

Photoelectron spectroscopy of nuclear transitions

Andrea Trabattoni

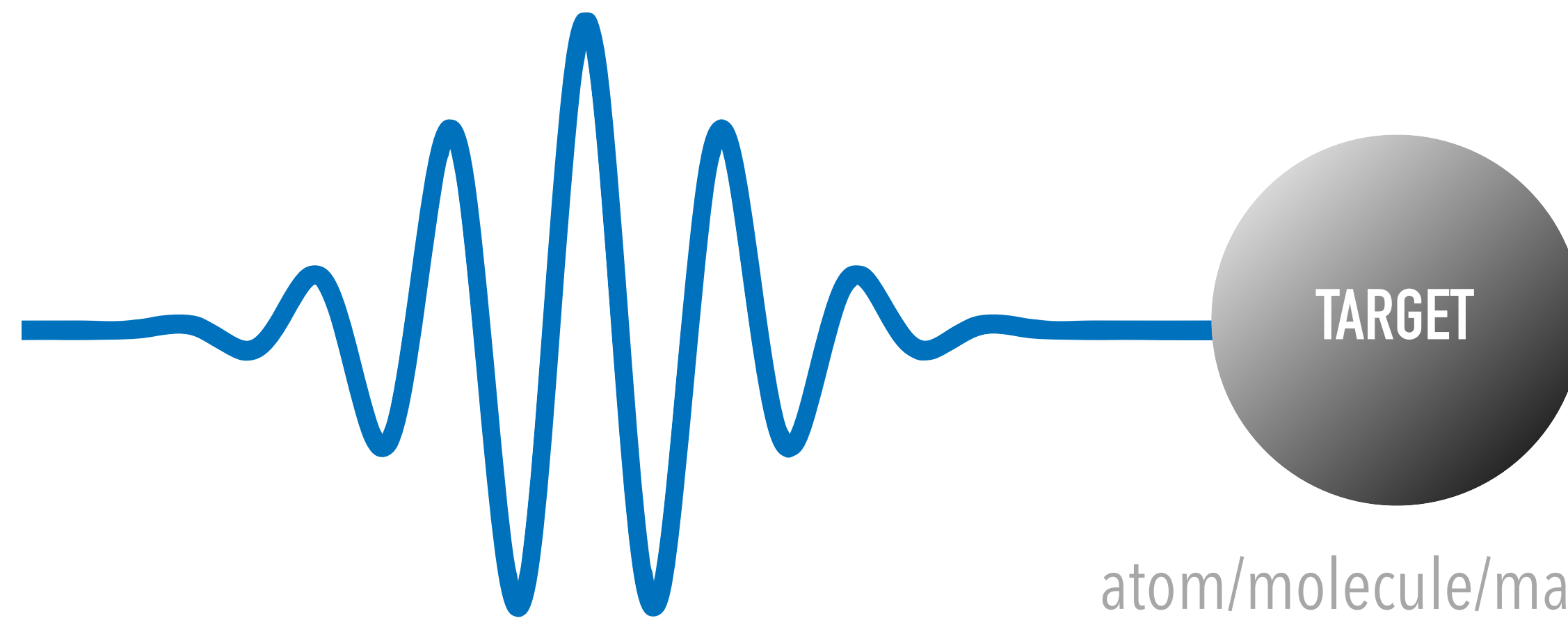
Institute of Quantum Optics, Leibniz University Hannover
Center for Free Electron Lasers, DESY





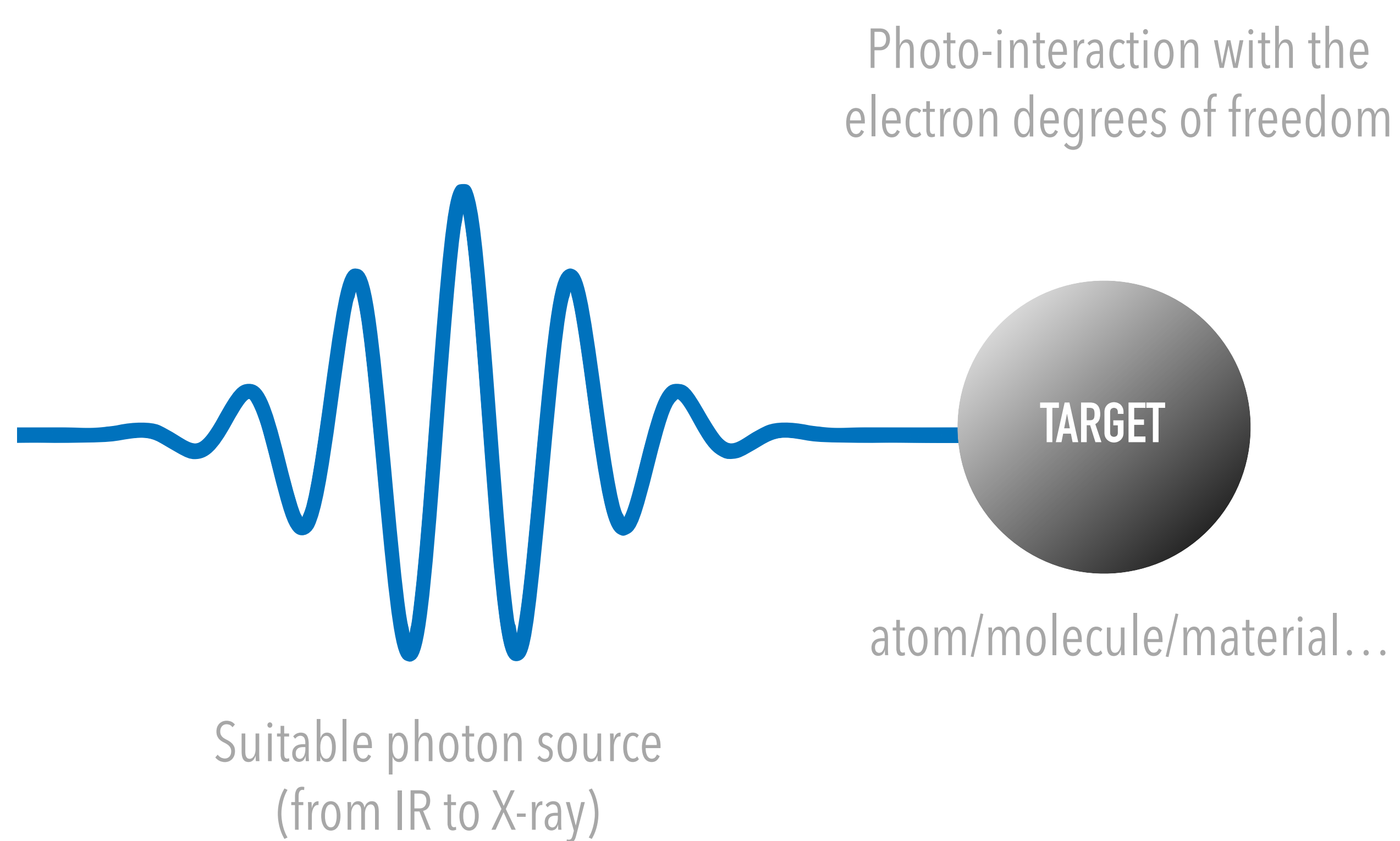
atom/molecule/material...

Photo-interaction with the
electron degrees of freedom

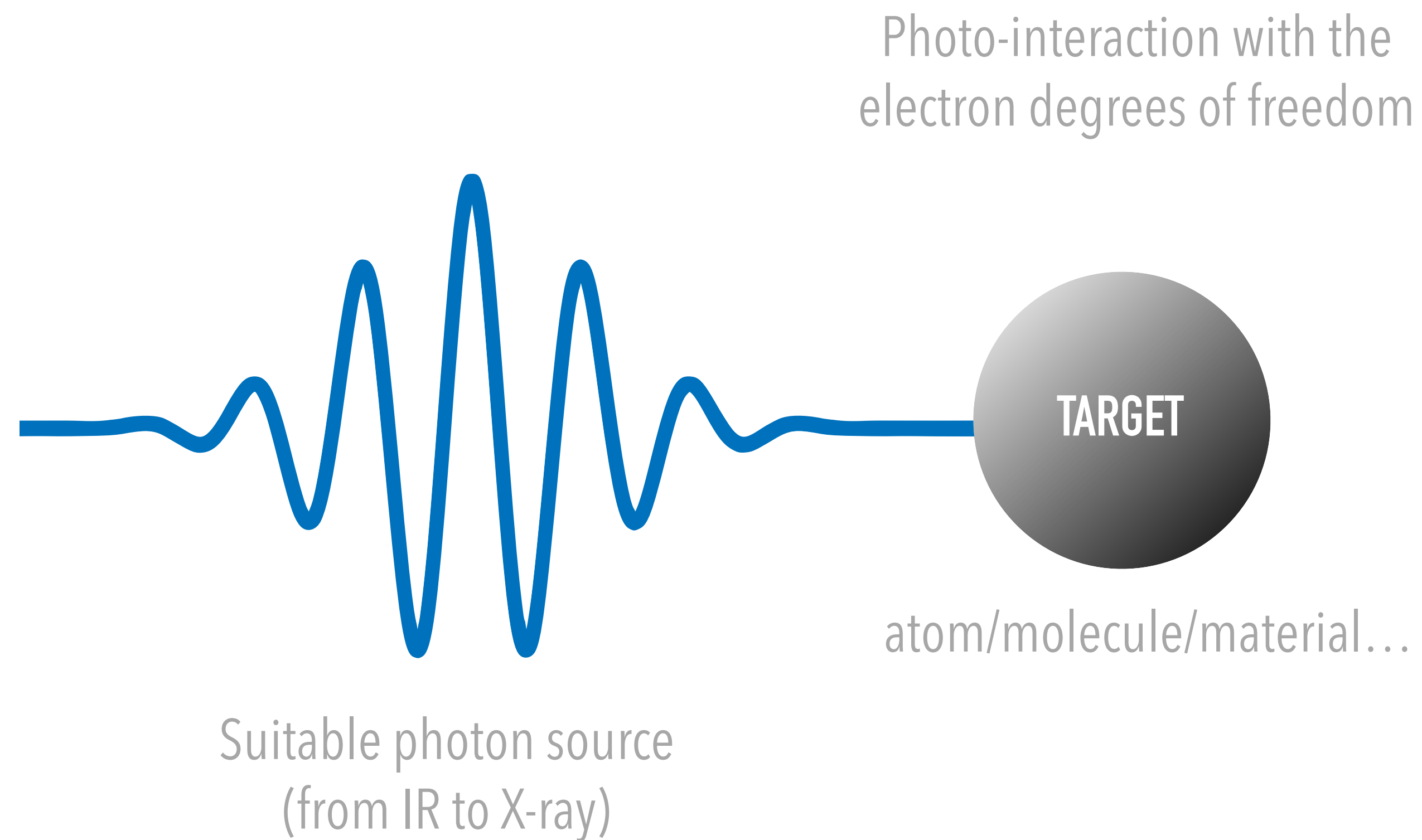


Suitable photon source
(from IR to X-ray)

atom/molecule/material...

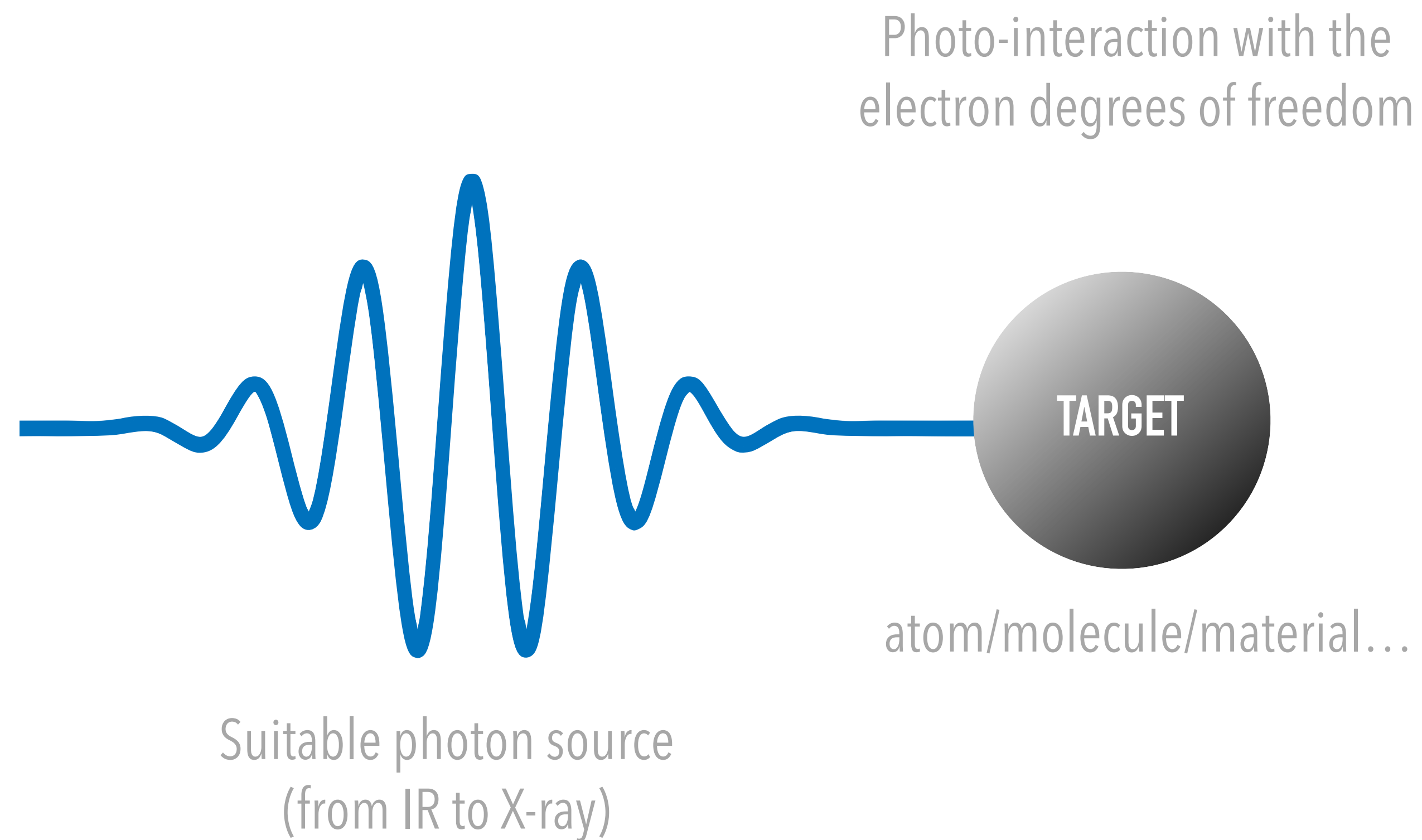


Probing the photo-induced electron dynamics.



Probing the photo-induced electron dynamics.

Understanding the role of electron dynamics in the time evolution of the system.



Probing the photo-induced electron dynamics.

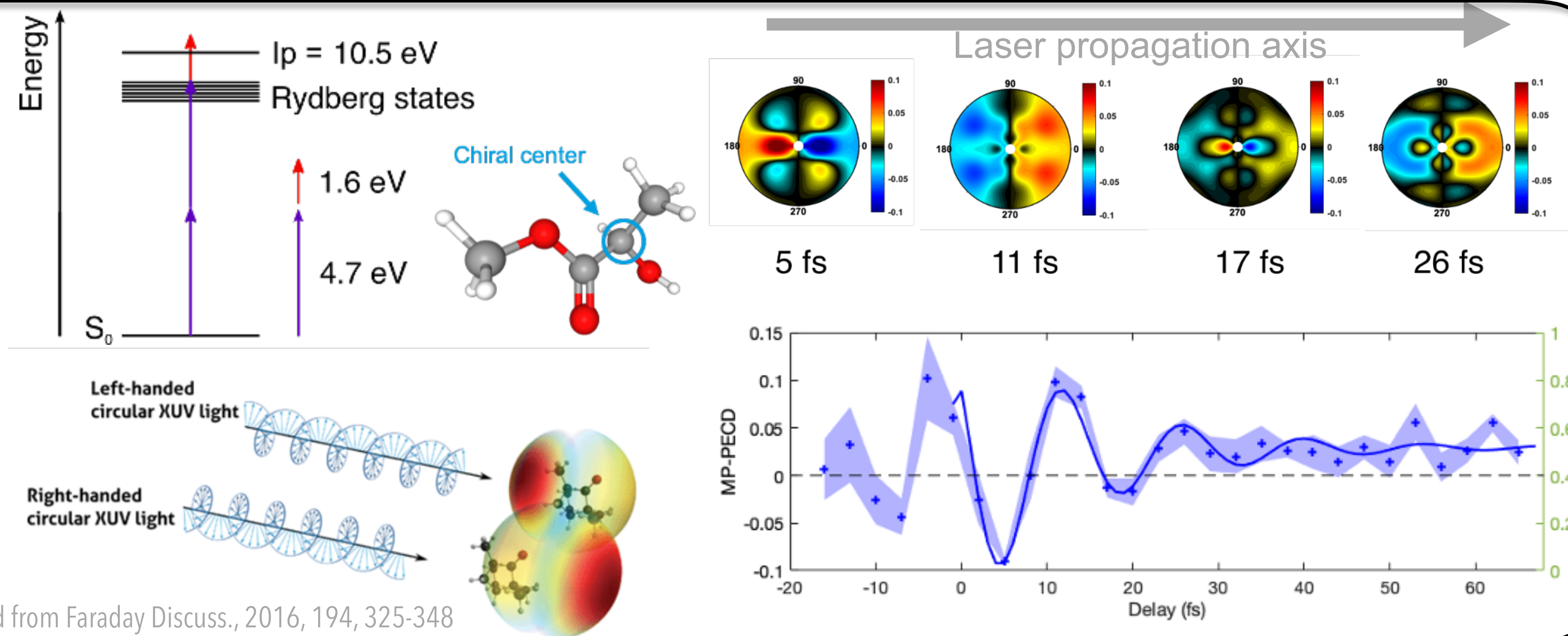
Understanding the role of electron dynamics in the time evolution of the system.

ULTIMATE AIM:

Triggering and controlling new functionalities.

Towards charge directed molecular reactivity in chiral molecules.

V. Wanie, et al., Nature 630, 109–115 (2024)



Towards charge directed molecular reactivity in chiral molecules.

