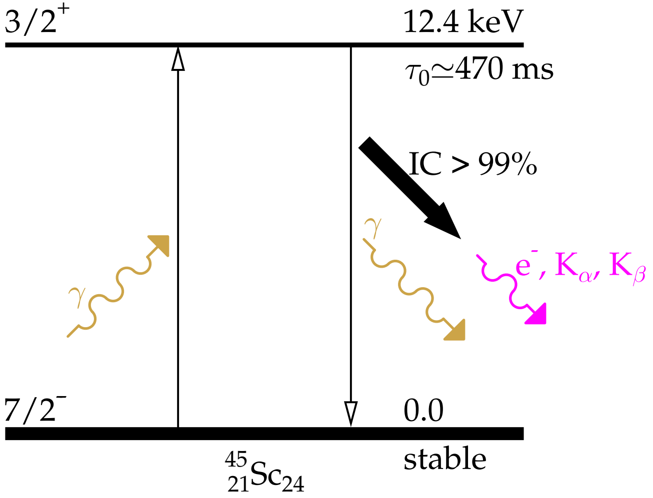






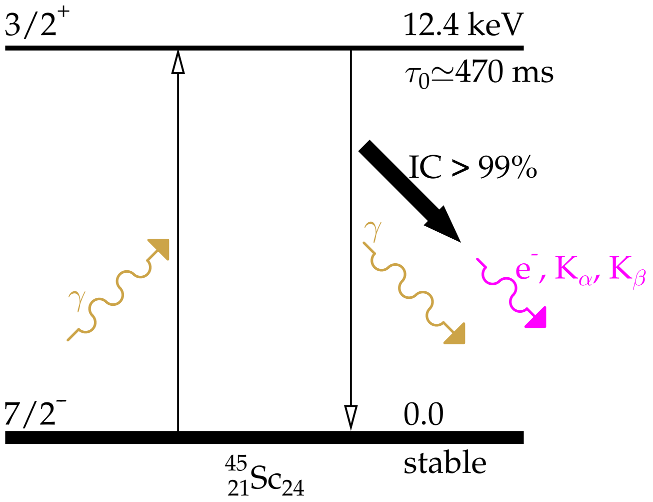
^{45}Sc Experiment in a Nutshell: Highest Spectral Flux – Lowest Detector Background

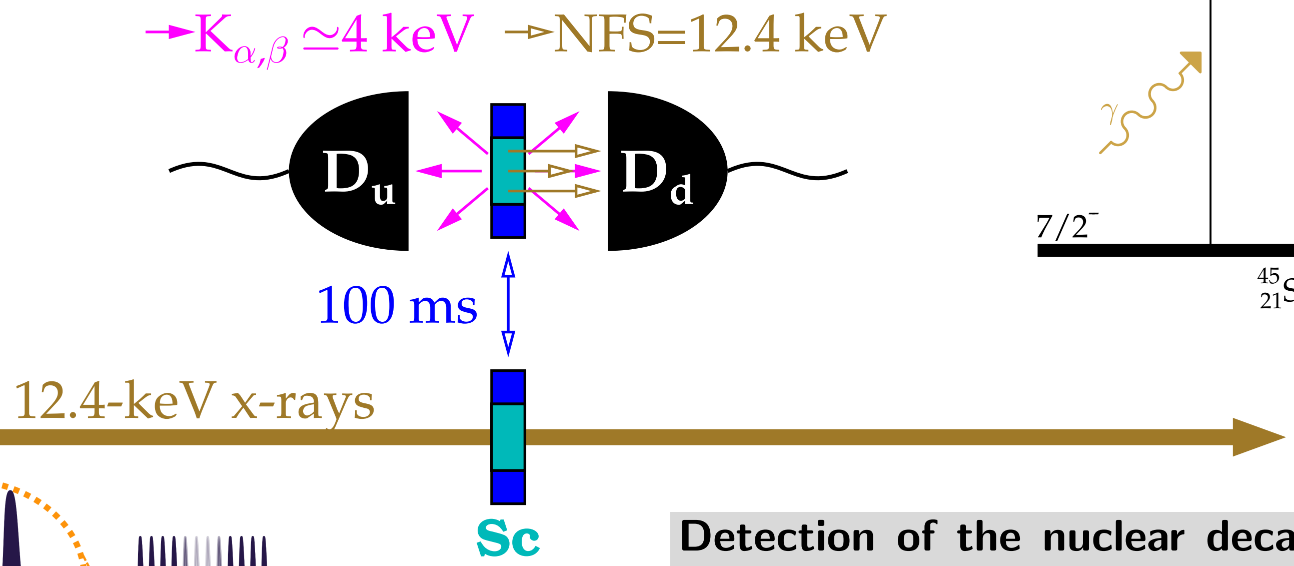


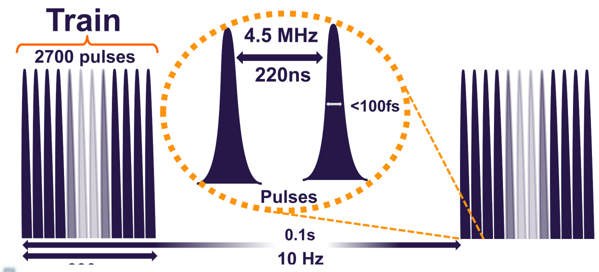


^{45}Sc Experiment in a Nutshell: Highest Spectral Flux – Lowest Detector Background

With a 25-μm thick Sc metal target exposed to an x-ray beam with $\simeq 1$ ph/s/$\Gamma_0 \simeq 10^{15}$ ph/s/eV, the expected count-rate in D_1 or D_2 of 4-keV $K_{\alpha,\beta}$-fluorescence is 1-5 ph/100 s.

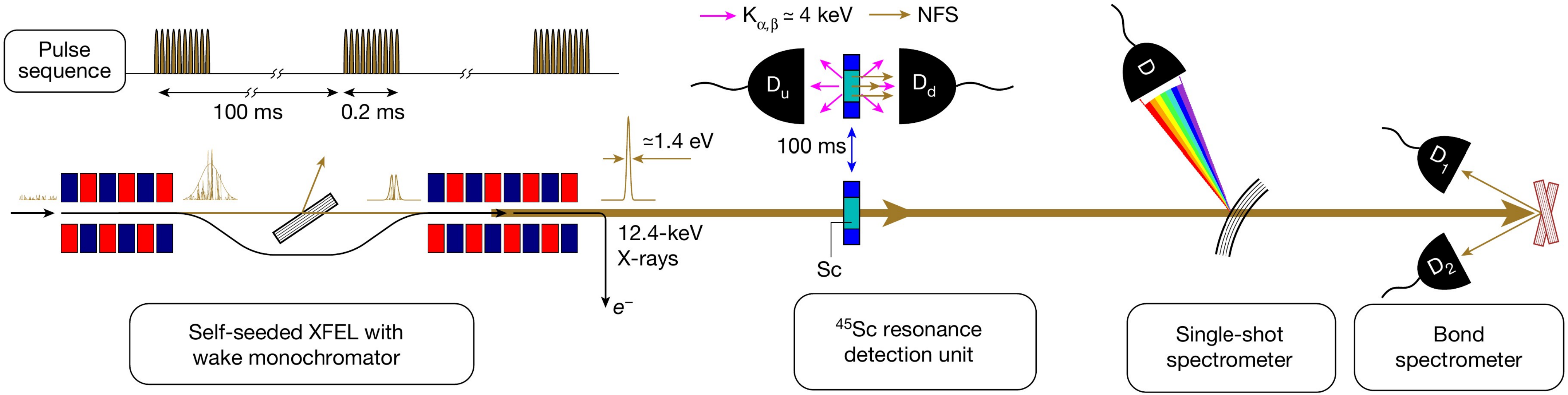




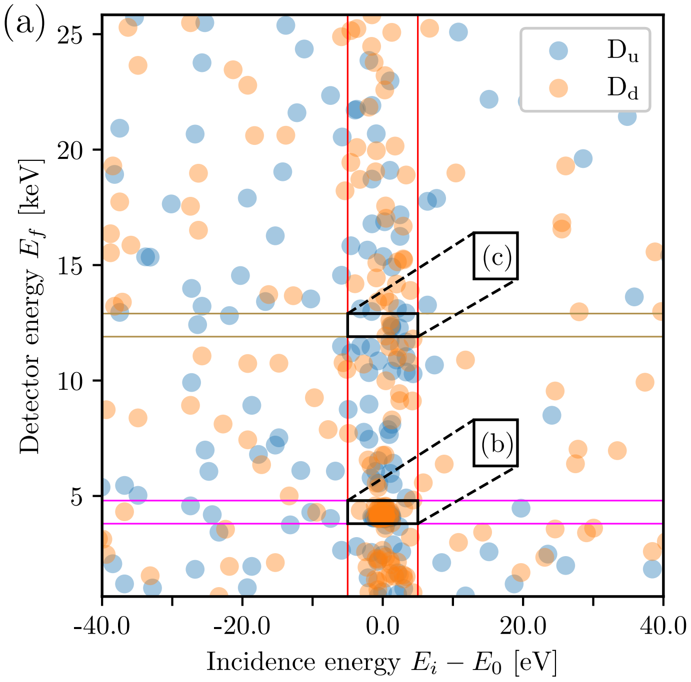


Detection of the nuclear decay products with time, spatial, energy, and polarization discrimination ensures very low detector noise floor <2 counts/10000 s.

