

X-ray FEL Lays the Groundwork for Scandium-45 Nuclear Clock

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Brief Introduction to Scandium-45

 \bullet 45 Sc nuclei exhibit an extraordinary narrowband $\Gamma_o^{(45\rm Sc)} \simeq 1.4$ feV = 1.4×10^{-15} eV x-ray resonance transition with transition energy $E \simeq 12.4$ keV and with exceptionally large quality factor $Q=E/\Gamma_{o}^{({\rm 45}_{\rm Sc})}\simeq 10^{19}$.

• It has succeeeded in driving this extremely sharp x-ray quantum transition with an x-ray source of highest average spectral brightness: a self-seeded high-repetition-rate XFEL.

• This opens up new horizons for exquisite applications such as nuclear clocks, extreme metrology, ultra-high-resolution spectroscopy, etc.

• These applications call for further development of the XFEL technology towards XFEL oscillators (XFELO) and x-ray combs.

Γ (⁵⁷Fe) o Γ (45Sc) o = 4.8 neV 1.4 feV ≃ 3 × 10⁶ ⁴⁵Sc nuclear clock *e -* XFEL drives ⁴⁵Sc August 23, 2024 foil 1/24 CRL² CRL¹ A B C D e undulator x-ray pulse sequence x-ray comb ⁴⁵Sc

Content

- Atomic and nuclear frequency standards.
- 45 Sc as a nuclear frequency standard and XFELs.
- Resonant x-ray excitation of 45 Sc.
- Next steps.
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Article

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Resonant X-ray excitation of the nuclear clock isomer⁴⁵Sc

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Frequency Standards Enable Time Measurements

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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

Nuclear Clock Isomer ²²⁹^mTh

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

Excitation of the 229m Th resonance with photons was achieved very recently: 8.355733(10) eV

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

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

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 $-$ ⁴⁵Sc.

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, 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/ $\Gamma_{\scriptscriptstyle 0}$,

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). [5] Blaugrund, A. E., Holland, R. E. & Lynch, F. J., Phys. Rev. 159, 926–930 (1967). [4] Jones, K. W. and Schwarzschild, Phys. Rev. 148, 1148-1150 (1966).

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

• A 13-keV gamma-emitting state of 0.44-sec mean lifetime was found in ⁴⁵Sc by (p, p') reaction on ⁴⁵Sc at Argonne National Laboratory in 1964. R.E. Holland, F.J. Lynch, K.E. Nystén, PRL 13 (1964) 241-243

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

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• 45 Sc was rediscovered and resurrected in 1990:

(1) resonant excitation of 45 Sc is feasible using 12.4-keV x-rays from accelerator-based x-ray sources of high spectral brightness and flux, which started to emerge in 1990s (ESRF, APS, SPring-8); (2) spectral width of the long-lived state can be accessed by measuring time dependence of coherent nuclear-resonant forward scattering (NFS) Yu. Shvyd'ko and G.V. Smirnov NIM 51 (1990) 452-457.

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Attempts to detect the ⁴⁵Sc 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

 $XFEL$ drives ^{45}Sc foil 10/24

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 (⁴⁵Sc) was demonstrated in selfseeded mode S. Liu et al. Nature Photonics 17 (2023) 984.

• 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.

 $XFEL$ drives 45 Sc foil 11/24

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

 $XFEL$ drives 45 Sc foil 12/24 A ugust 23, 2024 foil 12/24

Experimental Setup and Experiment Execution

⁴⁵Sc 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 \simeq 20-80 ms after every pulse-train excitation.

- ${\sf a\pm 50}$ eV range around ${\sf 12.4}$ keV. $\overline{}$ **EXFEL photon energy** E_i **was scanned in**
- rected to Sc targets. $\approx 10^{20}$ of 12.4-keV photons were di-

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- rected to Sc targets. $\approx 10^{20}$ of 12.4-keV photons were di-
- were detected with a $>$ 20-ms delay. Ef [keV] • \simeq 93 of 4-keV K_{α , β -fluorescence photons}
- 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 ⁴⁵Sc resonance.