V. Wanie, et al., Nature 630, 109-115 (2024)

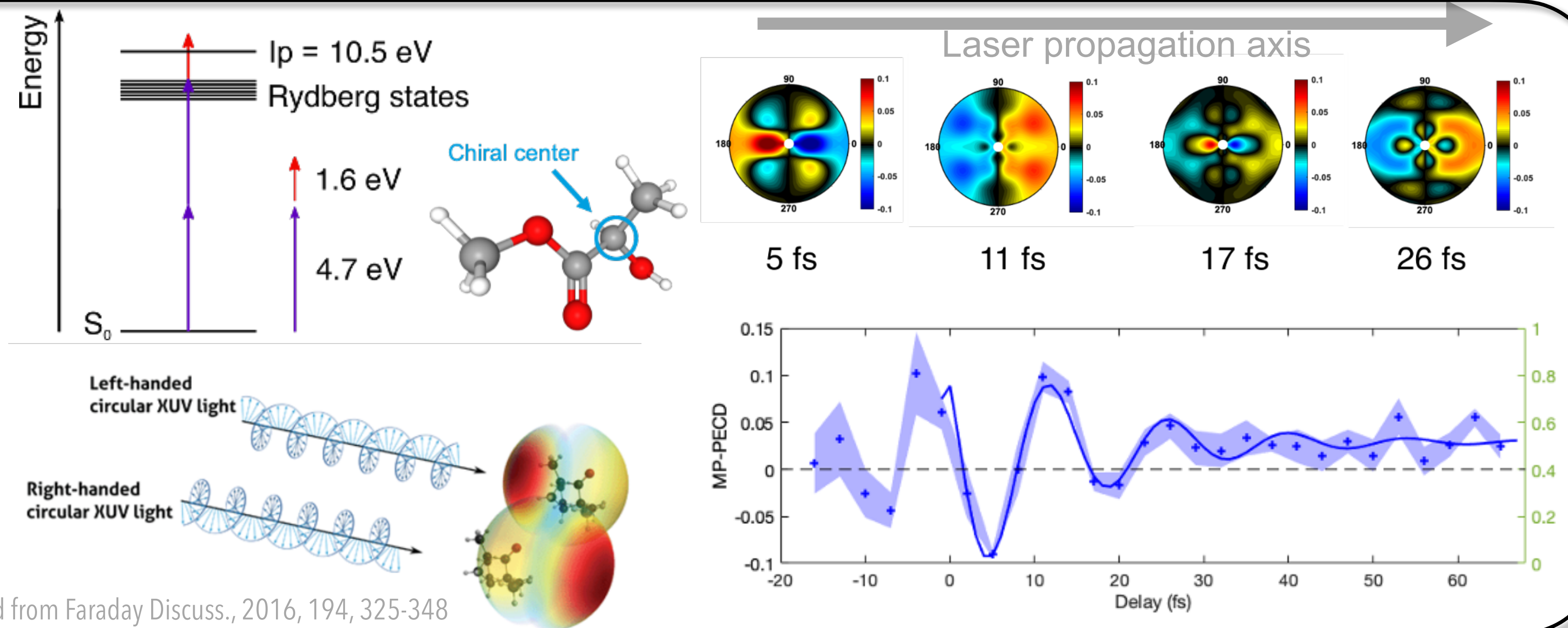
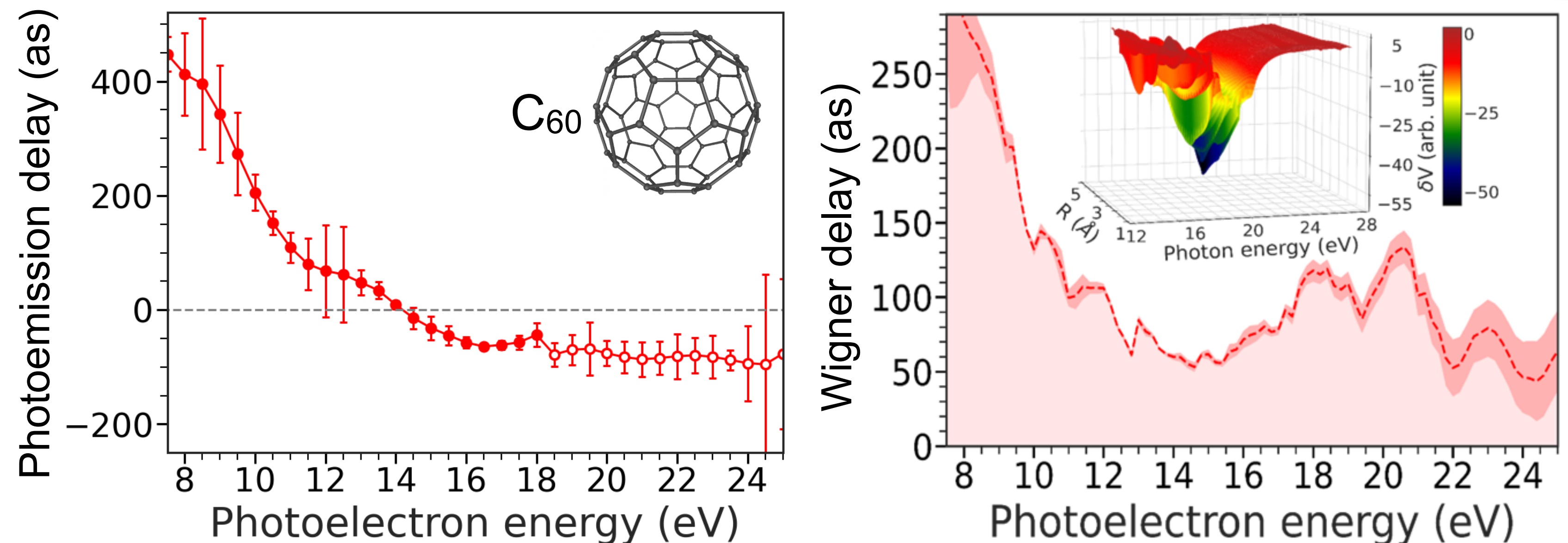


Figure adapted from Faraday Discuss., 2016, 194, 325-348

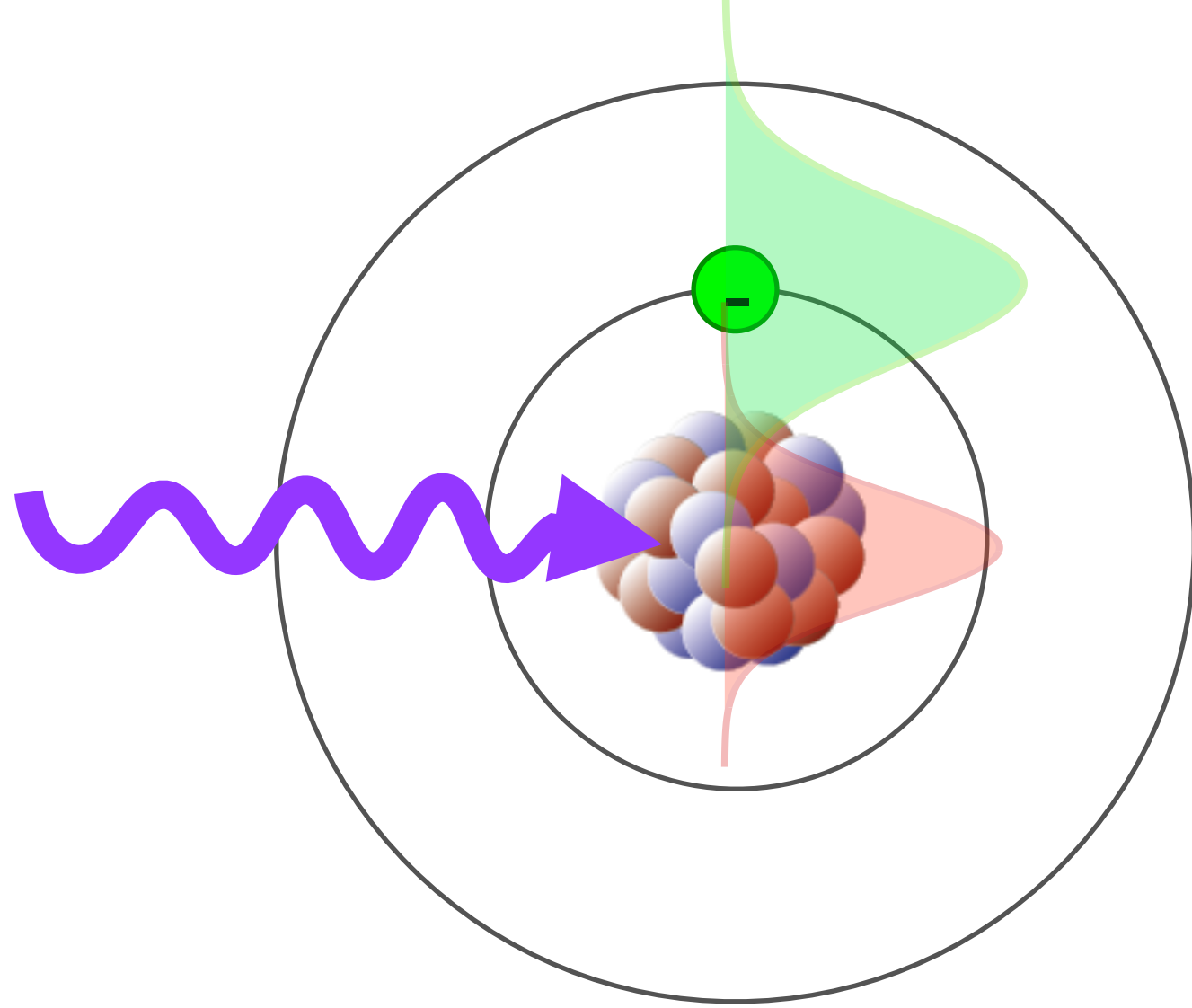
Realtime probing of plasmon dephasing time at the quantum level in fullerenes.

S. Biswas, AT, Sci. Adv. 11, eads0494 (2025)



Three-body problem: Photon-nucleus-electron

Photoexcitation of electron-nucleus couplings

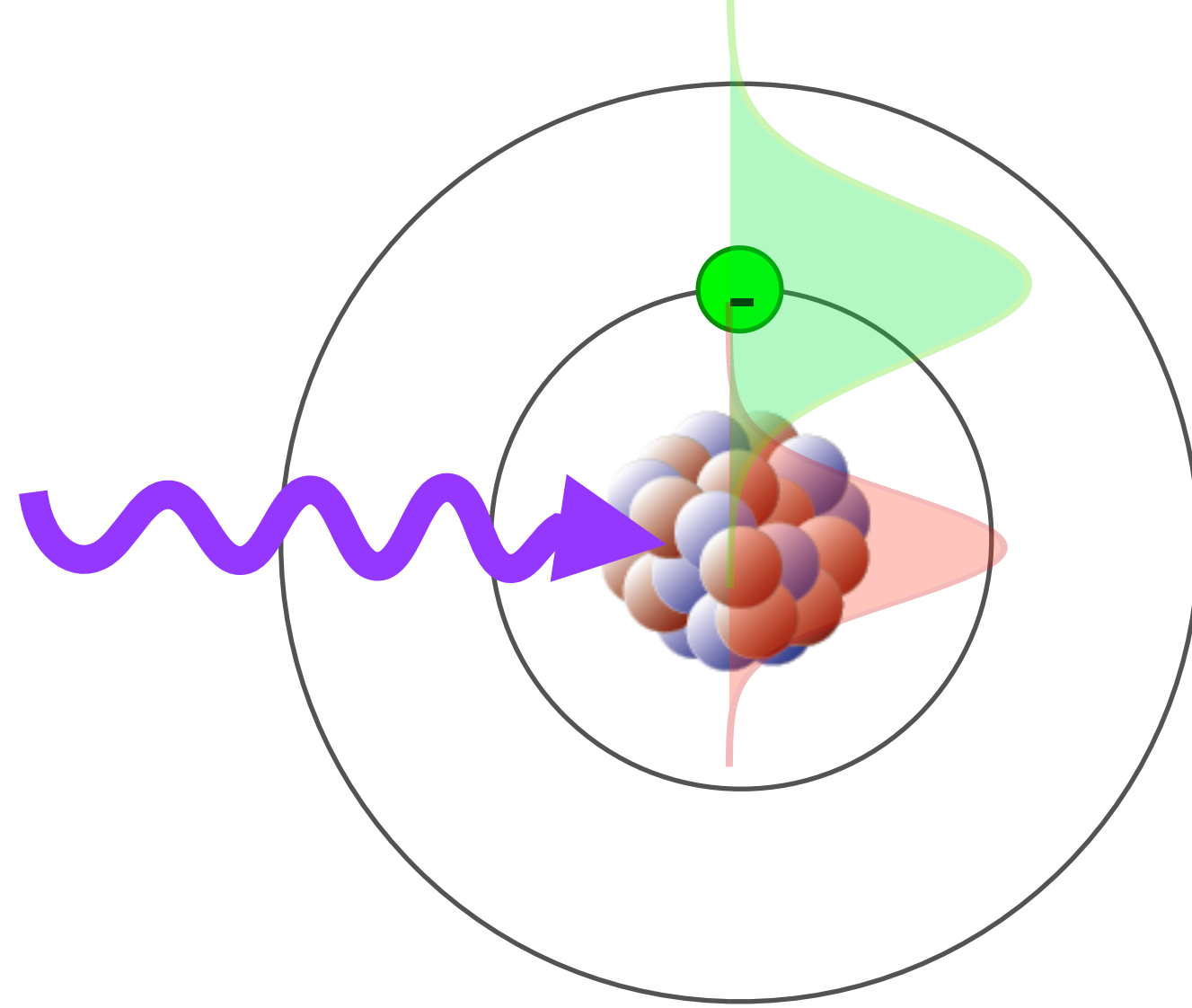


Relevant for nuclear states with $E_b < 100$ KeV.

- Internal conversion (IC), bound internal conversion (BIC)
- Nuclear excitation by electron capture (NEEC)
- Nuclear excitation by electronic transition (NEET)
- Electronic bridge (EB)

Three-body problem: Photon-nucleus-electron

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PhotoElement

HELMHOLTZ

RESEARCH FOR GRAND CHALLENGES

"Time-resolved spectroscopy of photo-induced electron dynamics in nuclear transitions"

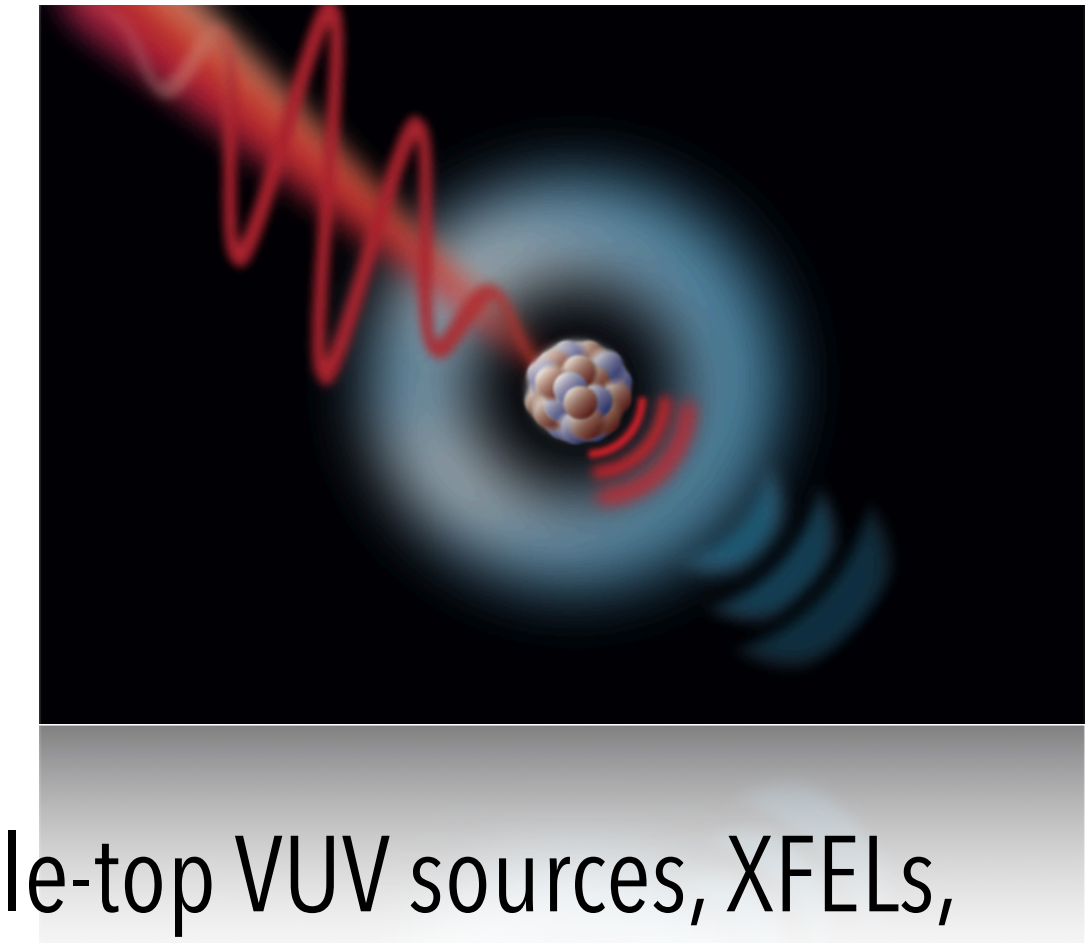
Employ advanced light sources (table-top VUV sources, XFELs, Synchrotron Mössbauer sources) to:

- 1) Better understand photo-induced electron-nucleus couplings.
- 2) Investigate the robustness of nuclear excited states against environmental perturbation.
- 3) Achieve quantum control of nuclear dynamics through electronic channels.

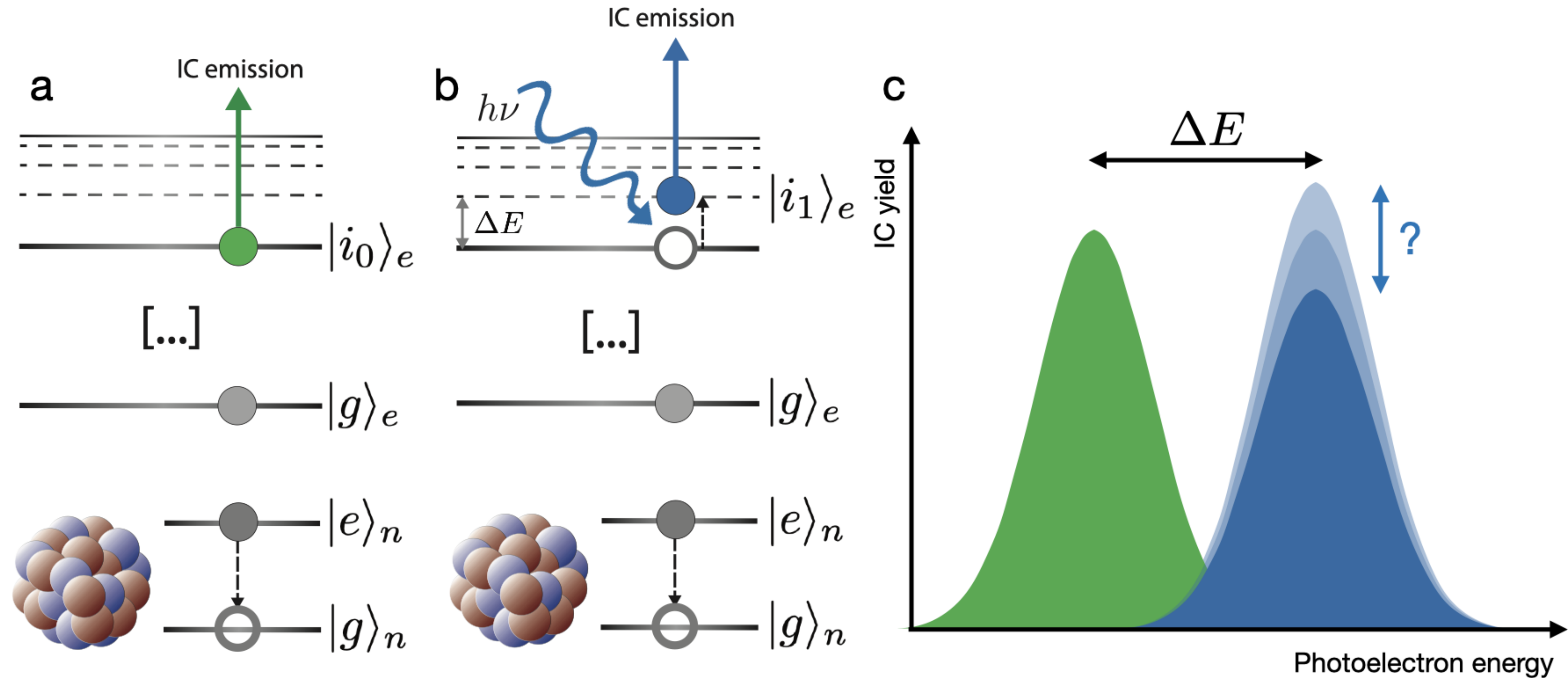
CEMS: A. V. K. Nomura and Y. Ujihira, J. Radiol. Nucl. Chem. 202, 103 (1996).