Experimental Setup and Experiment Execution (2022)



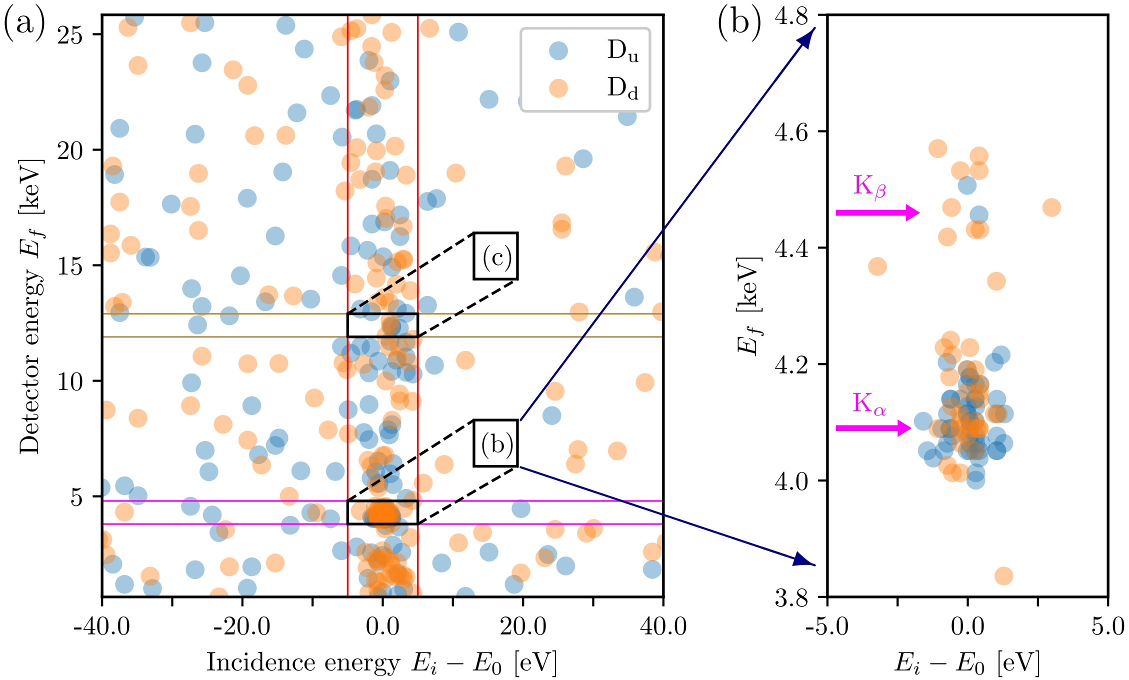
^{45}Sc Resonance Detected - 2D Picture



(a) Counts from the D_u and D_d x-ray detectors plotted as the energy E_f of the detected X-ray photons versus incident X-ray photon energy $E_i - E_0$. The photons were recorded in a time window of ≃20-80 ms after every pulse-train excitation.

- **XFEL photon energy E_i was scanned in a ± 50 eV range around 12.4 keV.**
- **≃ 10^{20} of 12.4-keV photons were directed to Sc targets.**

^{45}Sc Resonance Detected - 2D Picture



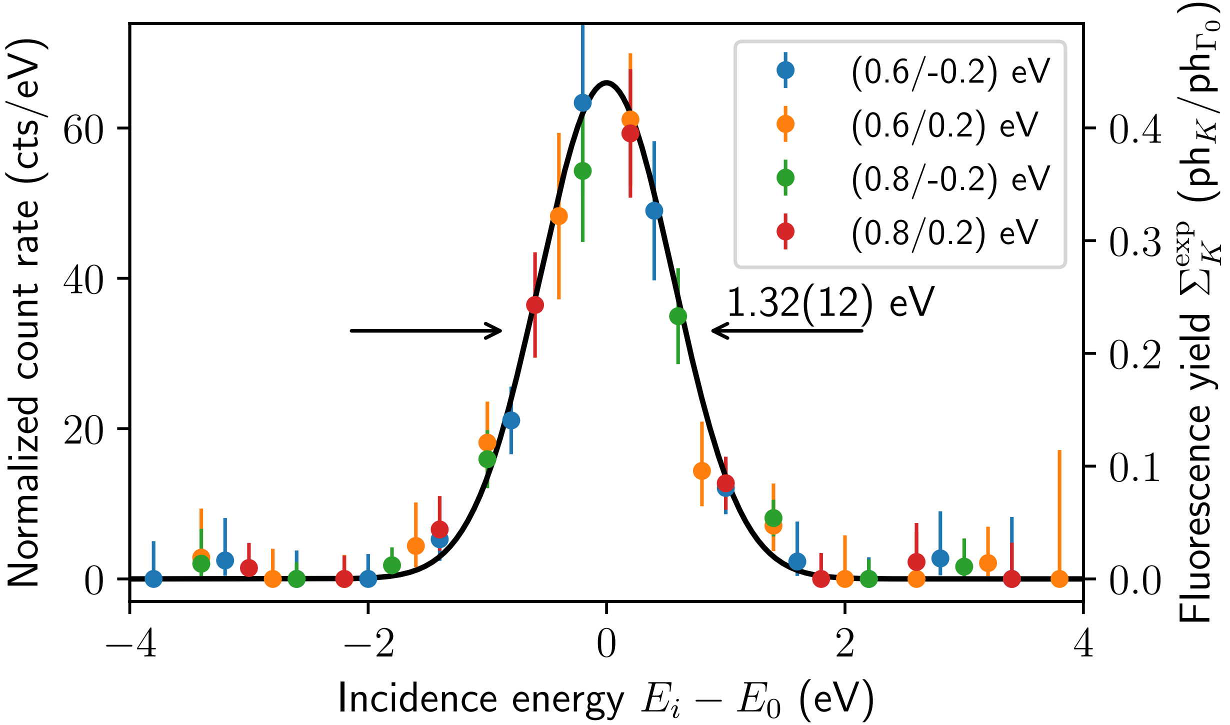
- XFEL photon energy E_i was scanned in a $\pm$ 50 eV range around 12.4 keV.
- $\simeq 10^{20}$ of 12.4-keV photons were directed to Sc targets.

- $\simeq$93 of 4-keV $K_{\alpha,\beta}$-fluorescence photons were detected with a $>$20-ms delay.
- Signal to noise ratio $\simeq 70$.

(a) Counts from the D_u and D_d x-ray detectors plotted as the energy E_f of the detected X-ray photons versus incident X-ray photon energy $E_i - E_0$. The photons were recorded in a time window of $\simeq$20-80 ms after every pulse-train excitation.

(b) Close-up of the 4.3-keV ROI, showing two clusters of counts centered at the energies of Sc K_α (4.09 keV) and K_β (4.46 keV) fluorescence as a direct confirmation of detection of the ^{45}Sc resonance.

^{45}Sc Resonance Detected & Resonance Energy Determined



Time-delayed ^{45}Sc K-shell fluorescence as a function of incoming X-ray photon energy E_i relative to resonance energy E_0.

The spectral width of 1.32(12) eV reflects the spectral width of the incoming XFEL radiation. The colored dots are exemplary binned data.