⁴⁵Sc Resonance Detected & Resonance Energy Determined

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

⁴⁵Sc Internal Conversion Coefficients α & α_K and Nuclear Resonance Cross-section σ_0 Refined

The measured nuclear-resonance-assisted K-shell fluorescence yield $\Sigma_K^{\rm exp} = 0.44(10)$ ph $_K$ /ph $_\Gamma$ for one incident resonant photon within Γ_0 is in a 100% disagreement with the value calculated with a theory, which uses published IC coefficients $[3-5]$.

A good agreement with the experiment is achieved with IC coefficients predicted by the state-of- the-art internal conversion theory [6]. The resonant cross-section σ_0 can be refined appropriately.

[1] Freedman, M. S., et al., Phys. Rev. 140, B563-B565 (1965). [2] Porter, F. T., et al., Phys. Rev. 146, 774-780 (1966). [3] Jones, K. W. and Schwarzschild, Phys. Rev. 148, 1148-1150 (1966). [4] Blaugrund, A. E., et al., Phys. Rev. 159, 926–930 (1967). [5] Burrows, T. W., Nuclear Data Sheets 109, 171-296 (2008).

[6] Kibedi, T., et al., NIM A 589, 202-229 (2008).

What are the Next Steps with ⁴⁵Sc?

⁴⁵Sc Nuclear Forward Scattering to Determine the Actual Resonance Width

• We do not know the actual width $\Gamma = \Gamma_0 + \Delta \Gamma$ **of the ⁴⁵Sc** resonance in a solid state target, and how much is it broadened $-\Delta\Gamma$ – compared to the natural linewidth $\Gamma_0=1.4$ feV?

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.

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Observation of ⁴⁵Sc NFS will not only allow us to determine Γ, but also enable important applications in ultra-high-precision coherent NFS spectroscopy, extreme metrology, etc. since the narrowness of the ⁴⁵Sc resonance is expected to surpass all currently accessible Mössbauer resonances by orders of magnitude.

Schematic of an Optical Atomic or Nuclear Clock

The laser oscillator frequency is stabilized to the atomic or nuclear resonance frequency and an optical clockwork is employed to produce a time signal.

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.

Path $#1$ to 45 Sc Mössbauer Nuclear Clock – HHG

• HHG inside a femtosecond enhancement cavity enables phase-coherent frequency combs in the XUV range:

R.J. Jones et al. Phys. Rev. Lett. 94, 193201 (2005).

A. Cingoz et al. Nature 482, 68–71 (2012).

• The intracavity HHG was extended into the XUV range with a spectral width of each harmonic of the order of 0.1 Hz:

C. Benko, et al. Nature Photonics, 8, 530–537 (2014).

This would perfectly match the spectral width of the 45 Sc resonance.

The cut-off photon energy in HHG has been pushed recently into the X-ray range (to 5.2 keV) from laser-induced multivalent ions of noble gas:

J. Gao et al. Optica 9, 1003–1008 (2022).

FIG. 1 (color). Schematic setup of intracavity high-harmonic generation. The incident pulse train is stabilized to a high finesse cavity, enhancing pulse energy nearly 3 orders of magnitude while maintaining a high repetition frequency. A gas target at the cavity focus enables phase-coherent HHG, resulting in a phasestable frequency comb in the vacuum utraviolet (VUV) spectral

Path $#2$ to 45 Sc Mössbauer Nuclear Clock – XFELO

• X-ray free-electron-laser oscillator (XFELO) [1] and hard X-ray comb generated by a nuclear-resonancestabilized 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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[3] G. Marcus et al., (2019) https://doi.org/10.18429/JACoW- FEL2019-TUD04

[4] P. Rauer et al., PRAB 26, 020701 (2023)

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

ANL-SLAC CBXFEL R&D Project (started 2019)

 $CBXFEL = Cavity-based XFEL$

 $XFELO = low-gain CBXFEL$

 $XRAFEL = high-gain CBXFEL$

ANL-SLAC CBXFEL R&D Project (started 2019)

1. Design, construct, and align a rectangular 4-crystal x-ray cavity (32-m long & 220-ns roundtrip-time) in 2-ray cavity (52-11 iong & 220-115 roundtrip-time) in ene ±e±e n ana

2. Perform 2-pass FEL gain in a 2-bunch cooper RF linac experiment as well as cavity ring-down measurements for both the low-gain XFELO and the high-gain XRAFEL schemes.