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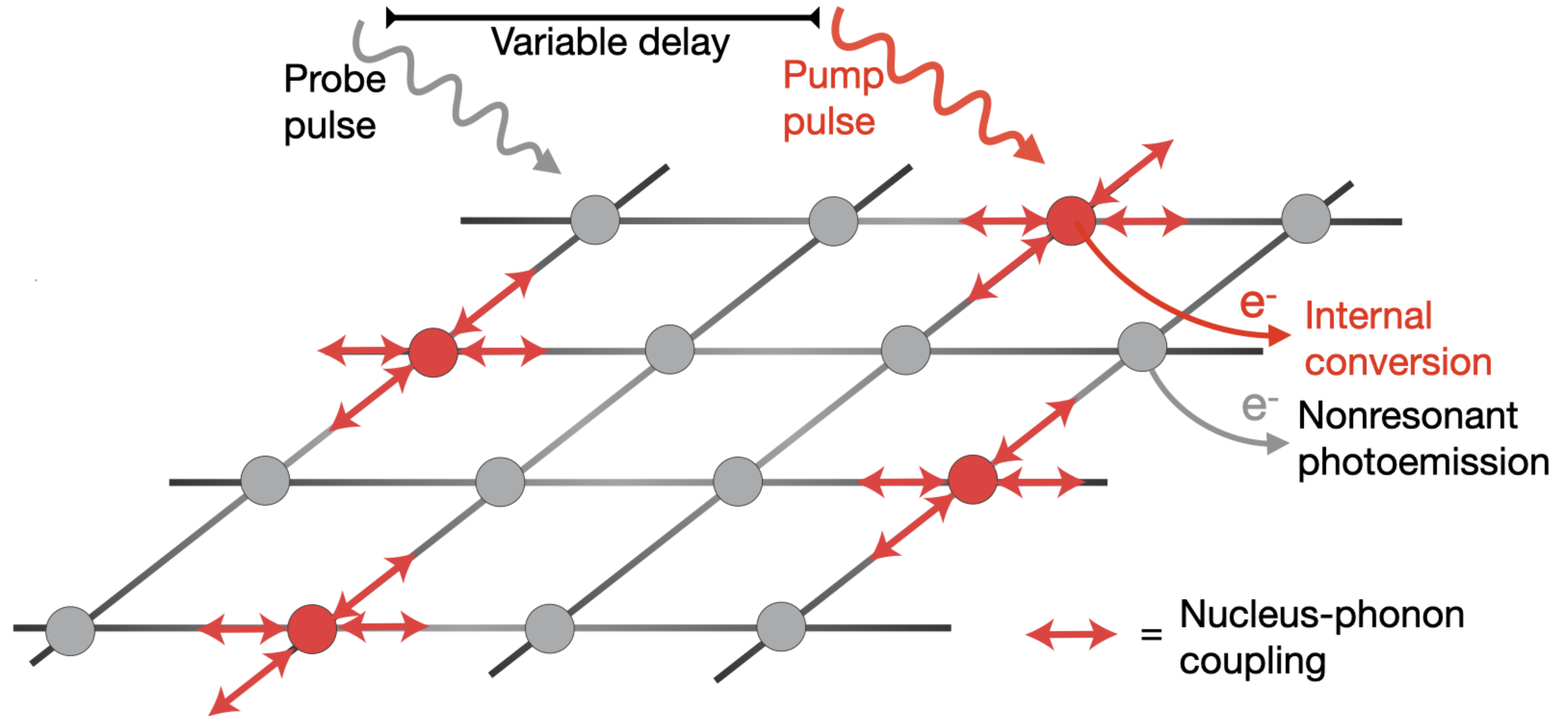
Be7@C60: T. Ohtsuki, et al., Phys. Rev. Lett. 93, 112501 (2004)



Examples of photo-controlled electron-nucleus couplings



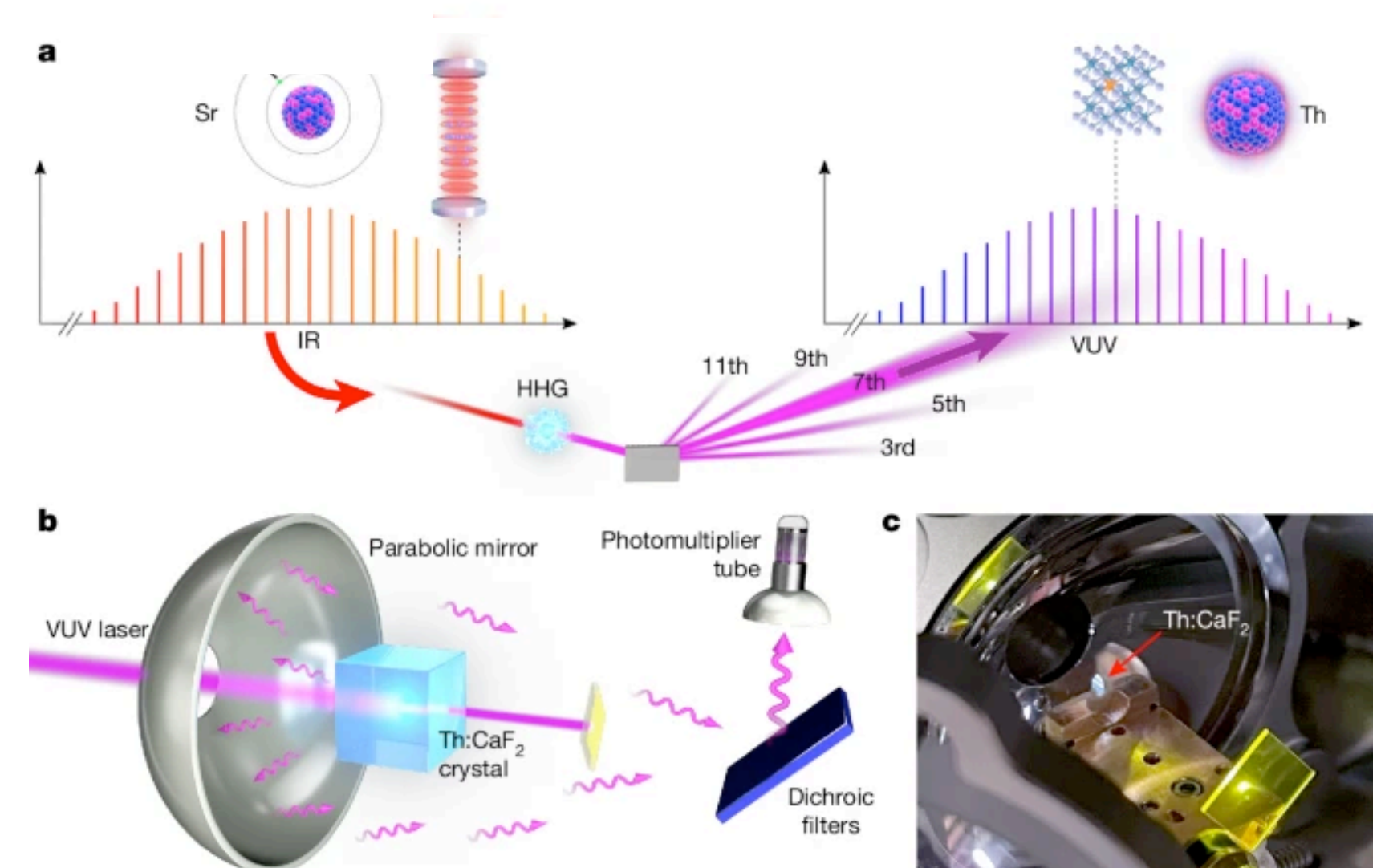
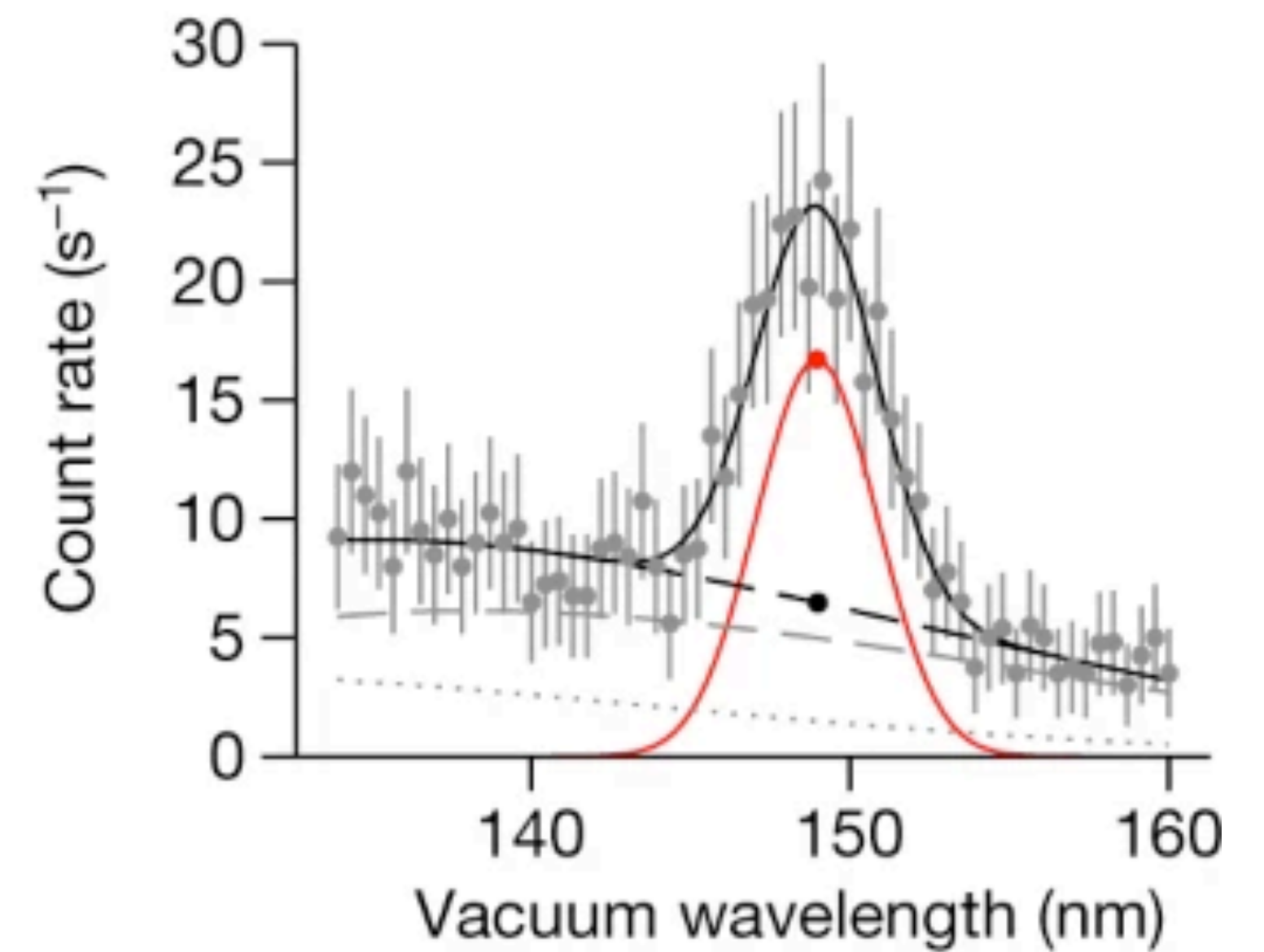
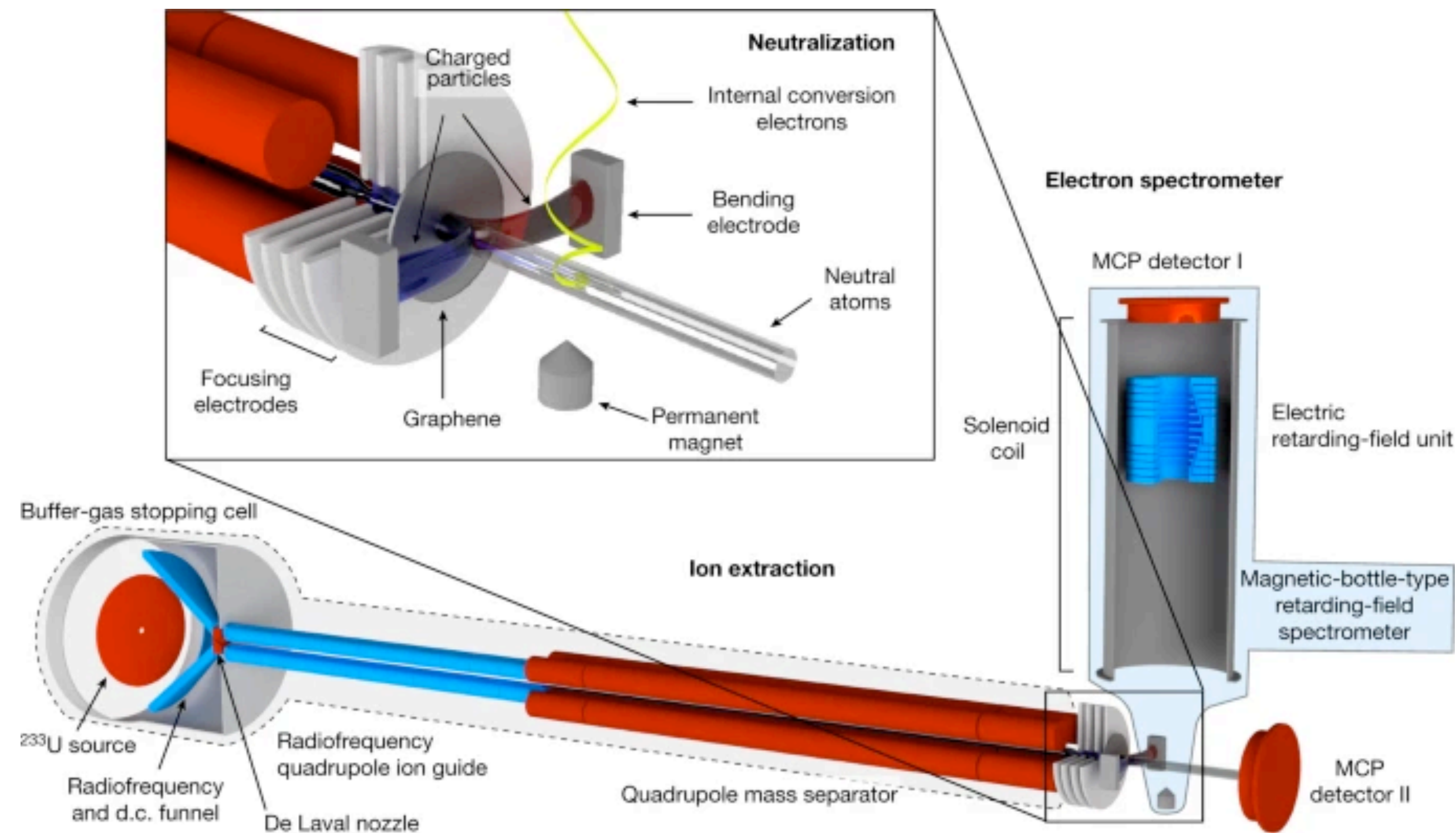
Examples of photo-controlled electron-nucleus couplings



M. Seitz, F. Calegari, P. G. Thirolf, A. Trabattoni, Towards the time-resolved spectroscopy of photoinduced electron dynamics in nuclear transitions, Phys. Rev. A., Accepted (2025). <https://doi.org/10.1103/31qf-pfvj>

State of the art: photoexcitation of electron–nucleus couplings

Thorium-229 $E \simeq 8.356$ eV, $\alpha = \Gamma_e/\Gamma_\gamma = 10^9$ (neutral atoms)



von der Wense, L., et al., Nature 533, 47–51 (2016)

B. Seiferle, et al., Nature 573, 243 (2019)

Kraemer, S., et al. Nature 617, 706–710 (2023)

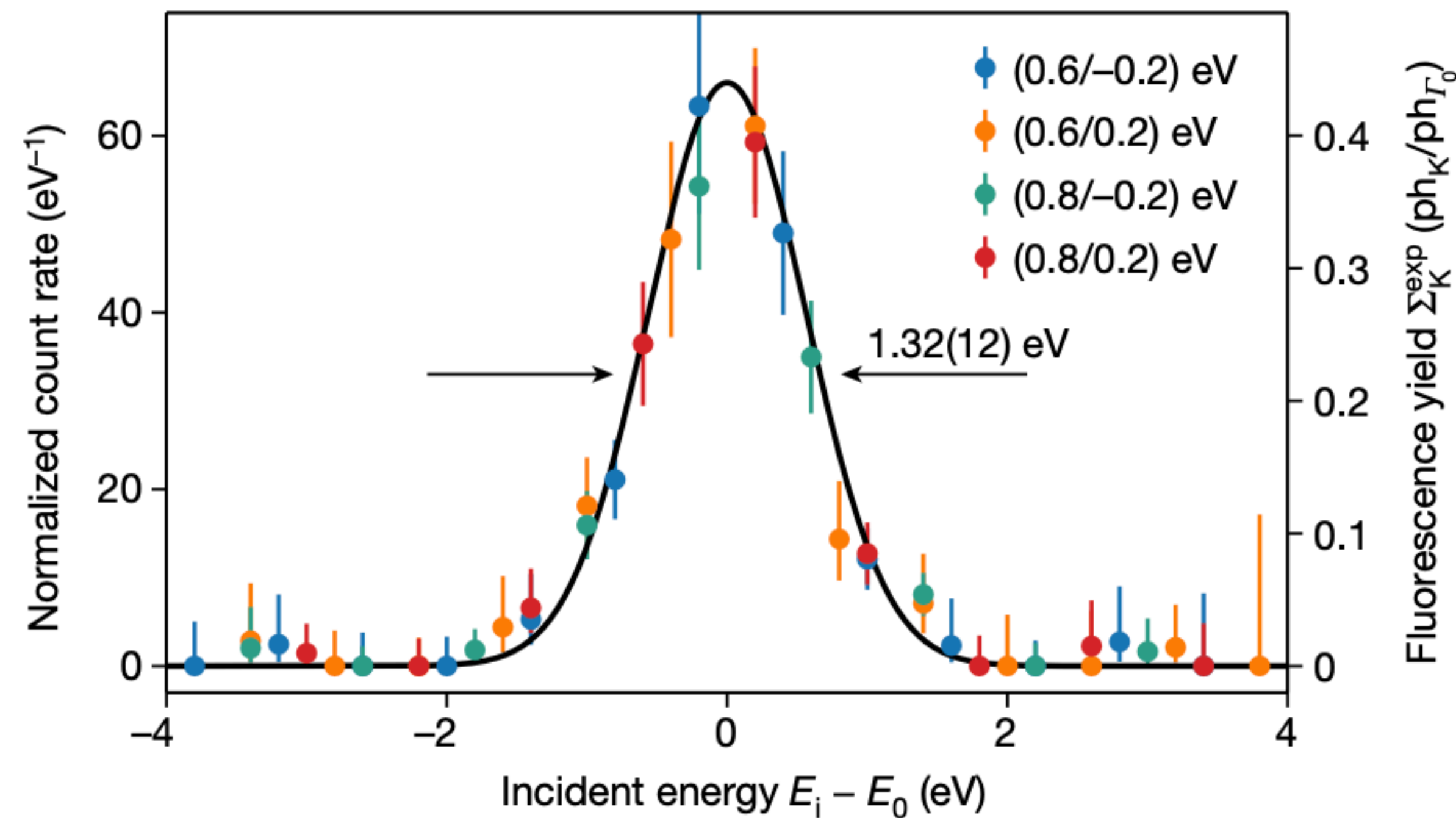
J. Tiedau, et al., Phys. Rev. Lett. 132, 182501 (2024)