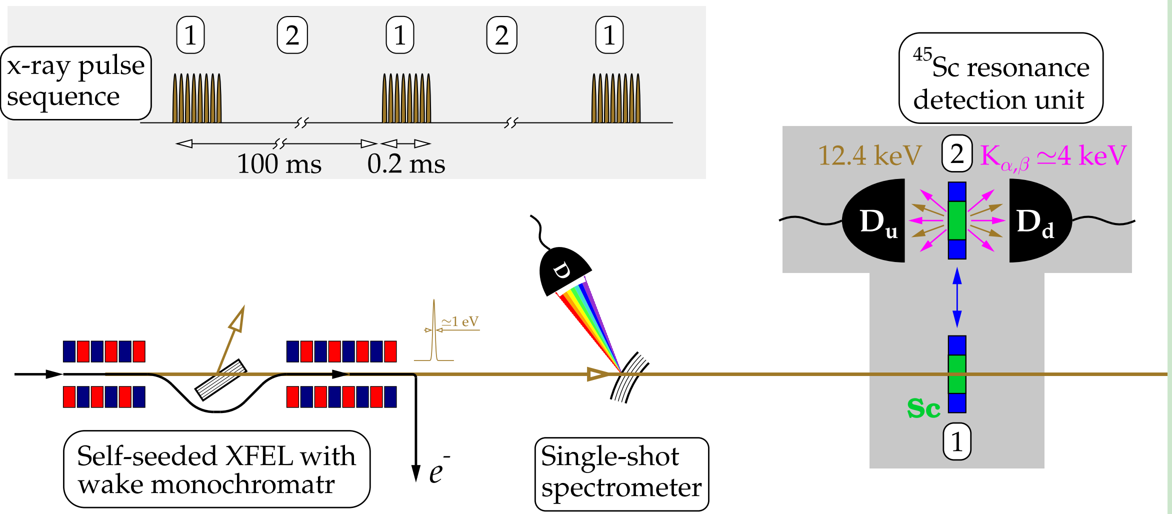
The ^{45}Sc nuclear resonance transition energy E_0 previously known to an uncertainty of $\simeq \pm 50$ eV, was determined with more than a hundred times higher accuracy as

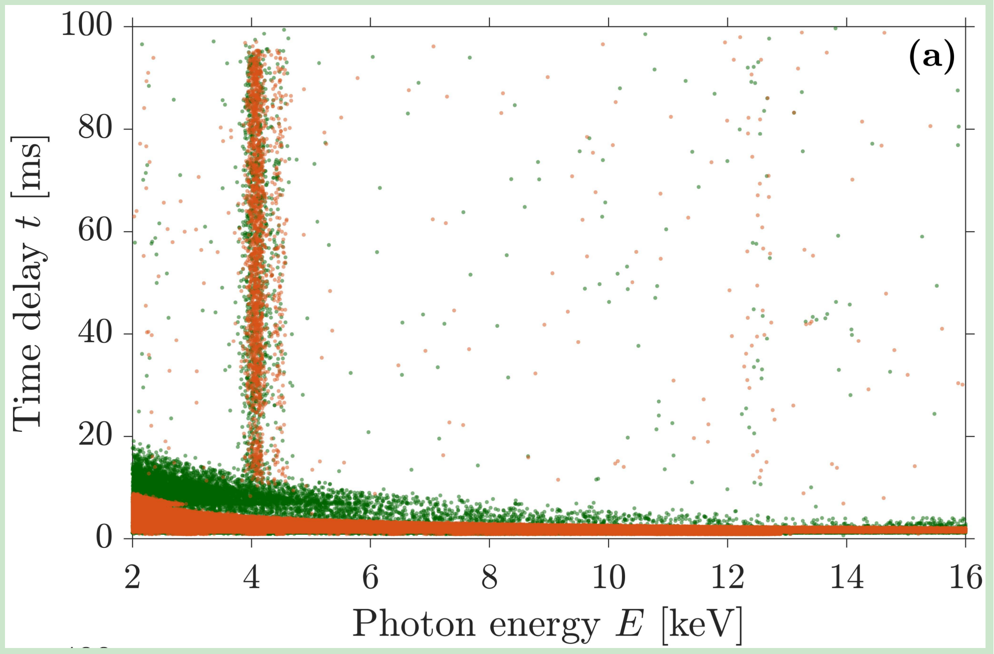
$$E_0 = 12{,}389.59^{\pm 0.15(\mathrm{stat})}_{+0.12(\mathrm{syst})}\ \mathrm{eV}$$

using Bond technique.

Incoherent Fluorescence of ^{45}Sc vs. Re-emitted Photon Energy E & Delay Time t (2024)

- ^{45}Sc fluorescence is detected with x-ray detectors D_u and D_d as a function of re-emitted photon energy E and photon delay time t after pulse excitation.
- Background (D_u+D_d): 1.8 counts/keV/10000 s.
- Total data acquisition time: 24.9 hours.