3. Demonstrate the x-ray optical cavity tolerance and stability requirements for future operation with the high repetition rate electron beam from a SCRF linac.

G. Marcus, F.-J. Decker, G. L. Gassner, et al, 39th Free Electron Laser Conf., Hamburg 2019, doi:10.18429/JACoW-FEL2019-TUD04

 $XFEL$ drives ^{45}Sc foil 23/24 A ugust 23, 2024 foil 23/24

Summary and Outlook

The long-lived nuclear transition of 45 Sc was resonantly excited from the ground to the long-lived 12-4-keV excited state by 12.4-keV x-rays for the first time.

The resonance energy that was known before the experiment to an uncertainty of ± 50 eV was determined with a sub-eV accuracy as $\mathbf{12{,}389.59}^{\pm 0.15 \mathrm{(stat)}}_{+ 0.12 \mathrm{(syst)}}$ eV.

• High repetition-rate, narrow-band XFELs is an ideal platform to drive and study longlived nuclear resonances at energies of hard X-rays. Development of XFELOs is greatly desired.

Successful resonant excitation of the 45 Sc resonance opens up new horizons for ultra-high precision spectroscopy, extreme metrology in the regime of hard x-rays, and for a solid state ⁴⁵Sc nuclear frequency standard (Mössbauer nuclear clock).

This was the first step. The next steps (NFS etc.) will be as exciting but not easier. The first ⁴⁵Sc NFS experiment is planned for October 2024 at EuXFEL.

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Supplementary Slides

Experimental Setup and Experiment Execution

Detectors D_1 - D_2 had very low background $\langle 2 \text{ counts}/10000 \text{ s} \rangle$ since they \Rightarrow were inhibited during irradiation; \Rightarrow placed 12-mm off the 12.4-keV beam; \Rightarrow had \lt 150-eV energy resolution.

Searching for ⁴⁵Sc Resonance

(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 about 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. This is a direct confirmation of detection of the ⁴⁵Sc resonance.

(c) Close-up of the 12.4-keV ROI. NFS signal at 12.4 keV is statistically insignificant. NFS is the prime next goal.

Measurement of ⁴⁵Sc Nuclear Resonance Transition Energy

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

$$
E_{_0}=12{,}389{.}59^{\pm0.15\text{(stat)}}_{+0.12\text{(syst)}}\text{ eV}
$$

using Bond technique.

Bond technique uses Bragg diffraction of X-rays with wavevector K_{0} from atomic planes with diffraction vectors (a) $H_1 = (8000)$ and (b) $H₂ = (0 8 0)$ from a Si crystal. The crystal is rotated by $\beta = \beta_1 + \beta_2$ between the angular positions of the (8 0 0) and the (0 8 0) Bragg reflections. X-ray wavelength $\lambda = 2\pi/K_0$ and photon energy E are calculated from the know crystal lattice parameter a_{si} using modified Bragg's law:

$$
\lambda = \frac{hc}{E} = \frac{a_{\rm Si}}{4} \sin(\pi/4 + \beta/2). \tag{1}
$$

W.L. Bond, Acta Crystallogr. 13 (1960) 814.

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⁴⁵Sc Parameters

Parameters of ⁴⁵Sc, taken from or based on Ref. [31] unless otherwise stated, with updated values measured or used in this work highlighted in colored boxes.

⁵⁷Fe Nuclear Forward Scattering Demonstrated in 1991