Zhang, C., et al. Nature 633, 63–70 (2024)

State of the art: nuclear transitions can be photo-excited

Scandium-45

Measurement at European XFEL, Hamburg



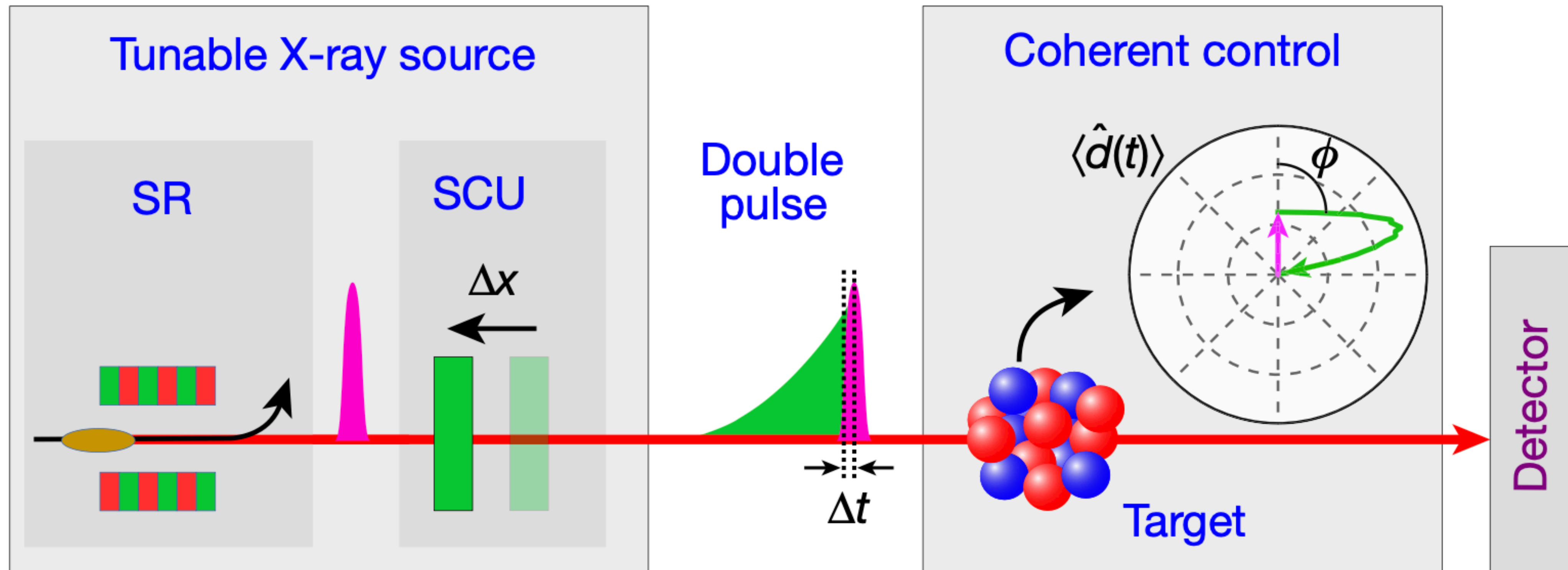
$\lambda \simeq 0.1 \text{ nm}, E \simeq 12.4 \text{ keV}$

Parameter	Notation	Magnitude	References
Ground state			
Spin and parity	I_g^π	$7/2^-$	
Magnetic moment	μ_g	$+4.756487(2)\mu_N$	
Quadrupole moment	Q_g	$-0.22(1)\text{b}$	
Isotopic abundance	η	100%	
Excited state			
Energy	E_0	$12,400(50)\text{eV}$ $12,389.59^{+0.15(\text{stat})}_{+0.12(\text{syst})}\text{eV}$	32,49,50
Lifetime	τ_0	$470(6)\text{ms}$	10,32,51
Natural linewidth	$\Gamma_0=\hbar/\tau_0$	$1.40(2)\text{feV}$	
Quality factor	$Q=E_0/\Gamma_0$	$8.85(10)\times 10^{18}$	
Multipolarity		M2	
Spin and parity	I_e^π	$3/2^+$	
Magnetic moment	μ_e	$+0.368(5)\mu_N$	52
Quadrupole moment	Q_e	$+0.318(22)\text{b}$	52
Internal conversion coefficients	α, α_K	$632(71), 474$	32,51,53
		424, 363	38
Cross-section	σ_0 in equation (3)	$1.26(15)\times 10^{-20}\text{cm}^2$	
		$1.9(5)\times 10^{-20}\text{cm}^2$	
Wavelength corresponding to measured ^{45}Sc energy: $\lambda_0=1.00071\text{\AA}$.			

State of the art: nuclear transitions can be photo-excited

Fe-57

Measurement at European Synchrotron Radiation Facility (ESRF), Grenoble



$$\lambda \simeq 0.08 \text{ nm}, E \simeq 14.4 \text{ keV}, \alpha \simeq 10$$

K. P. Heeg, et al., Nature 590, 401 (2021)

D.P.E. Dickson and F.J. Berry, Mössbauer Spectroscopy, Cambridge University Press, Cambridge, (1986)

Two examples

Energy-resolved/time-resolved photoelectron spectroscopy on ^{57}Fe

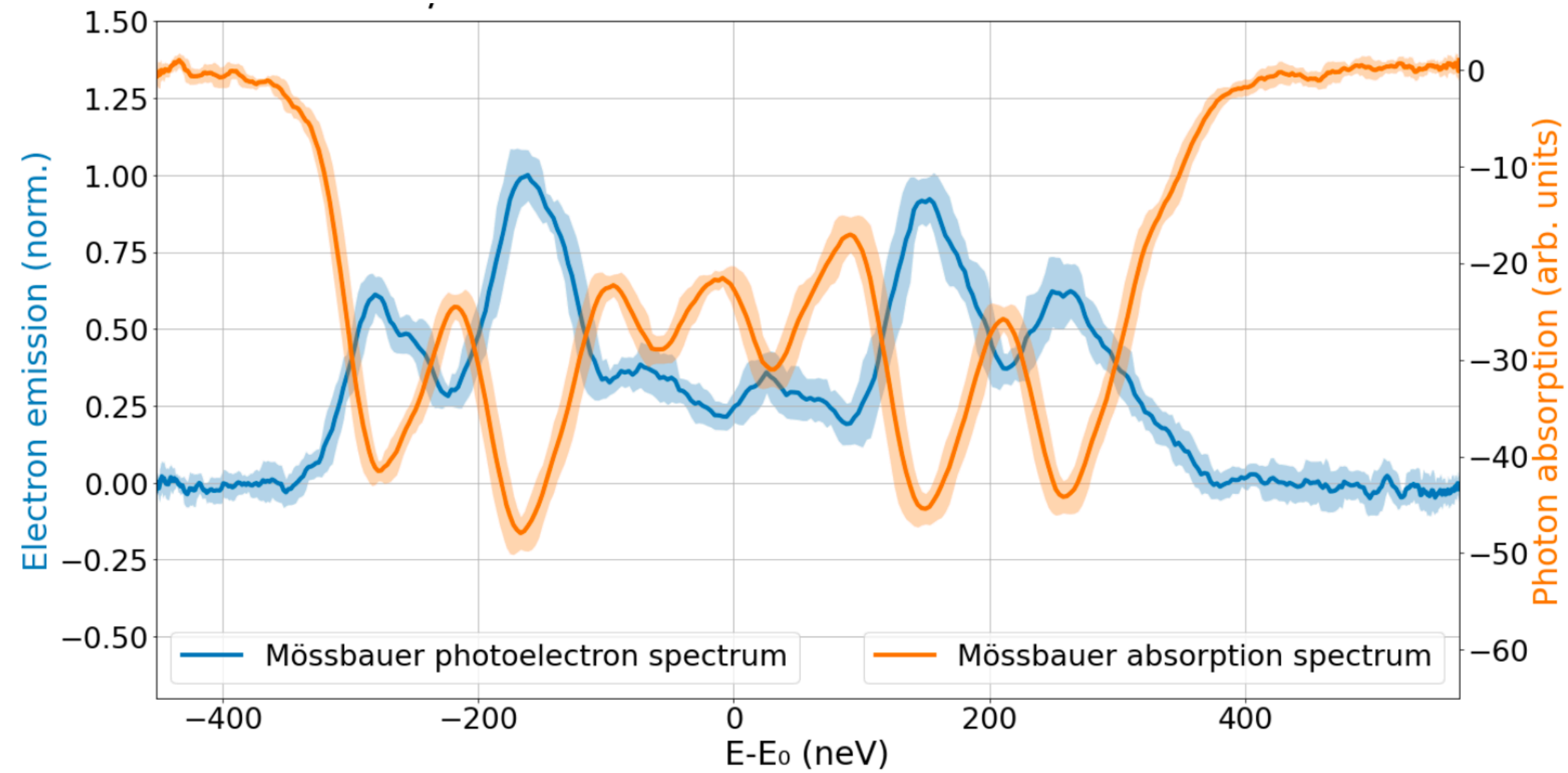
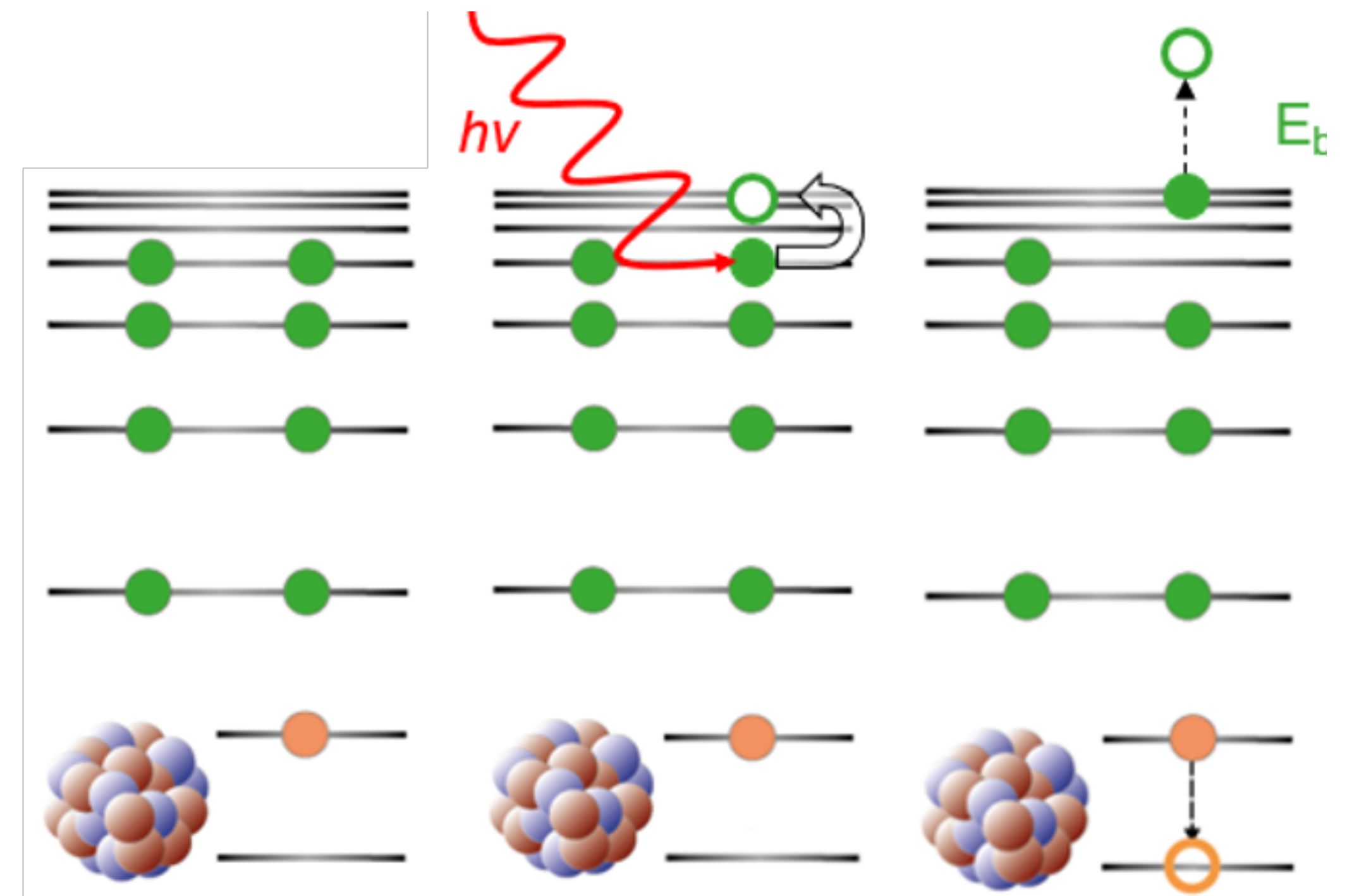
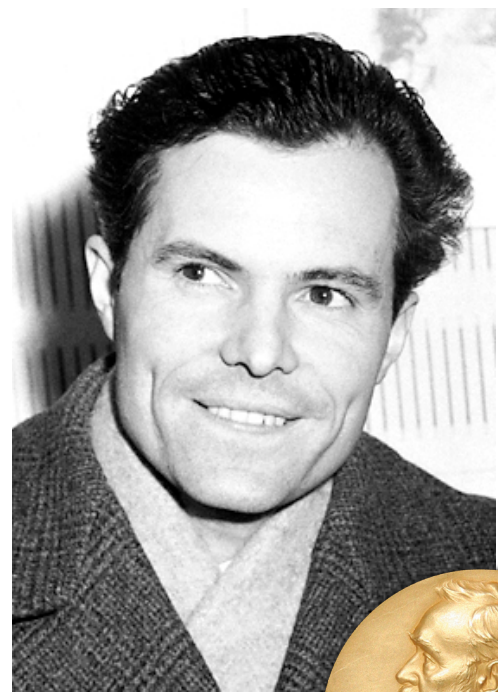
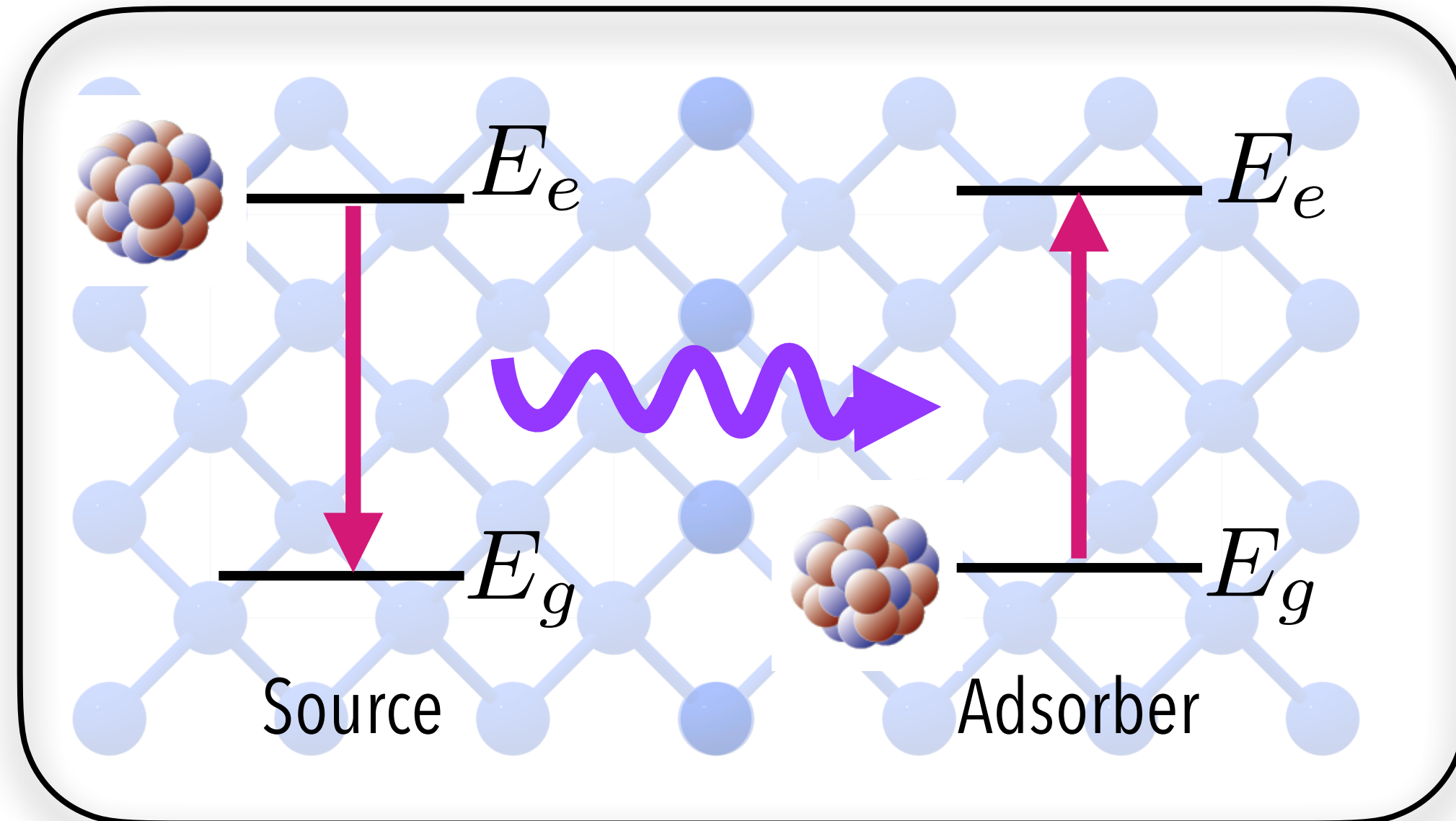