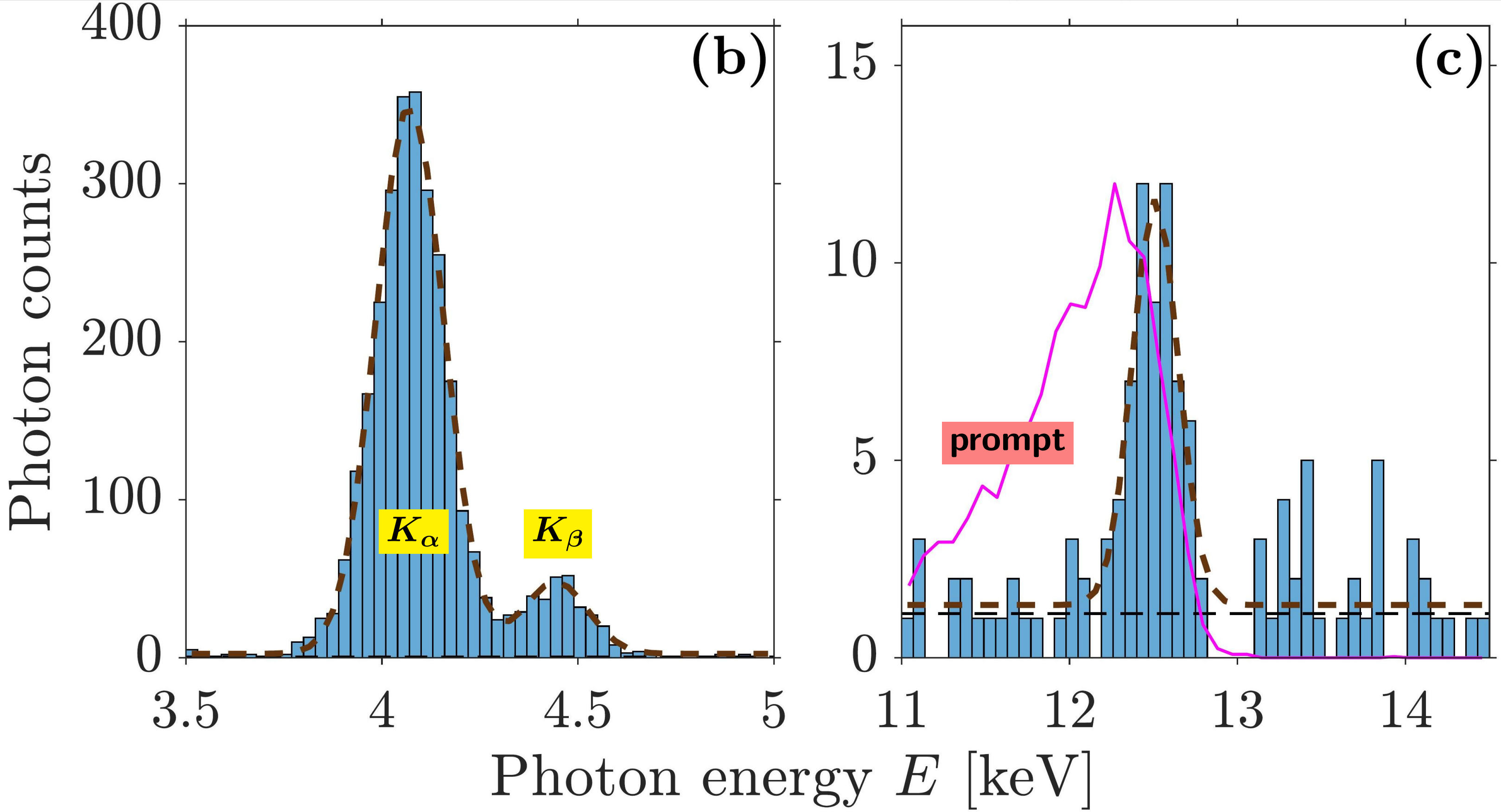




Incoherent Fluorescence of ^{45}Sc vs. Photon Energy E

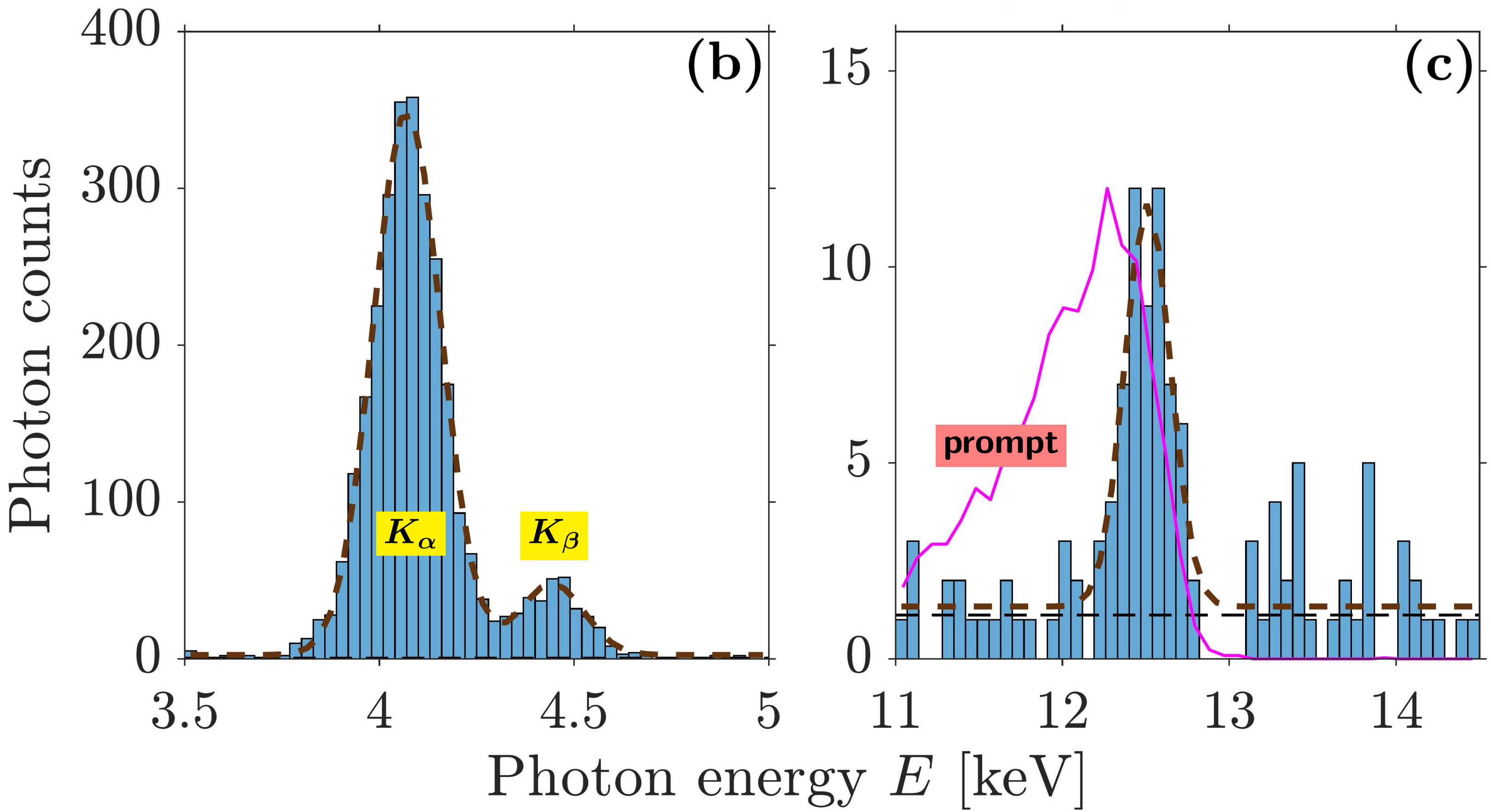
Countrates in detectors D_u+D_d [delay time integration: 15-100 ms, energy integration: 1 keV].

- Delayed 4-keV ($K_{\alpha,\beta}$) photons: R_4=328(6) ph/10000 s ⇒SNR=183 (SNR=65 in 2022)
- Delayed 12.4-keV photons: R_{12}=10(1) ph/10000 s ⇒SNR=5



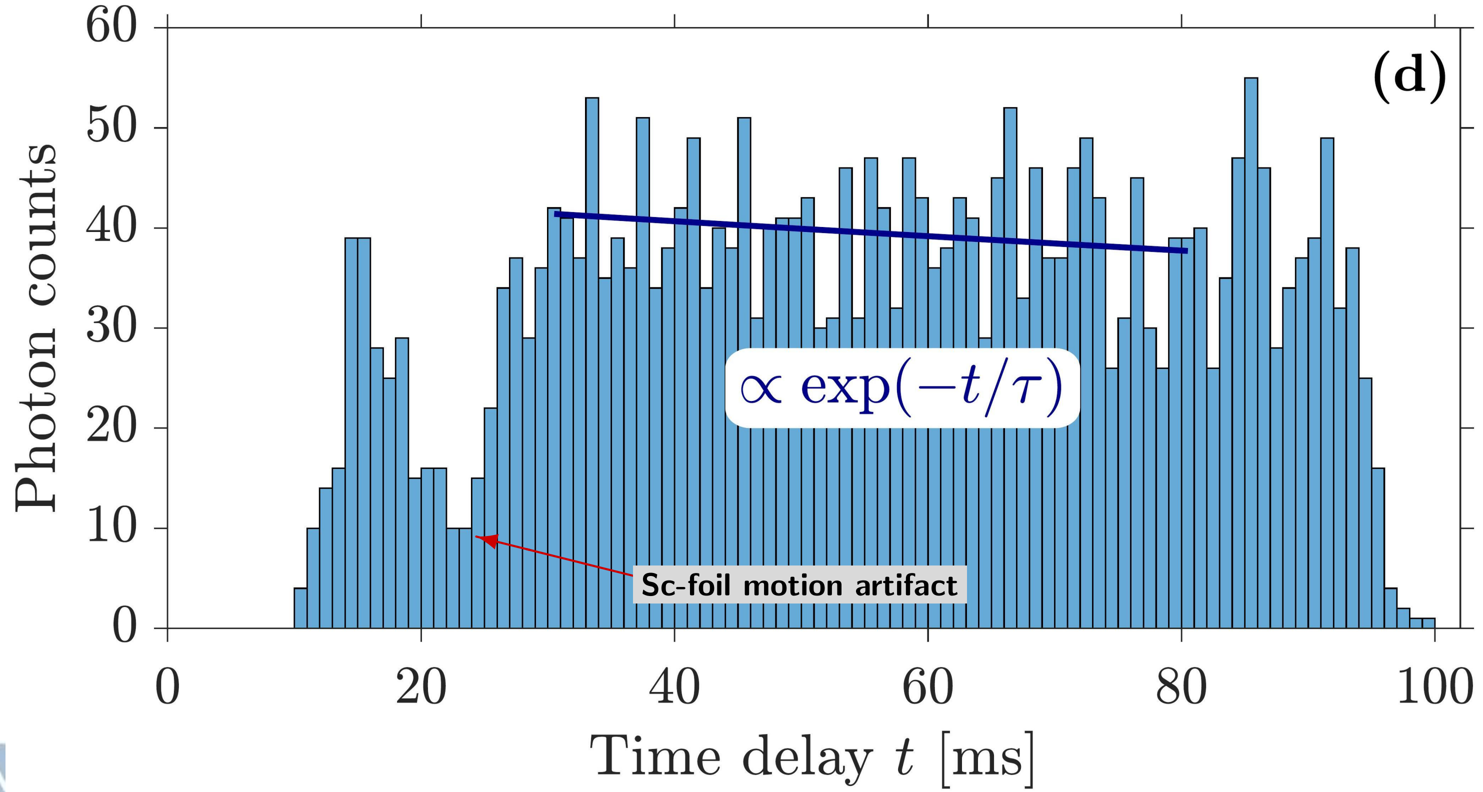
Internal Conversion Coefficient α_K for Isomer ^{45}Sc.

Measured fluorescence countrates of $K_{\alpha,\beta}$ vs. 12.4-keV allows us to determine α_K=350(65).
It agrees with α^*_K=363 predicted by the state-of-the-art theory Kibedi, T., et al., NIM A 589, 202-229 (2008)



Incoherent Fluorescence of ^{45}Sc vs. Time Delay t

- **Photon energy integration: 3.75 keV - 4.75 keV.**
- **Fit to $\propto \exp(-t/\tau)$ in a $\simeq$(30 – 80 ms) range is sensitive to: start/end and # of bins.**
- **Best fit: τ=460(250) ms.**
- **Agrees with τ=470(6) ms Blaugrund, A. E., et al., Phys. Rev. 159, 926 (1967). (via Coulomb excitation)**



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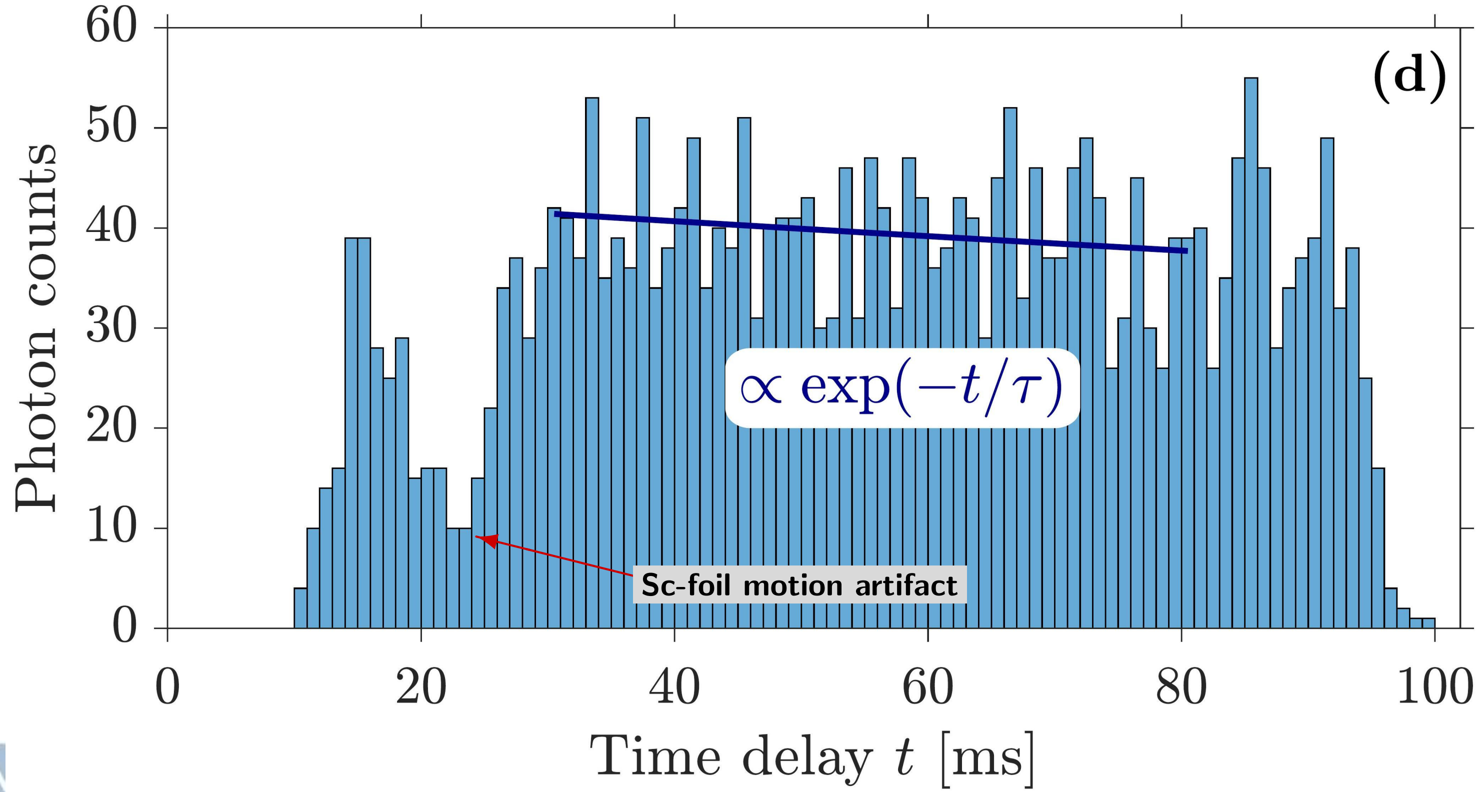

This confirms the ultra-narrow natural resonance width $\Gamma_0 = \hbar/\tau \simeq$ 1.4 feV of ^{45}Sc.

But, it is not yet the solid-state nuclear resonance width $\Gamma = \Gamma_0 + \Delta\Gamma$.

^{45}Sc Nuclear Forward Scattering to Determine the Solid-State Resonance Width

- We do not know the actual width $\Gamma = \Gamma_0 + \Delta\Gamma$ of the ^{45}Sc resonance in a solid state target, and how much is it broadened – $\Delta\Gamma$ – compared to the natural linewidth Γ_0=1.4 feV?
- Measuring $\Gamma \simeq$ feV directly is a formidable challenge.

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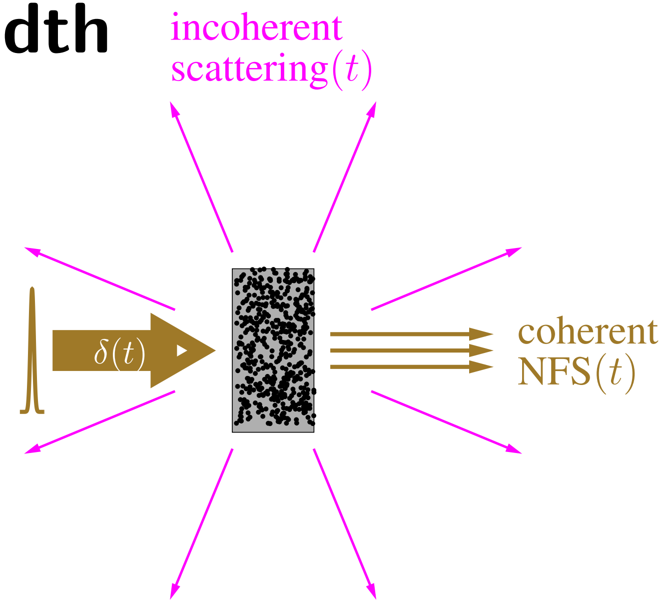
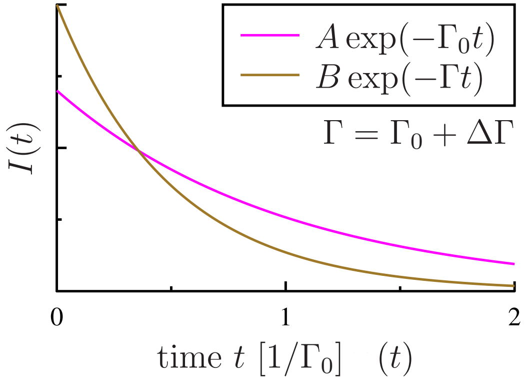
- Measuring $\Gamma \simeq$ feV directly is a formidable challenge.

- Measuring complementary time dependences on the millisecond-scale instead of energy dependences on the feV-scale is a more straightforward approach.
- It requires measuring time dependence of coherent nuclear forward scattering (NFS) to access Γ.

Yu. Shvyd'ko and G.V. Smirnov NIM 51 (1990) 452-457





Nuclear Forward Scattering (NFS) Set-up

- NFS crystal targets (Sc, Sc_2O_3, ScN, and $ScAlMgO_4$) are in a cryostat at 40 K.

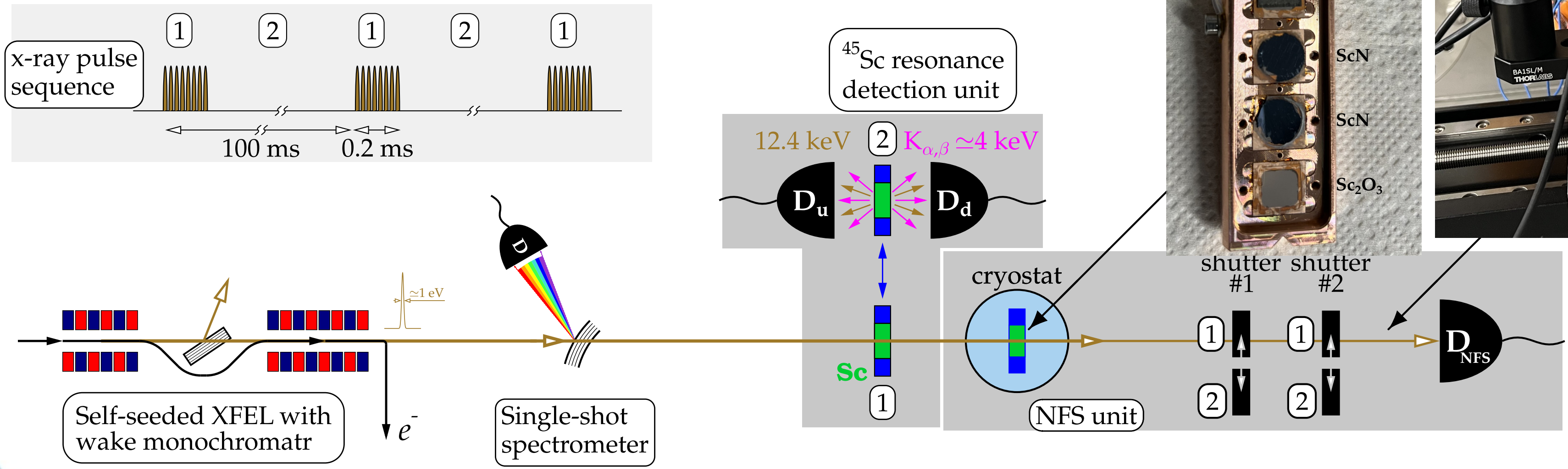
Each target is placed into the beam sequentially and raster-scanned to reduce radiation damage.