Photo-perturbation of IC in $^{229\text{m}}\text{Th}$



Mössbauer spectroscopy

Recoilless nuclear resonant excitation/emission

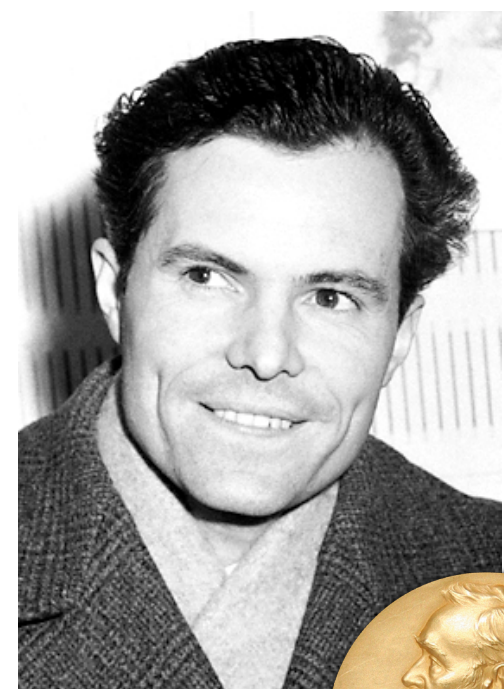
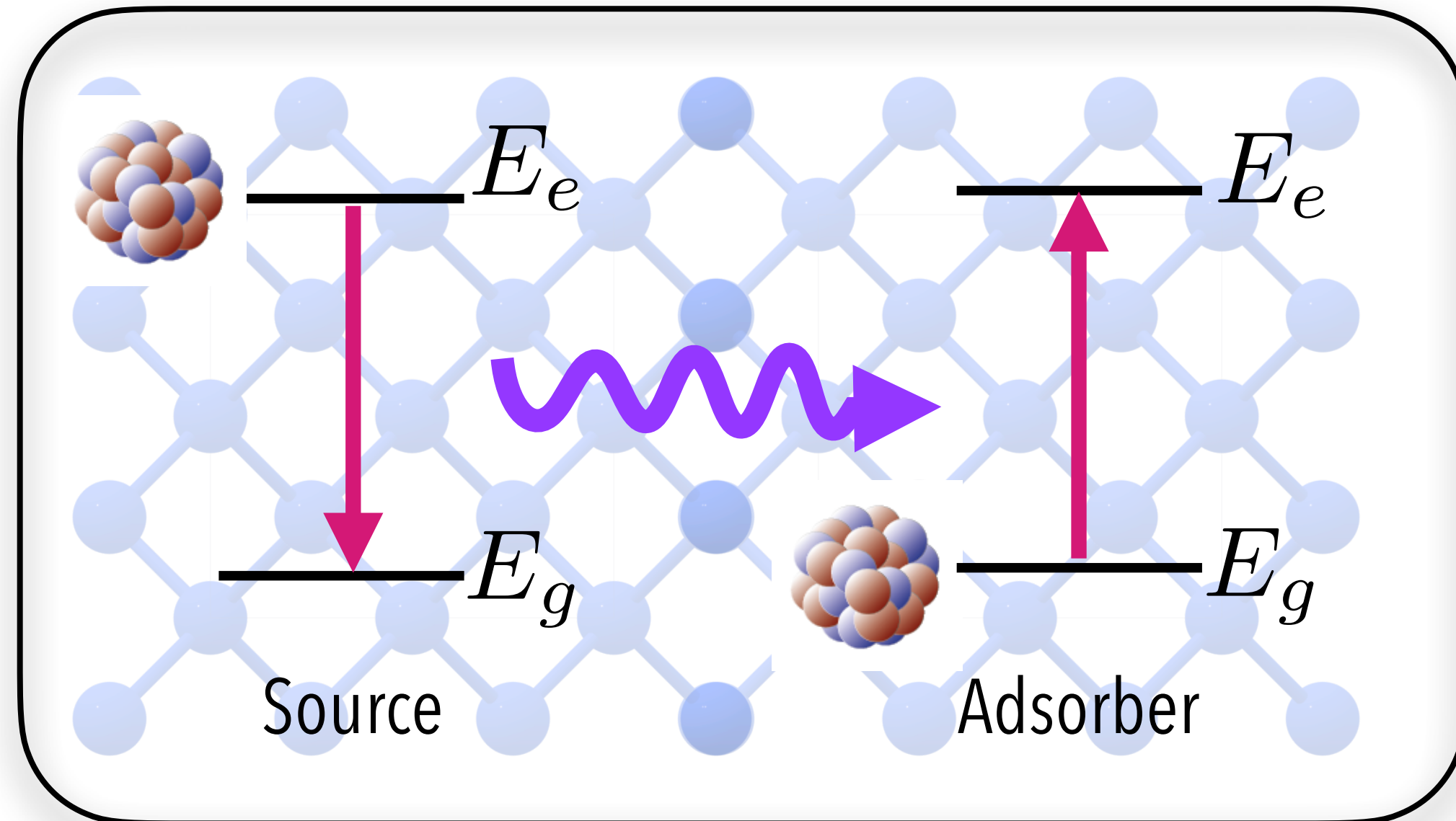


R.L. Mössbauer, Z. Physik, 1958, 151, 124
Nobel Prize for Physics in 1961



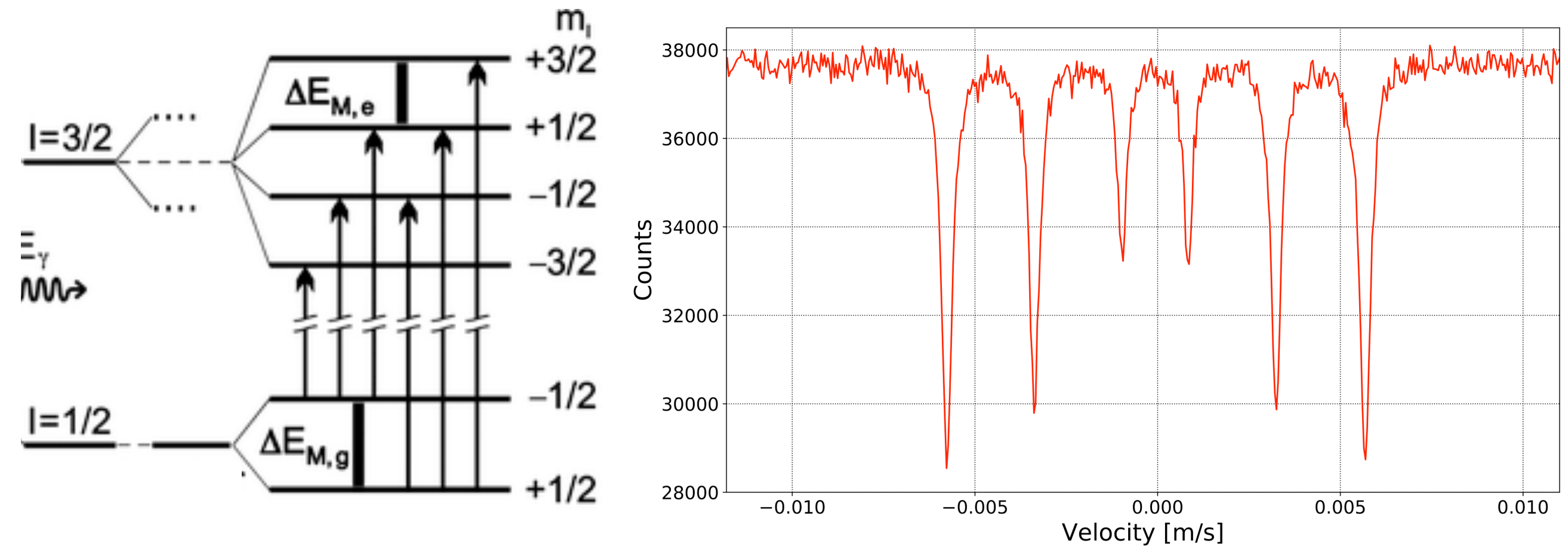
Mössbauer spectroscopy

Recoilless nuclear resonant excitation/emission

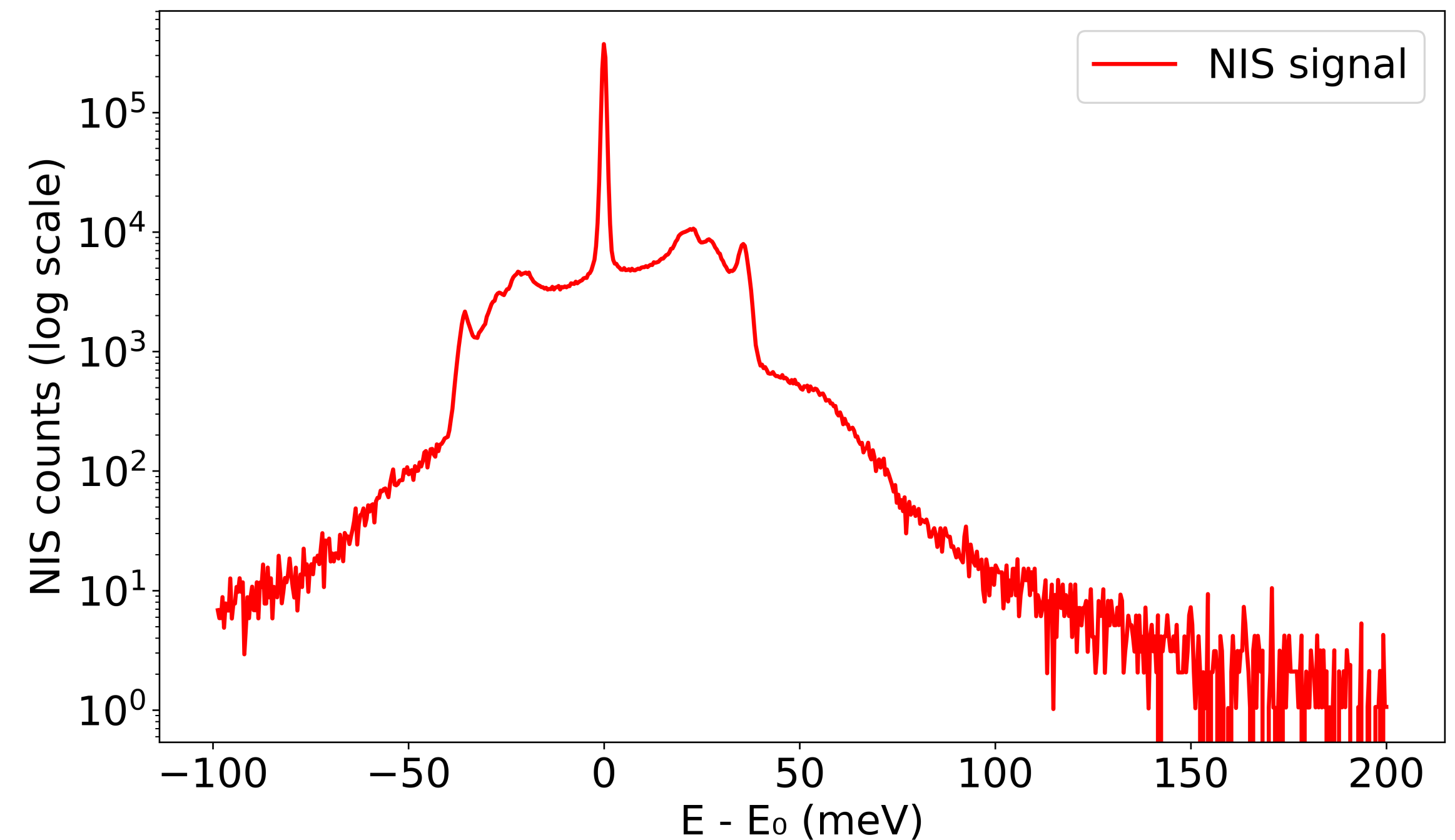


R.L. Mössbauer, Z. Physik, 1958, 151, 124
Nobel Prize for Physics in 1961

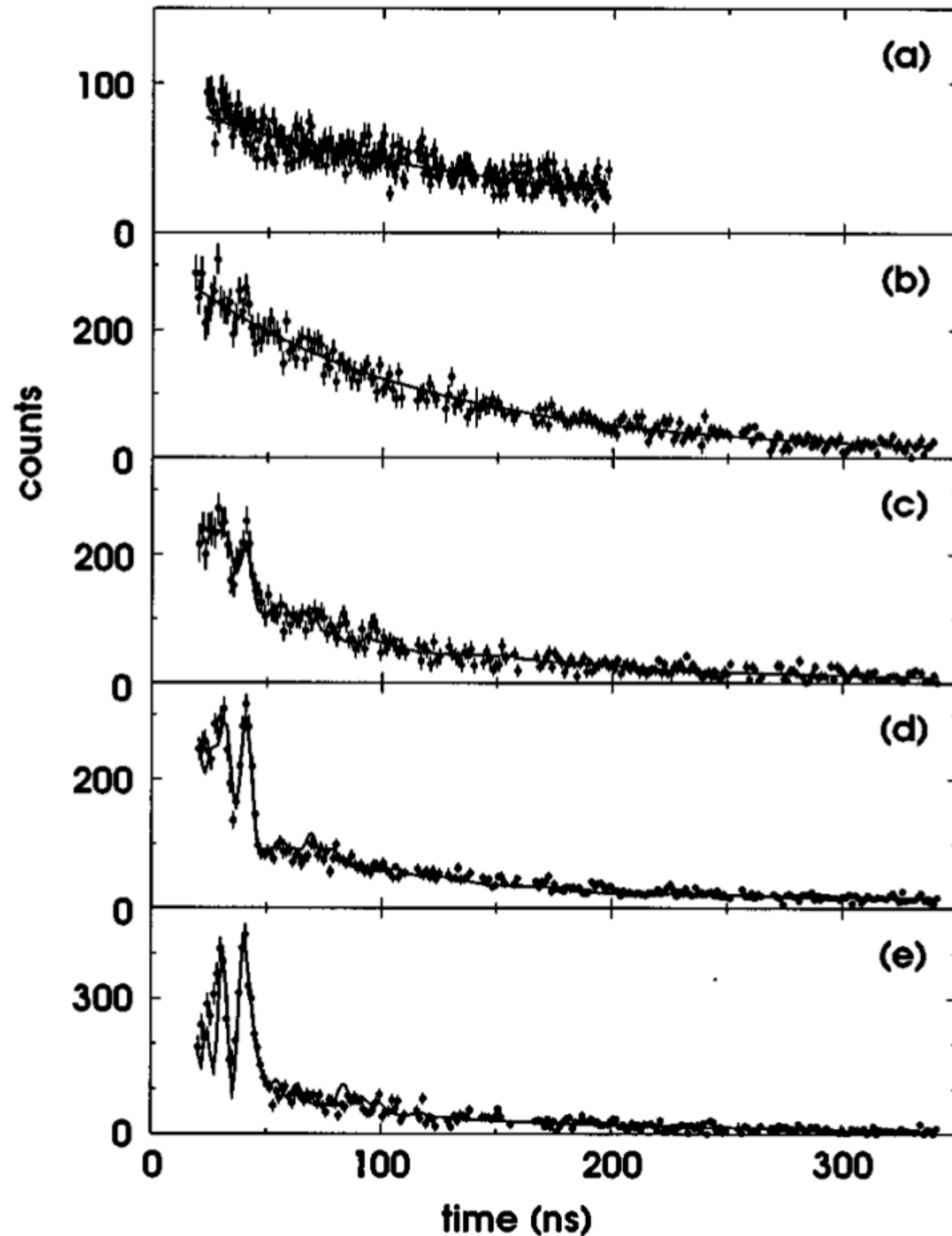
Mössbauer Spectrum



Inelastic nuclear scattering spectrum



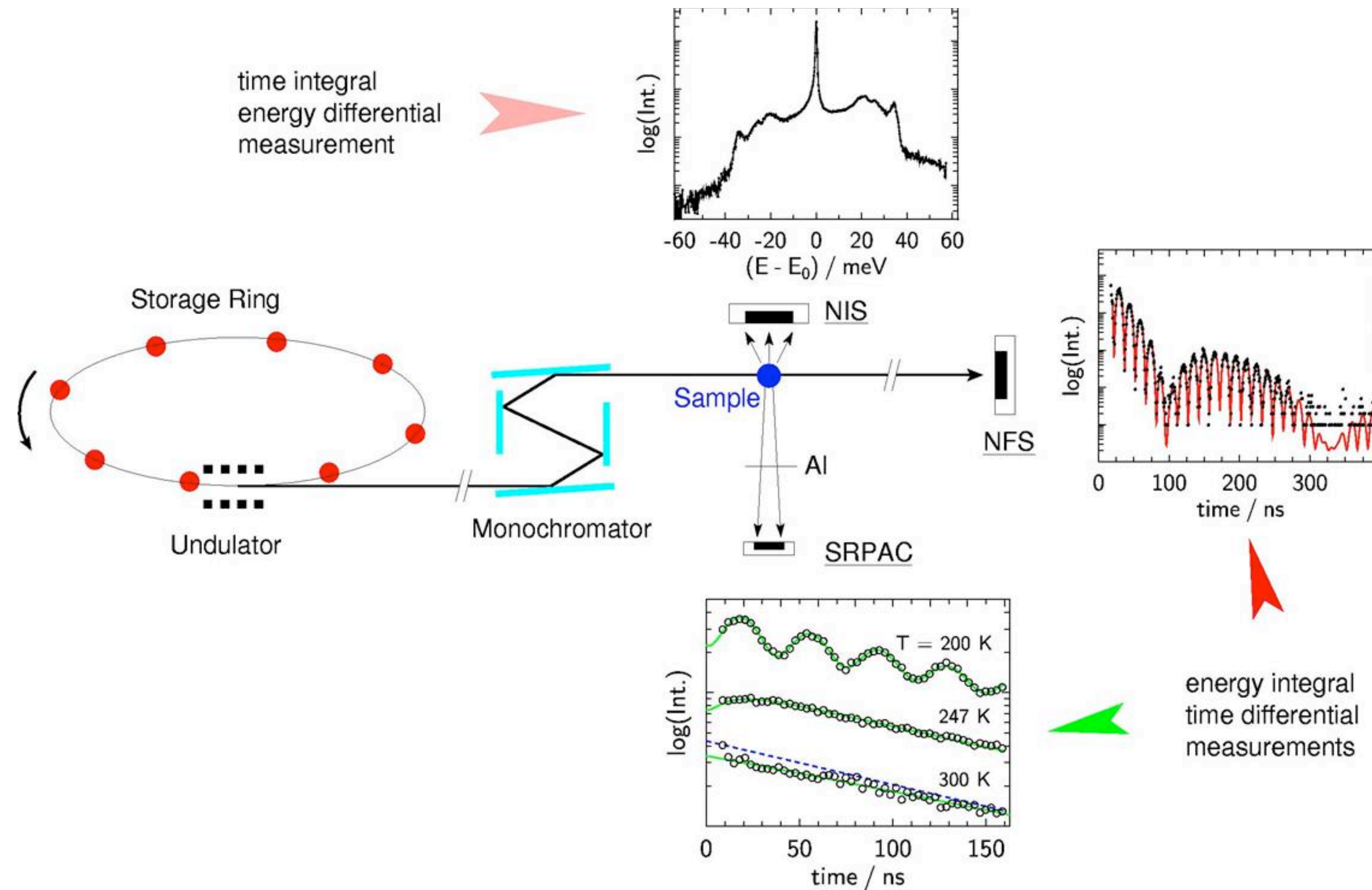
Conversion electron Mössbauer spectroscopy



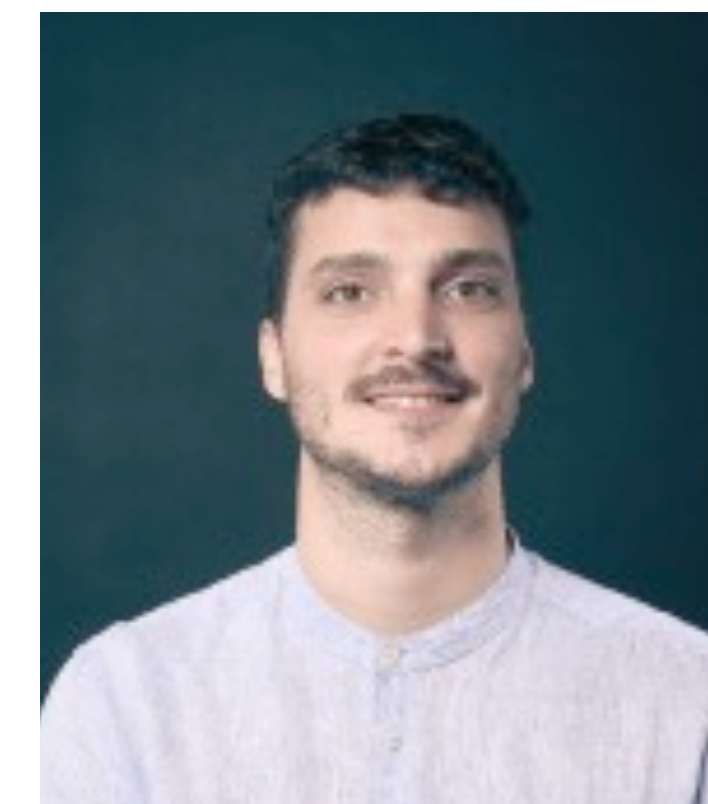
To our knowledge, the measurement of conversion electrons upon photo excitation by a synchrotron or a free electron laser is largely unexplored.