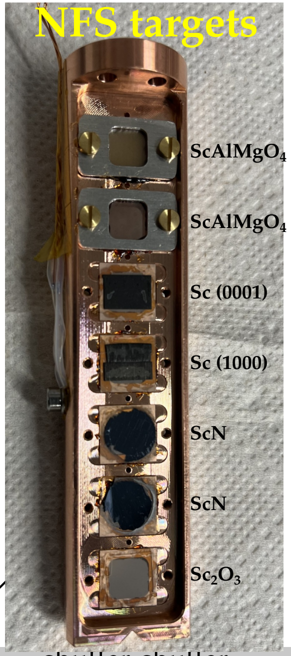
- Two synchronized shutters #1 & #2 open NFS path

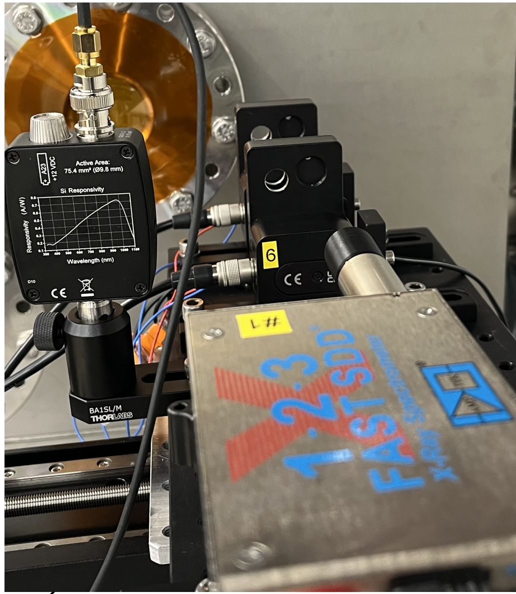
from the target to the NFS detector 2 ms after the excitation.

- The resonance detection unit is always on:

to confirm that the XFEL is on resonance.







^{45}Sc NFS Targets (only crystal samples are used)

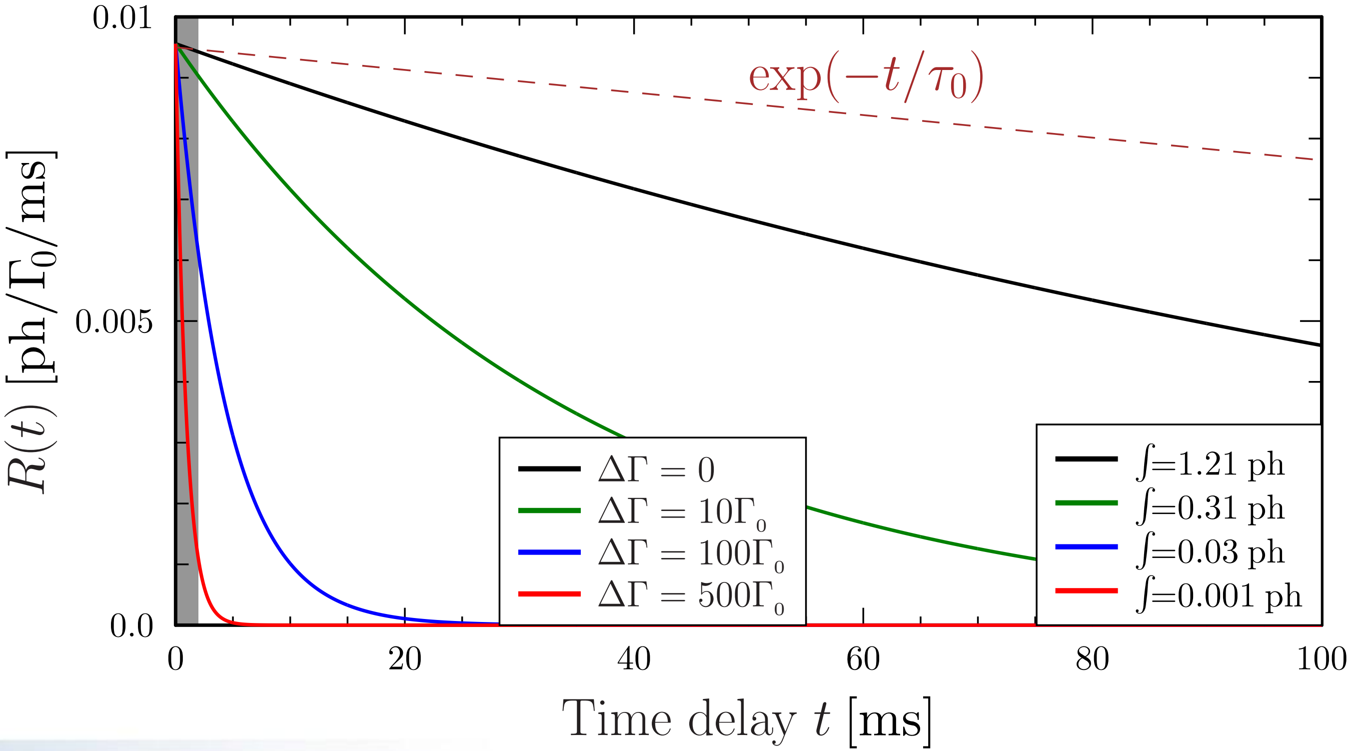
Selection criteria: minimal magnetic and electric interactions; high density of Sc; low density and photo-absorption of ballast elements; high thermal conductivity; chemical purity and stoichiometry; crystal perfection, etc.

substance	Sc	ScN	Sc_2O_3	$ScAlMgO_4$	ScF_3 (not used)
space group	$P63/mmc$ (194) hcp	$Fm\bar{3}m$ (225) NaCl	$Ia\bar{3}$ (206)	$R\bar{3}m$ (166) [?]	$Pm\bar{3}m$ (221) [? ?]
magnetism	paramag.	diamag.	diamag.	diamag.	diamag.
Quadrupole interaction parameters eQ_gV_{zz}/h [MHz]	$\simeq 2$ [?]	0 [? ?]	15.5 - 24.4	?	0
η	0?	0	0.69 - 0 [? ?]	?	0
density [g/cm^3]	2.985	4.28	3.86	3.64	2.57
Photo-absorption length L_a [μm]	60	54.5	69.7	133	146
Sc number dens. $N_0 \times 10^{22}$ [1/cm^3]	3.98	4.37	3.18	0.88	1.54
Optimized optical thickness $L_{eff} = \sigma_R N_0 2L_a$	9.06	9.04	8.42	4.45	8.57
Optimized $\xi = L_{eff}/4$	2.26	2.26	2.10	1.11	2.14
Thermal conductivity @300 K [W/m K]	15.8	51-56 [?]	17.3	4 [?]	9.6 [?]

^{45}Sc Nuclear Forward Scattering (NFS) - Expected Results

- $R(t)$: NFS rate as function of time delay after resonant excitation with ultra-short x-ray pulse.
- $R(t)$ is calculated for

– a single resonance for different values of inhomogenious resonance broadening $\Gamma = \Gamma_0 + \Delta\Gamma$
– a NFS target with nuclear resonance optical thickness parameter $\xi = 2$ (as in the experiment)
– Incident spectral flux: 1 ph/Γ_0/pulse.