We tackled the challenge of developing new tools (and concepts) of photoelectrons spectroscopy of nuclear transitions.

P01 beamline @ Petra III, Hamburg



Sven Velten

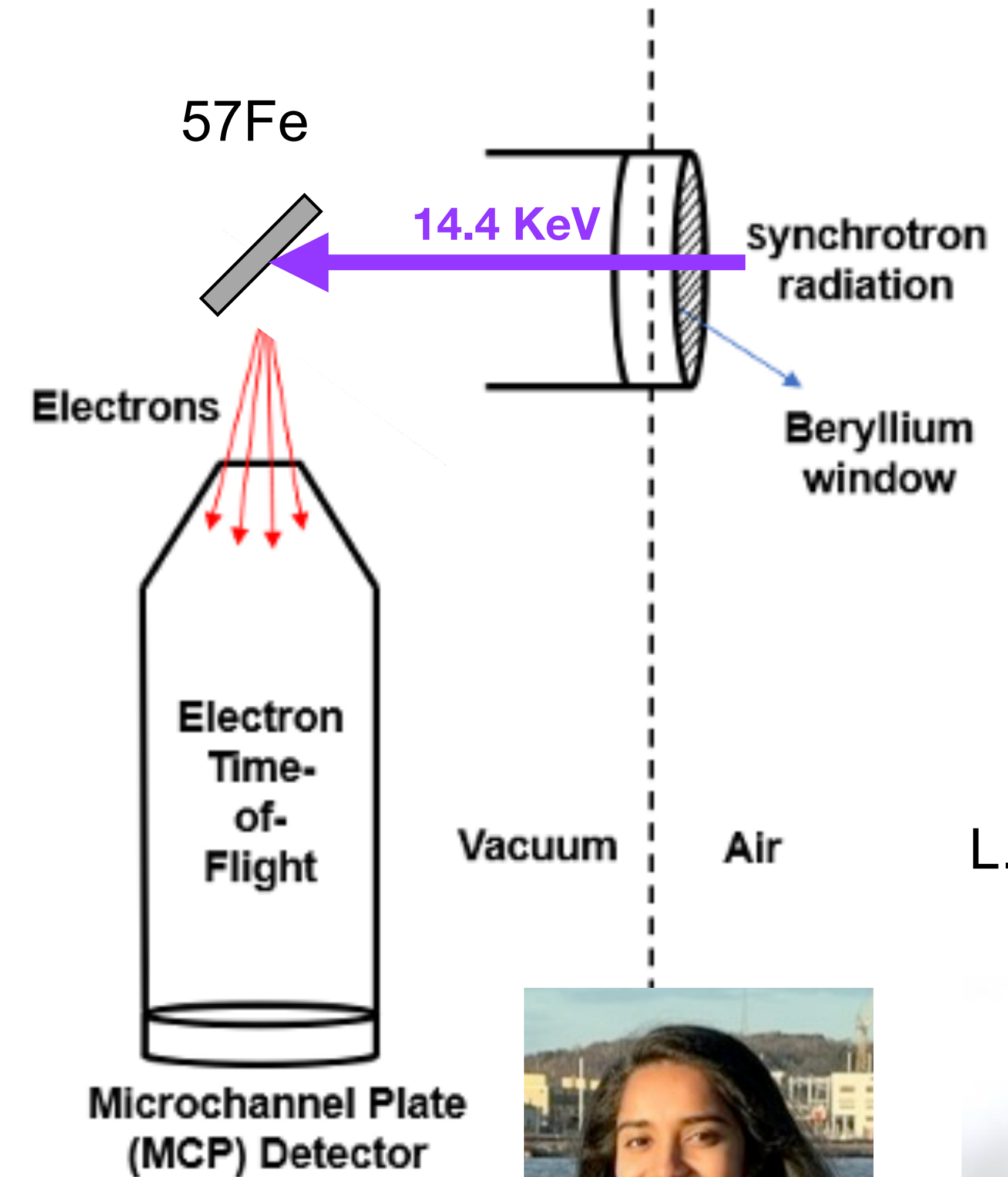


Ilya Sergeev

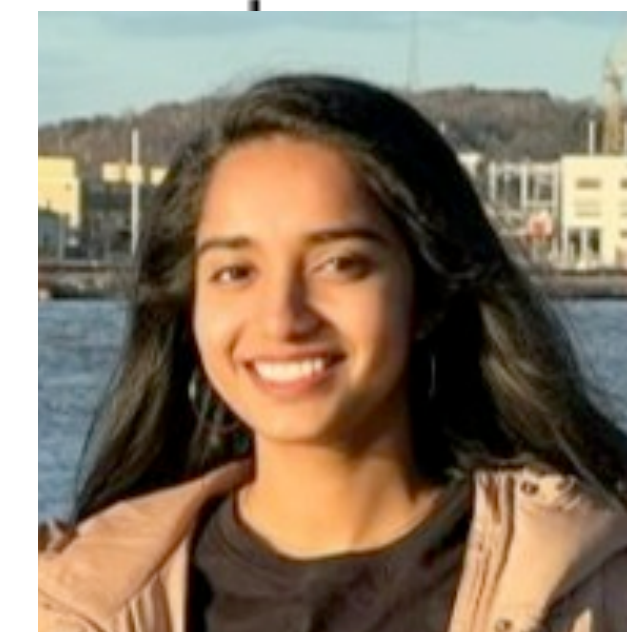


Nuclear Resonant Scattering station

The experimental setup - energy-resolved photoelectron spectroscopy



L. Bocklage

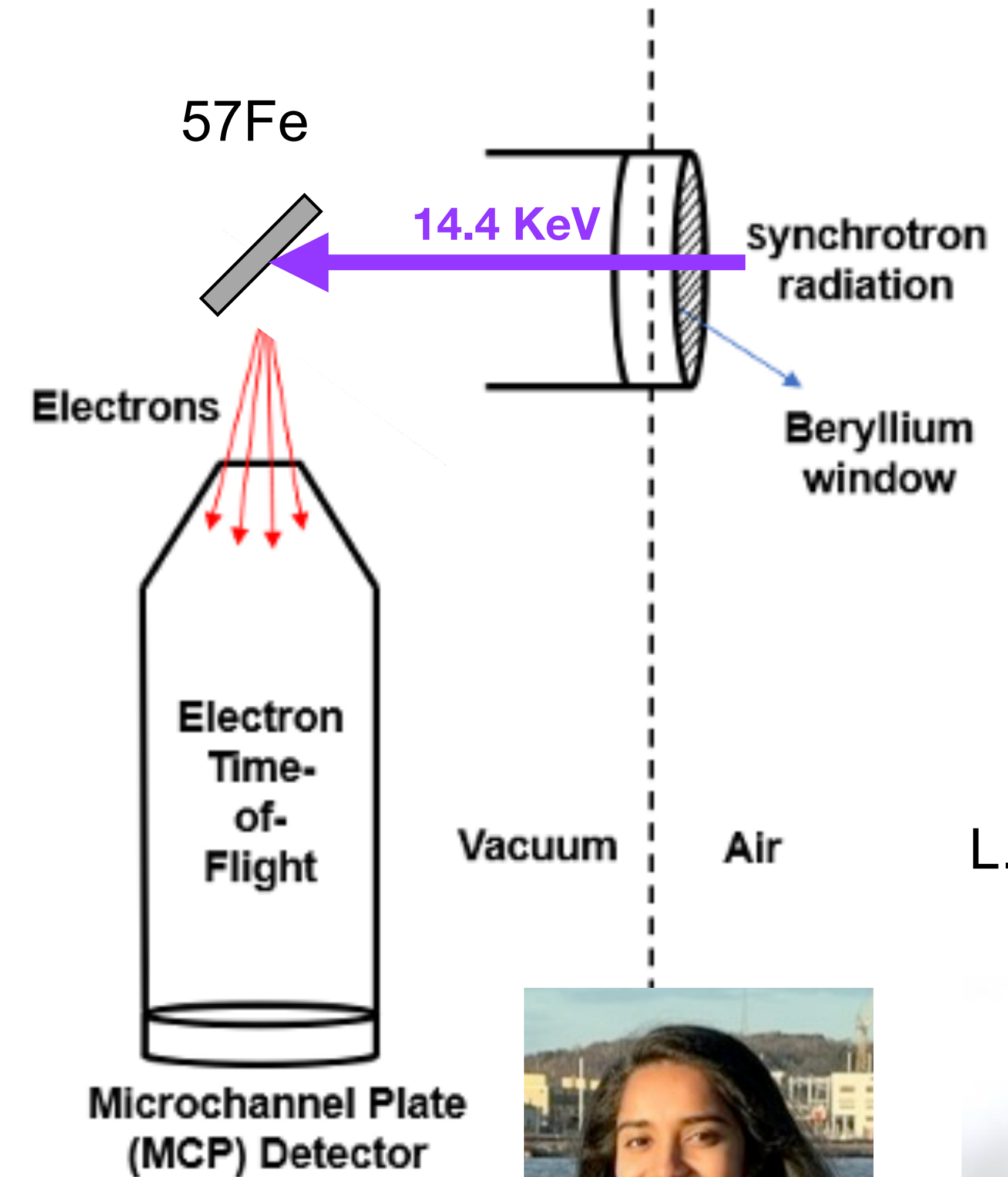
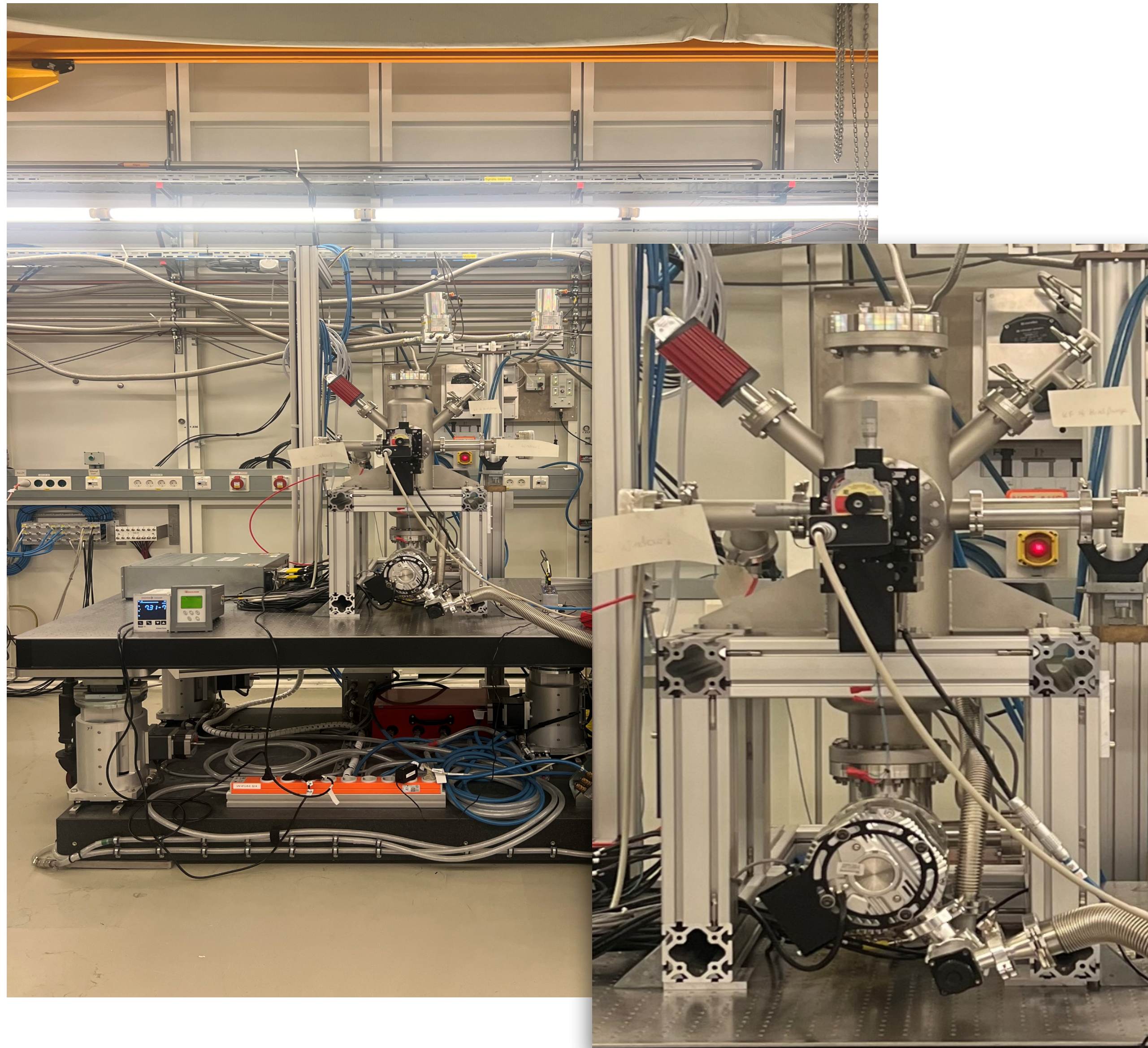