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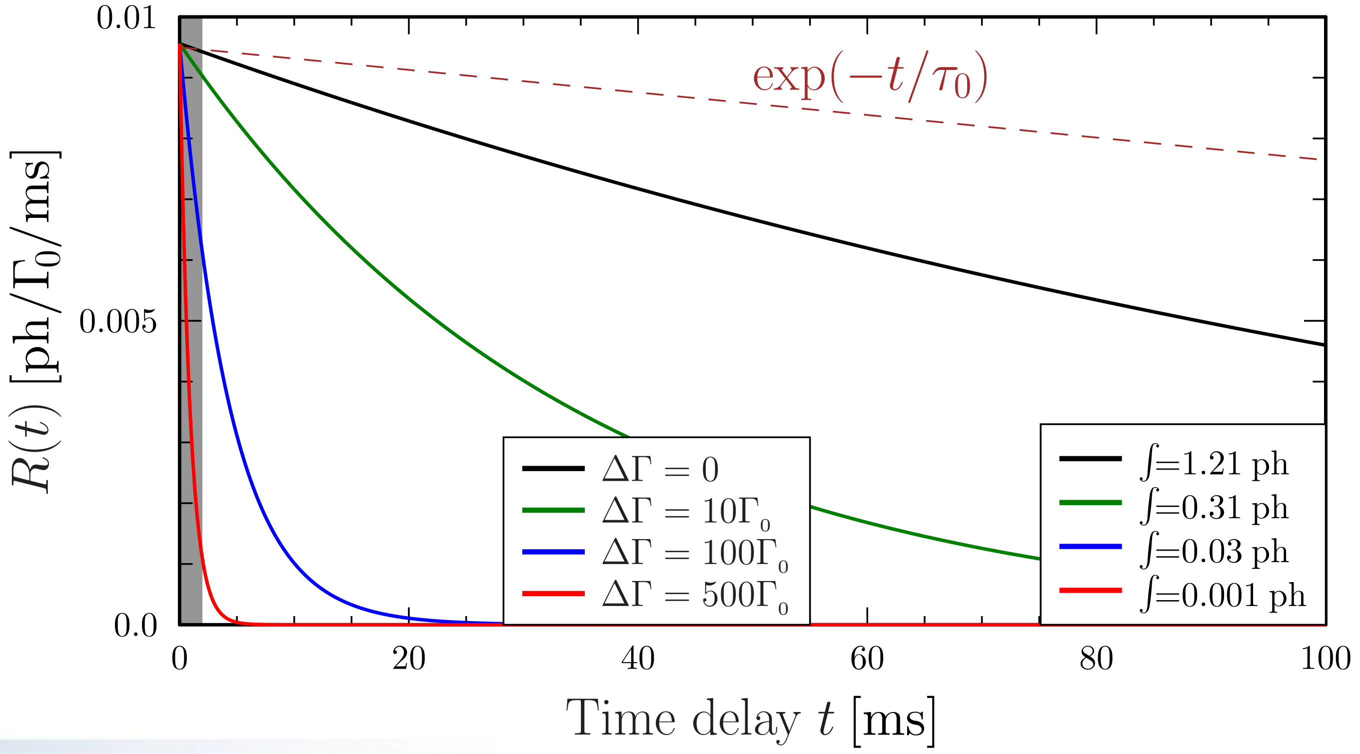
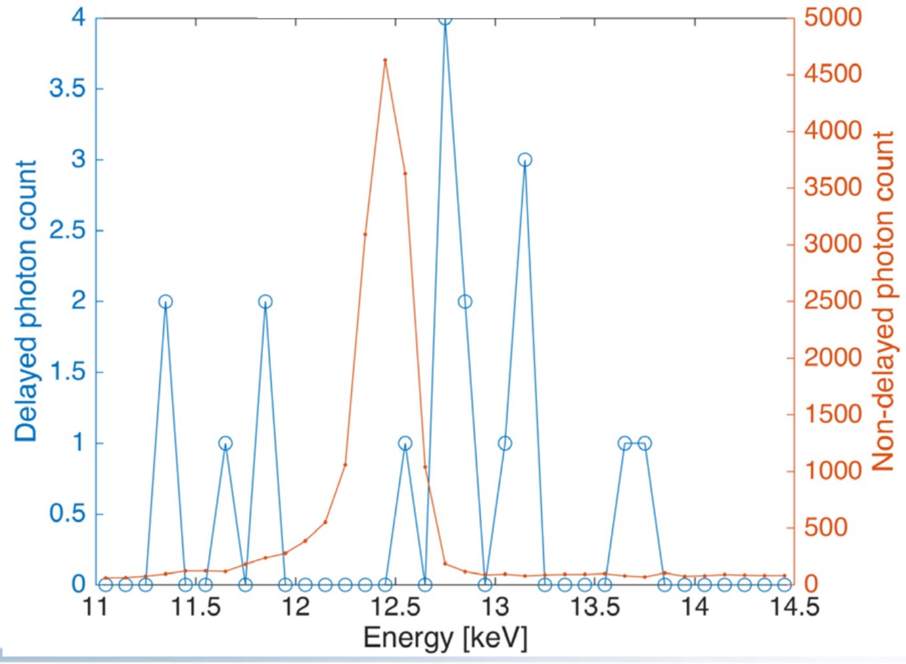
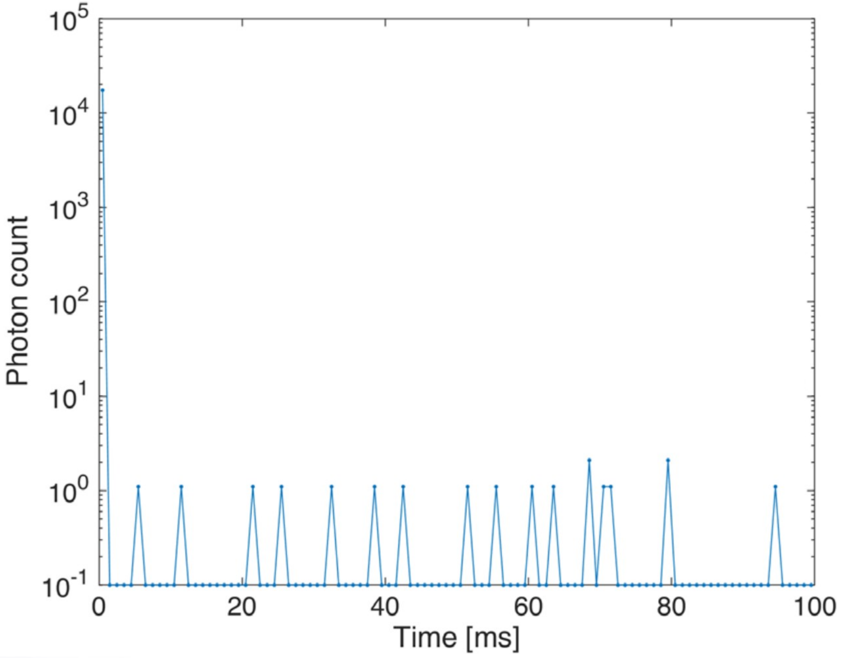
- With a minimum measurable delay time of 2 ms, NFS can be detected if $\Delta\Gamma < 500\Gamma_0$.



^{45}Sc Nuclear Forward Scattering (NFS) - First Results

- Delayed ^{45}Sc $K_{\alpha,\beta}$ fluorescence countrate $\simeq$300 ph/10000 s: **XFEL is on ^{45}Sc resonance.**
- 12.4-keV prompt photons (at $t = 0$) leaking through the shutter blade are detected by D_{NFS}.





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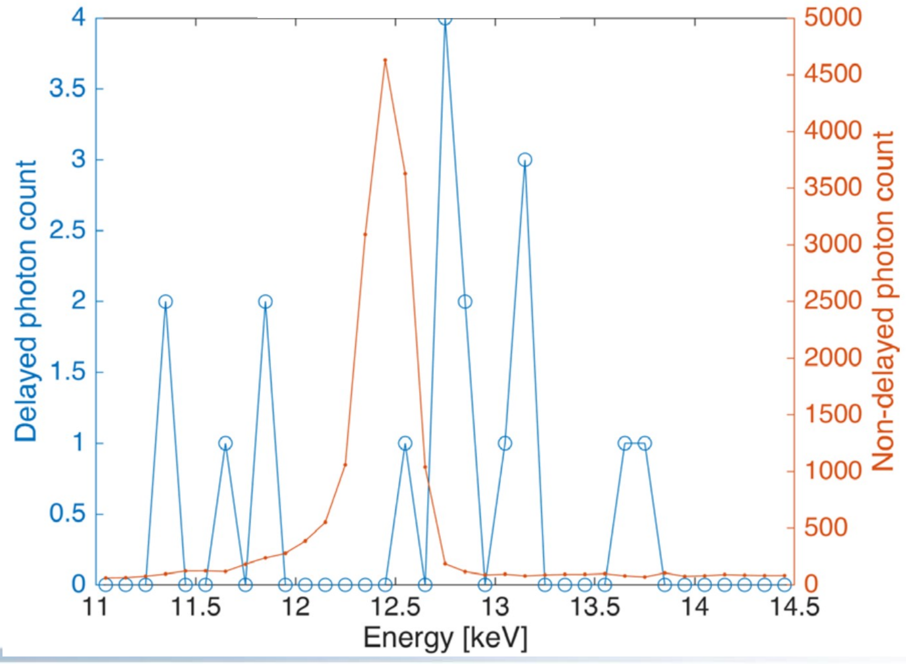
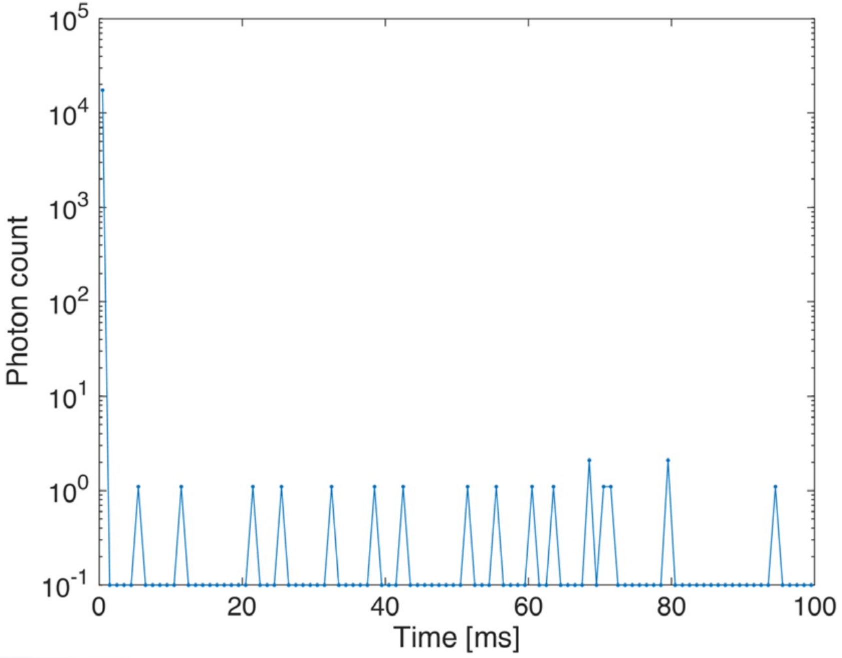
- 12.4-keV prompt photons (at $t = 0$) leaking through the shutter blade are detected by D_{NFS}.
- Delayed photons are very rare $\simeq$2 ph/10000 s, homogeneously distributed in time, and have wrong energy.
- NFS signal obviously decays faster than the smallest measurable delay time of 2 ms meaning that $\Gamma > 500\Gamma_0$.