Keerthana Ravi



R. Röhlberger

The experimental setup - energy-resolved photoelectron spectroscopy



L. Bocklage

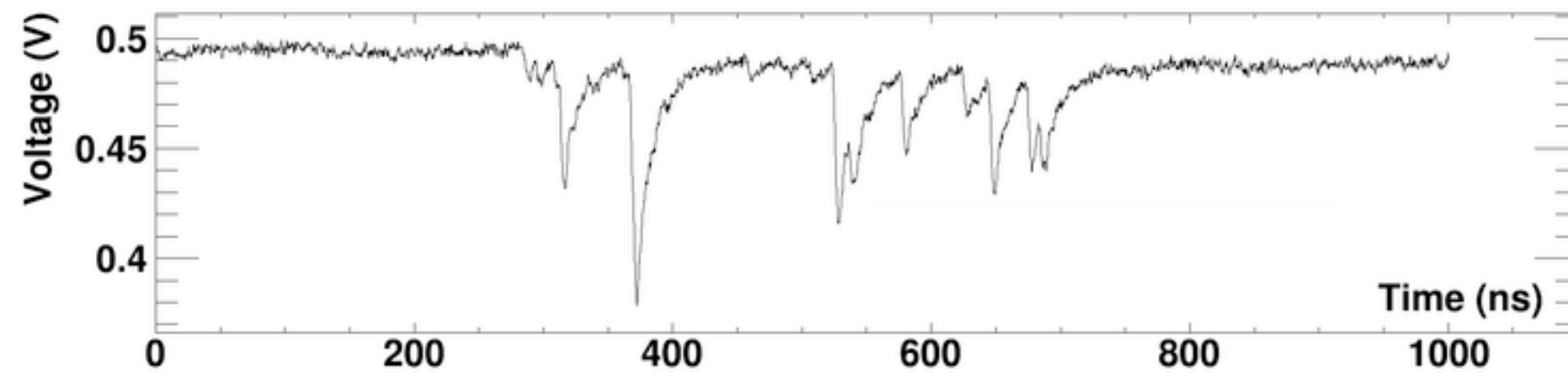


Keerthana Ravi



R. Röhlberger

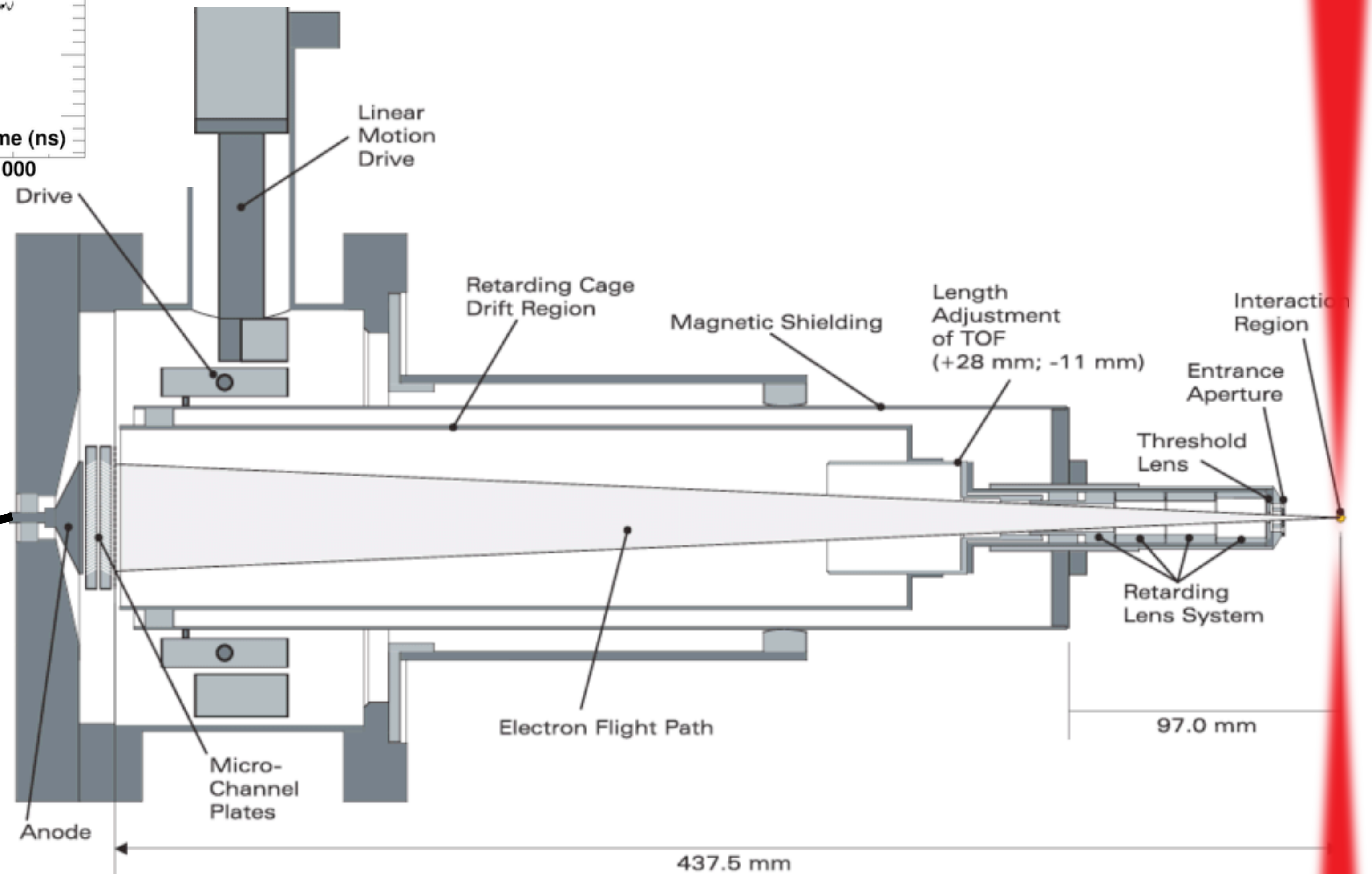
Electron time of flight spectrometer



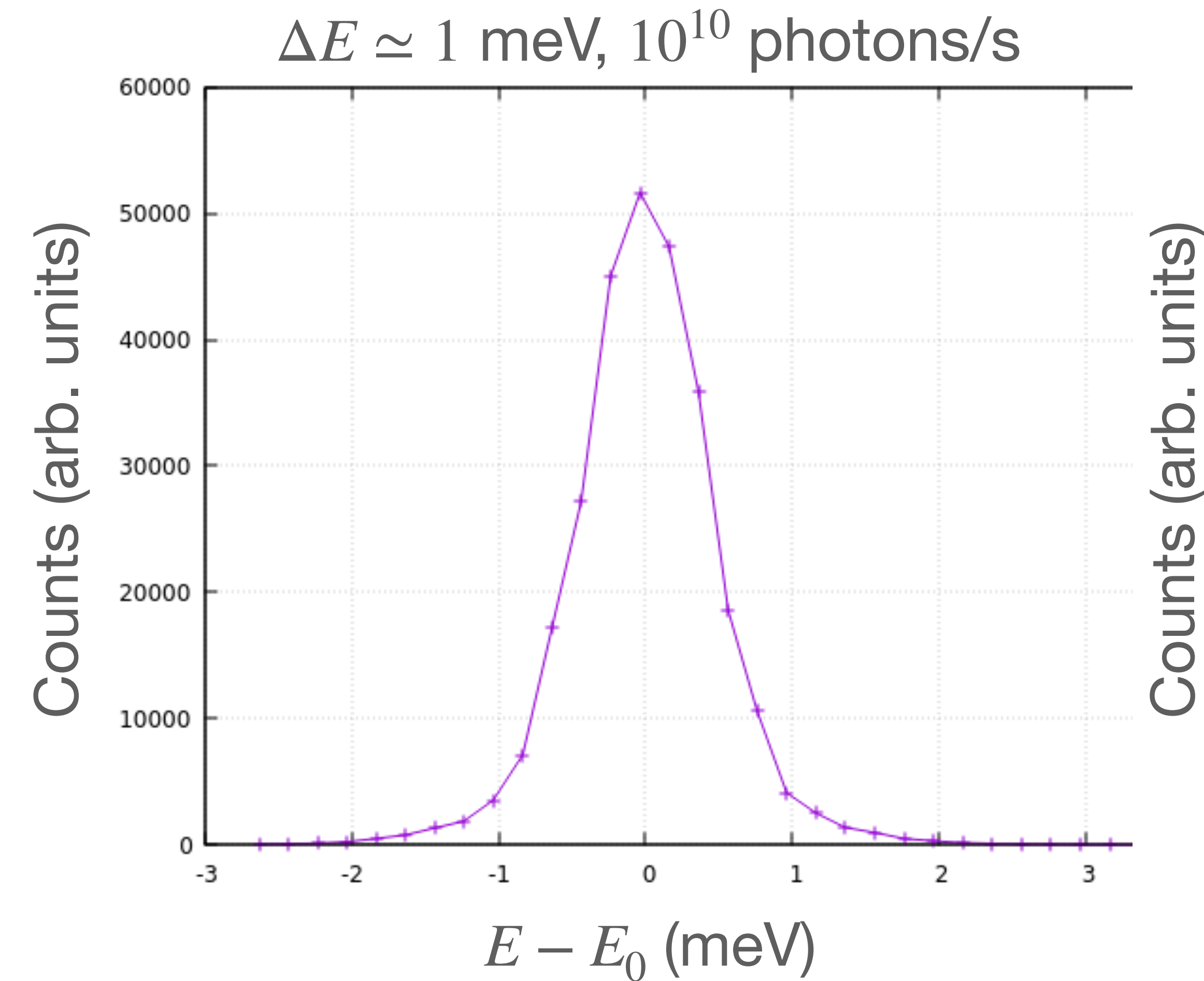
From time of flight to photoelectron kinetic energy

$$E_k = A(t - t_0)^{-2} + B(t - t_0)^2$$

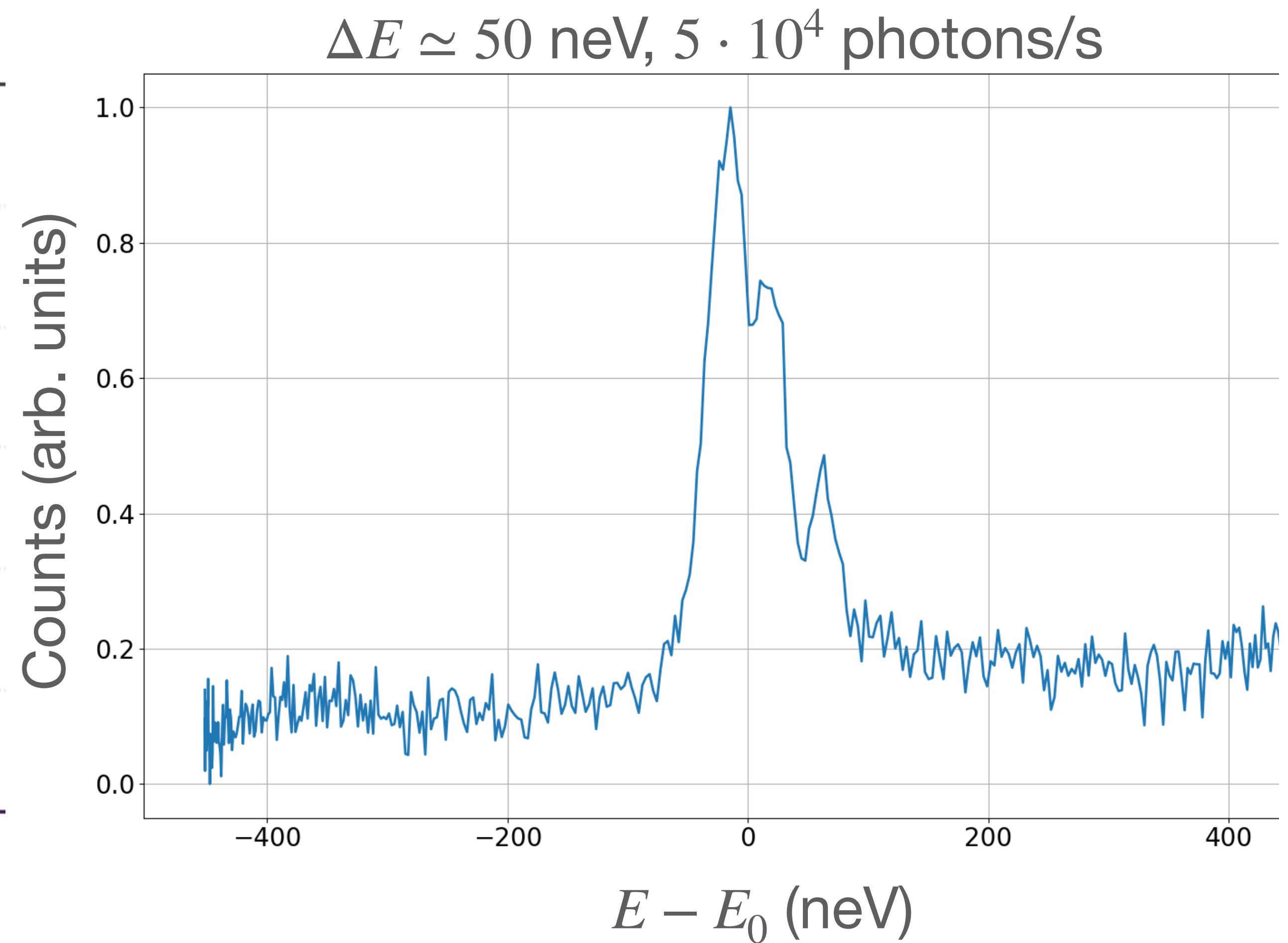
$$E_k = \frac{md^2}{2(t - t_0)^2} + \frac{E^2 e^2 (t - t_0)^2}{8m}$$



Highly monochromatized synchrotron source



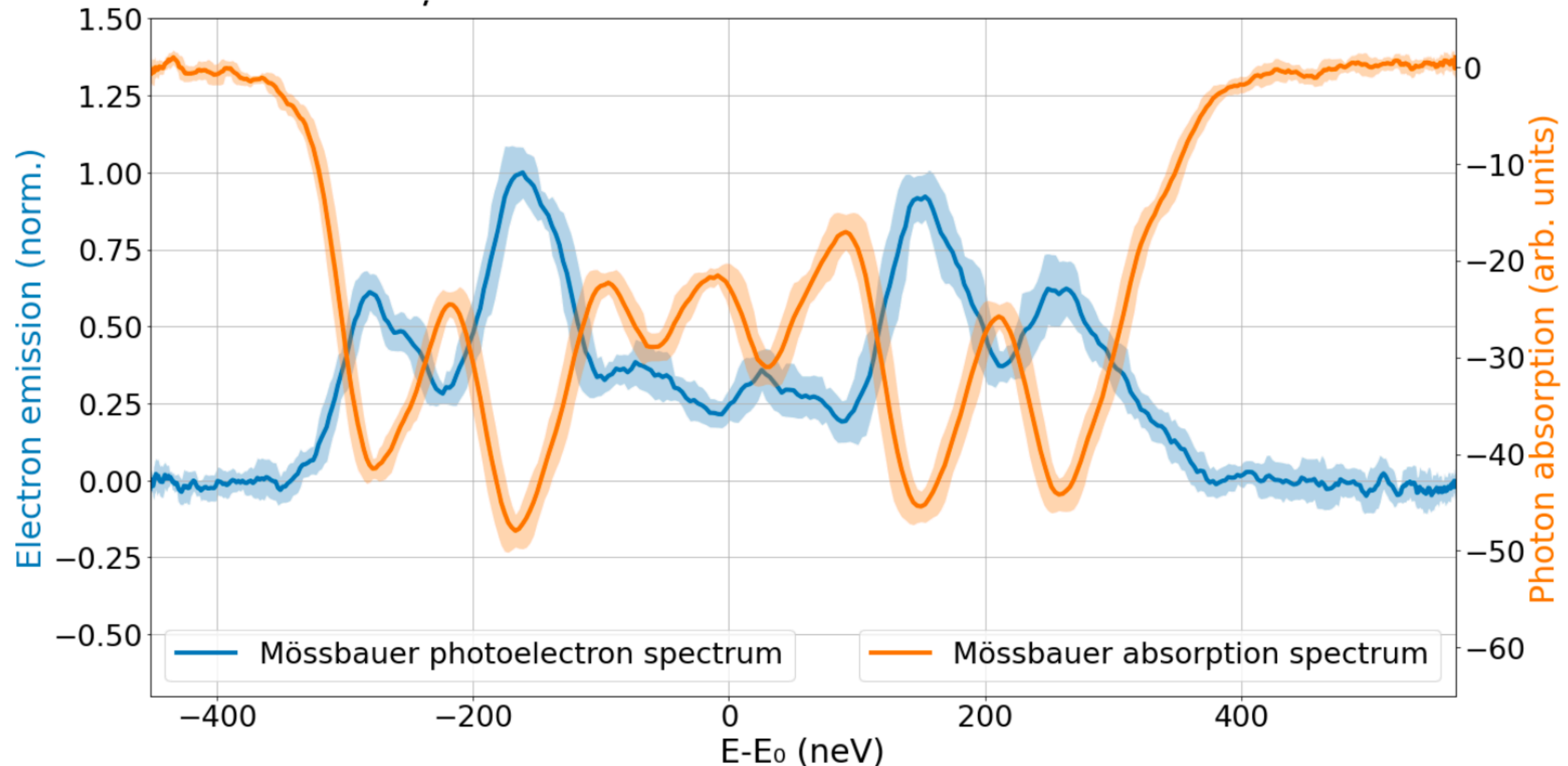
For inelastic spectrum measurements



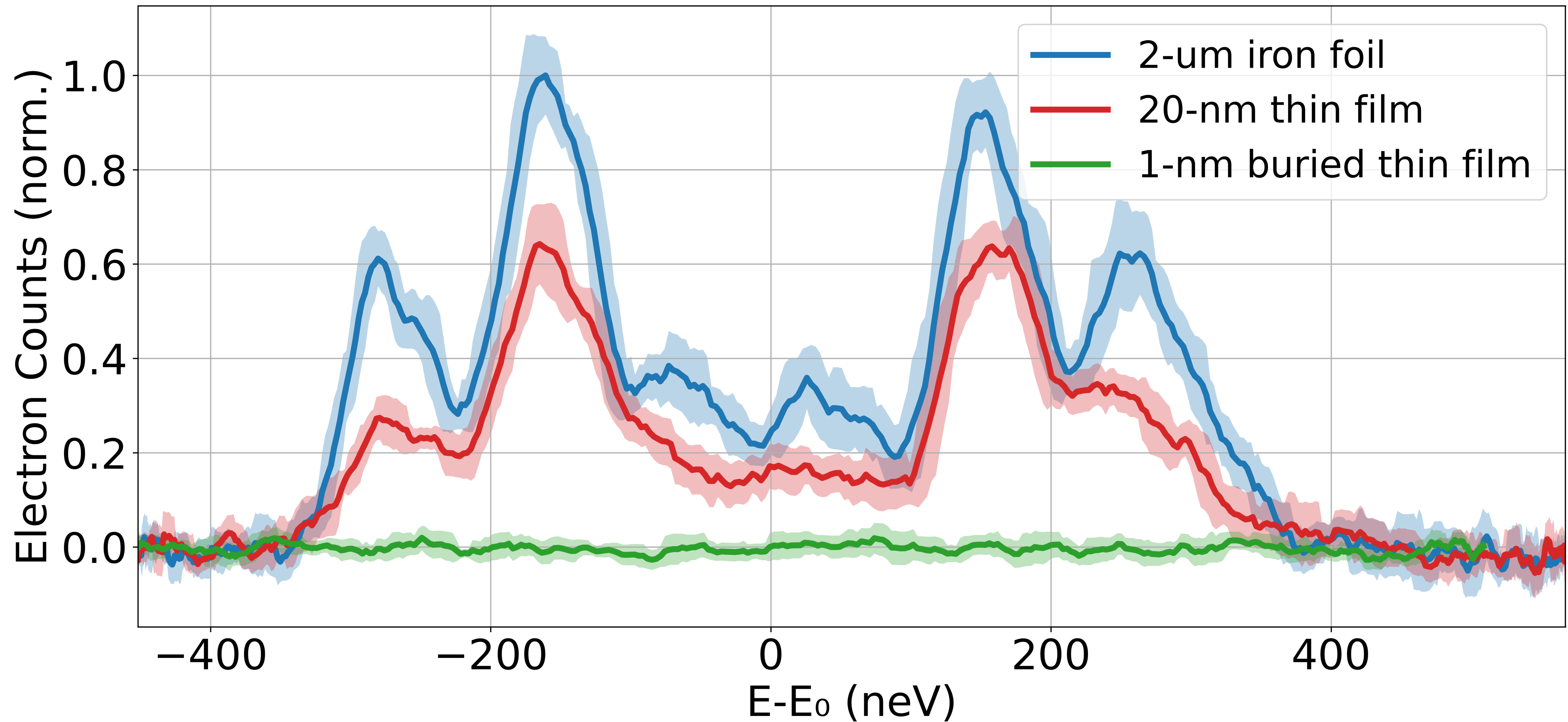
For hyperfine structure measurements

Hyperfine structure – photons vs. electrons

X-ray absorption spectrum measured with an APD detector, photoelectron yield measured with the eTOF