Why $\Gamma > 500\ \Gamma_0$ and How this Could be Improved?

Why?

- Magnetic dipole-dipole interaction $\mu_e \Leftrightarrow \mu_g$ results in a maximum energy shift of

$$U_{max} = 2\mu_e\mu_g/R^3 \simeq 1500\ (3000)\ \Gamma_0 \text{ or } 500\ (1000)\ \text{Hz}$$

for ^{45}Sc in Sc_2O_3 (ScN)

- Unhomogeneous broadening due to distribution of HFI parameters in imperfect NFS crystals.
- Heatload induce further crystal imperfections.

How?

- NFS crystal targets of highest crystallinity and chemiocal purity characterized with NMR, X-rays, EDS etc.
- Milder excitation conditions.
- Dynamic resonance narrowing, etc. ....

^{45}Sc Mössbauer Nuclear Clock and XFELO

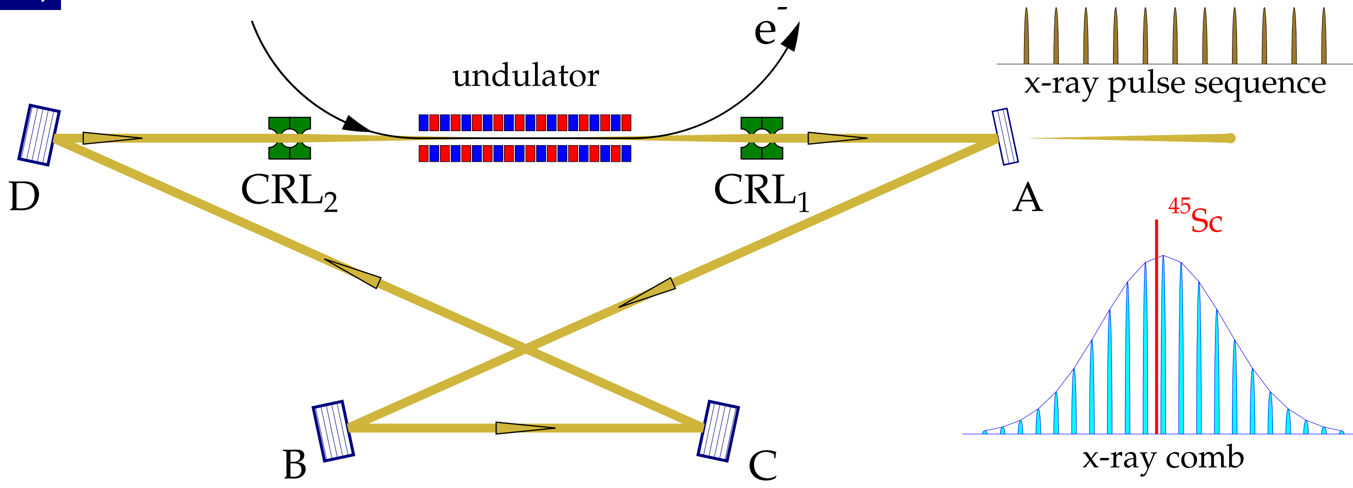
Realization of the ^{45}Sc Mössbauer nuclear clock will require
(1) a further increase of the resonant spectral flux
using improved narrow-band 12.4-keV X-ray sources and
(2) frequency combs stretching up to this energy.

^{45}Sc Mössbauer Nuclear Clock and XFELO

- X-ray free-electron-laser oscillator (XFELO) [1] and
hard X-ray comb generated by a nuclear-resonance-stabilized XFELO [2].

[1] K.-J. Kim, Yu. Shvyd'ko, S. Reiche PRL 100, 244802 (2008)

[2] B. Adams and K.-J. Kim PRAB 18, 030711 (2015)



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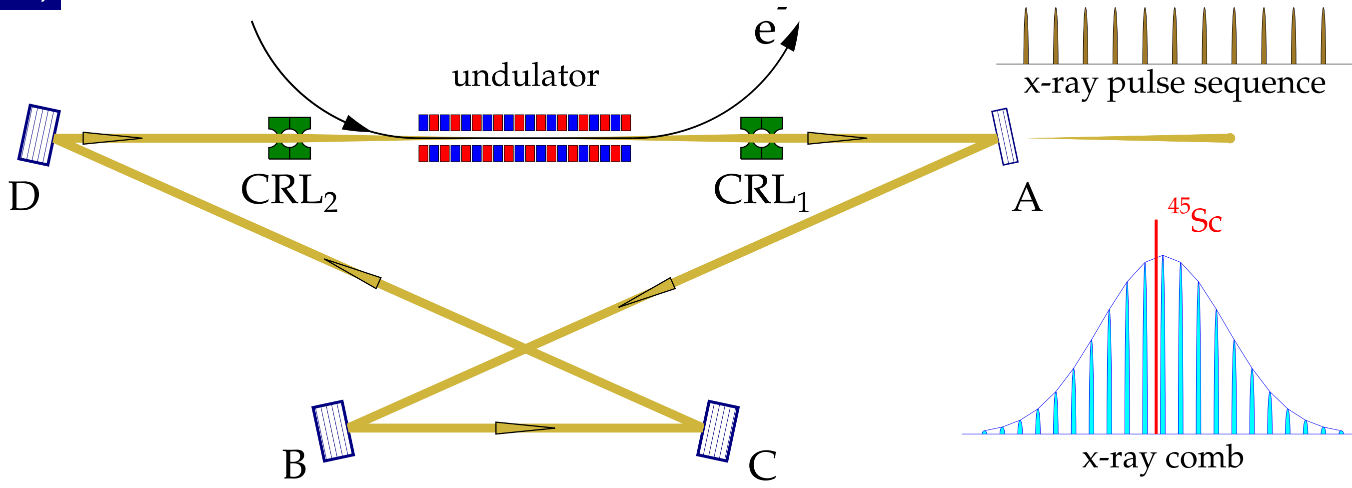

- R&D on the realization of such devices is presently in progress at ANL/SLAC (USA) [3], at EuXFEL (Germany) [4], and is considered at SHINE (China) [5].

[3] G. Marcus et al., (2019) https://doi.org/10.18429/JACoW- FEL2019-TUD04

[4] P. Rauer et al., PRAB 26, 020701 (2023) + recent demonstration (2025)

[5] N.-S. Huang et al., Nuclear Science and Techniques (2023) 34:6

Summary and Outlook

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- No NFS signal was observed indicating that the solid state ^{45}Sc resonance Γ was broadened at least by $500\ \Gamma_0$. Observation of NFS is the primary current goal.

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  - internal conversion coefficient $\alpha_K = 350(60)$
  - decay time constant $\tau_0 = 480(250)$ ms

- No NFS signal was observed indicating that the solid state ^{45}Sc resonance Γ was broadened at least by $500\ \Gamma_0$. Observation of NFS is the primary current goal.

- Progress with the seeded XFELs and XFELOs is essential for the realization of the ^{45}Sc Mössbauer nuclear clock.