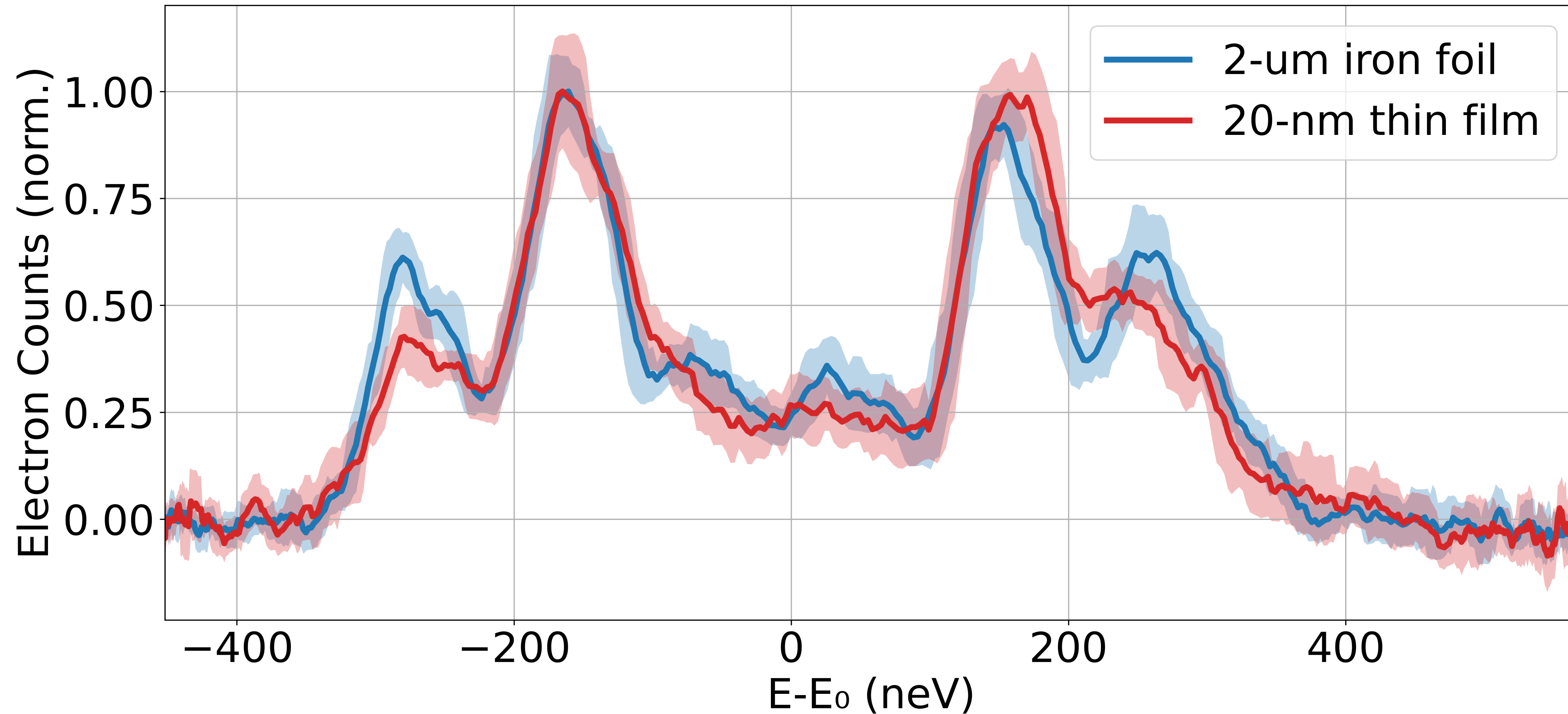


Hyperfine structure – Foil vs Thin Film



Hyperfine structure – Foil vs Thin Film

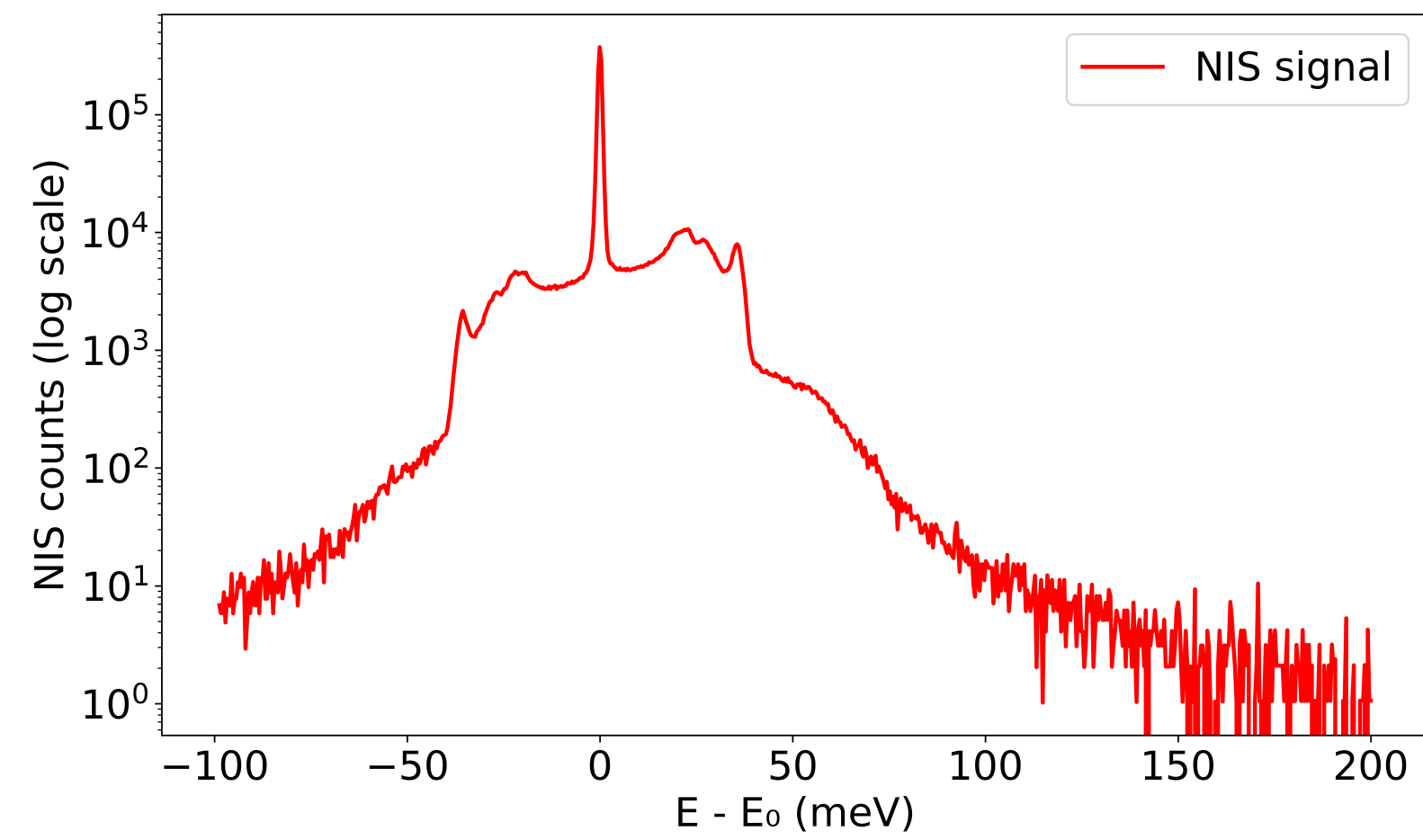
Signature of planar magnetisation at the surface for the 20-nm film



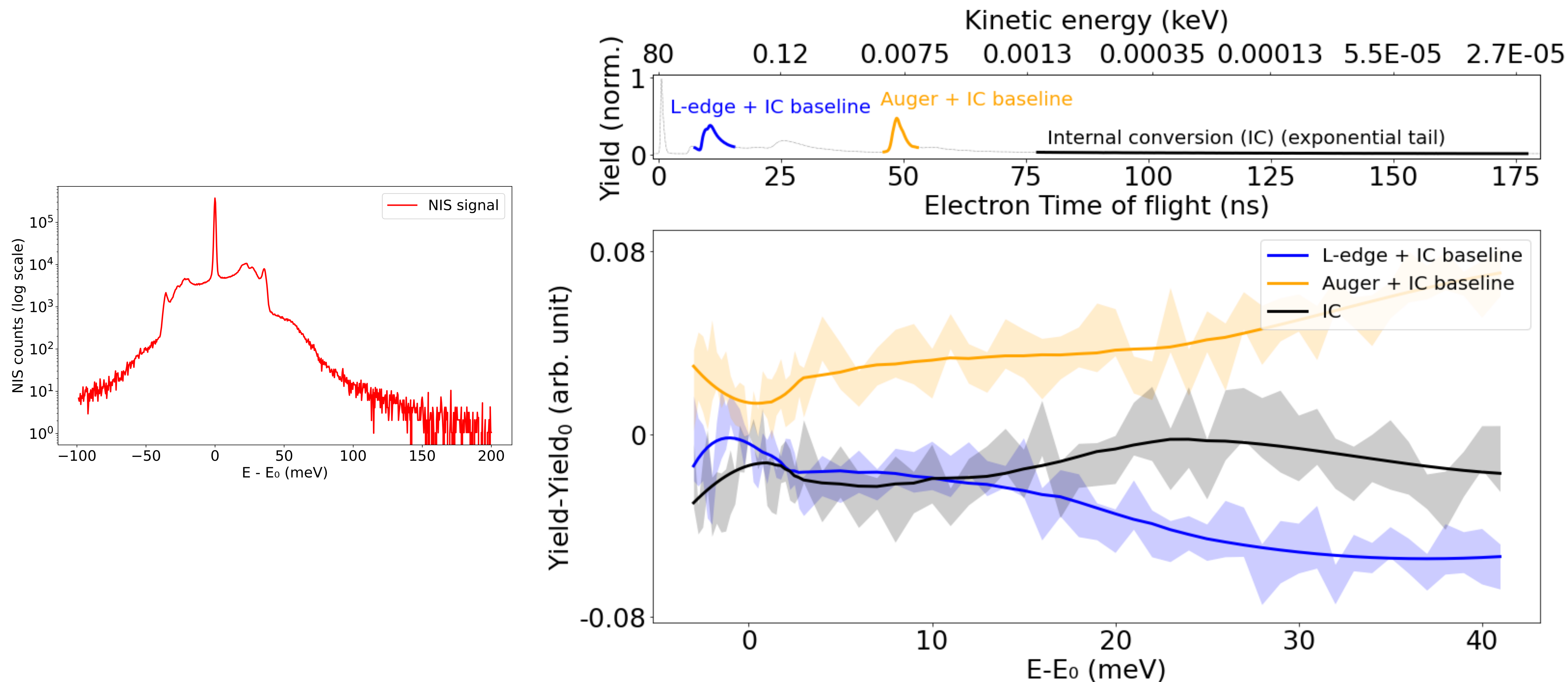
Ongoing analysis, simulations performed with NEXUS (Lars Bocklage, DESY): <https://helmholtz.software/software/nuclear-nexus>

What is the contribution of the different electronic states?

Inelastic spectrum – energy-resolved photoelectron spectrum

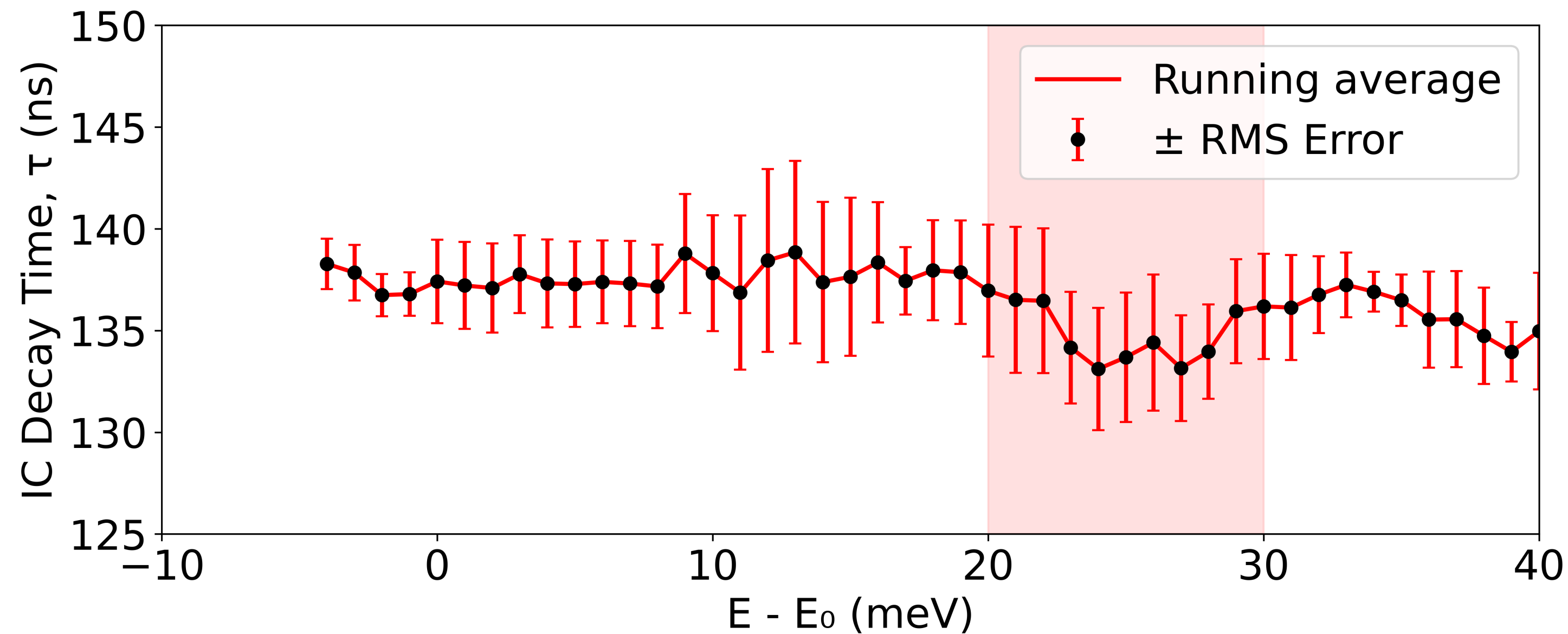


Inelastic spectrum – energy-resolved photoelectron spectrum

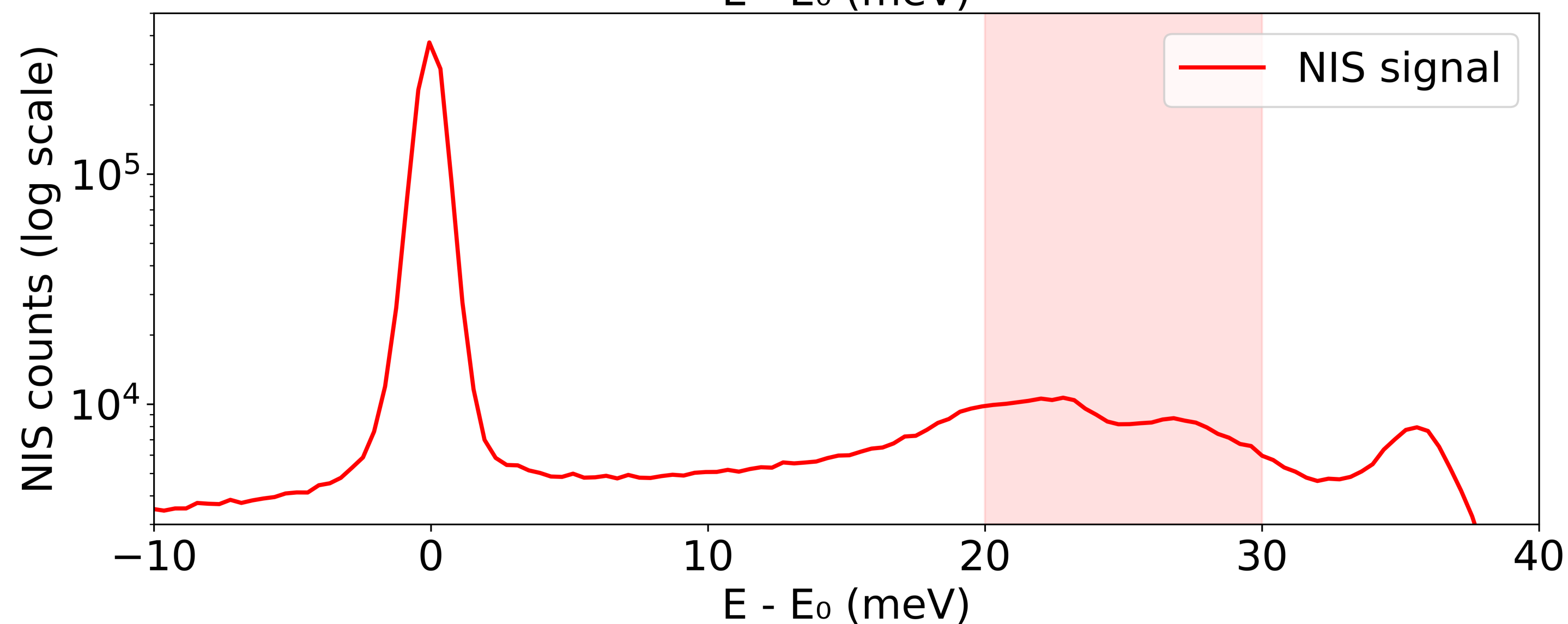


Coupling between photoemission and phonon modes. Population transfer between complementary channels.

Internal conversion decay time for the dressed nuclear state



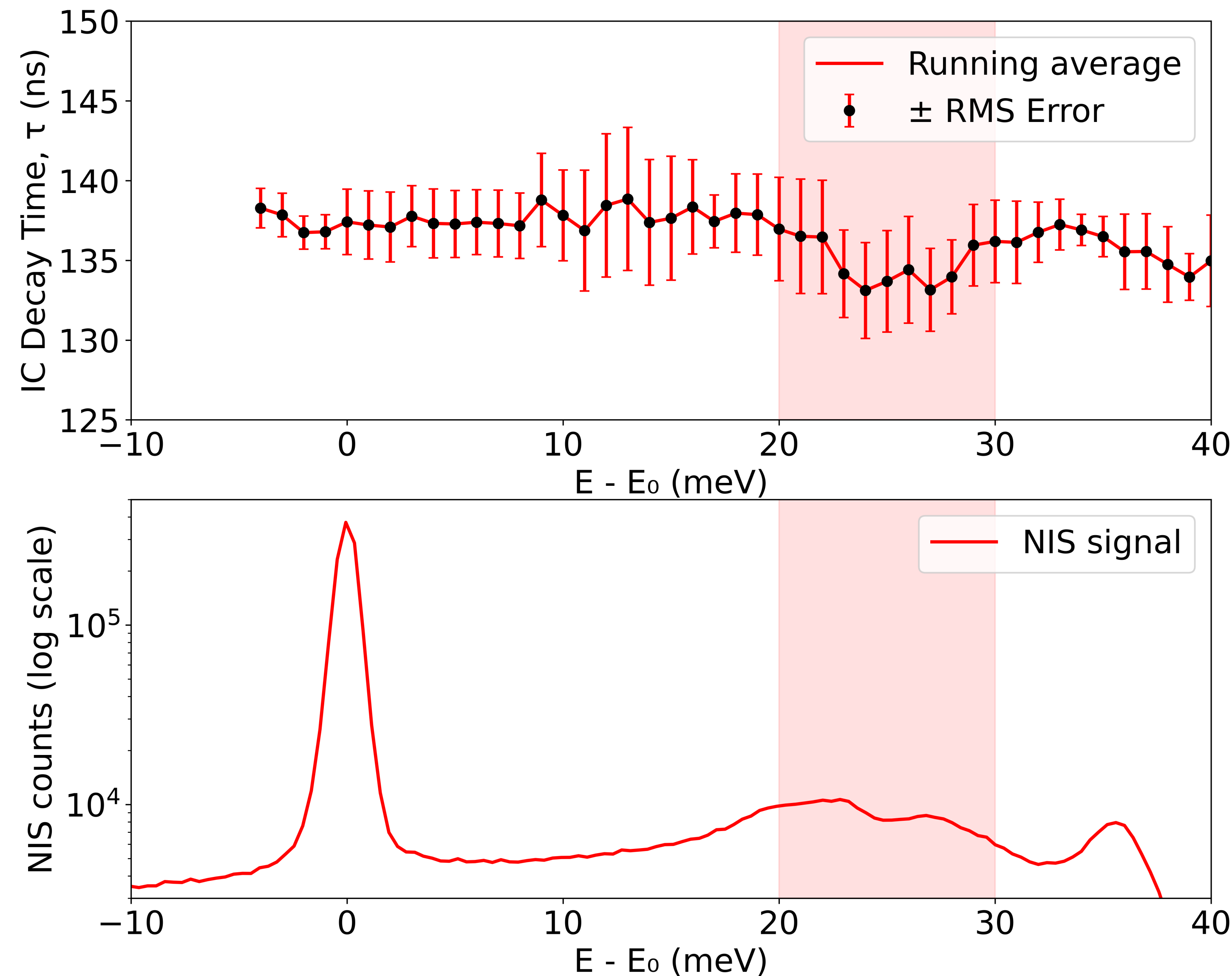
The IC decay time drops at around 25 meV energy detuning.



Consequence of the phonon coupling?

Internal conversion decay time for the dressed nuclear state

POSTER 37



Photoelectron Spectroscopy of the Mössbauer Transition in ^{57}Fe .

K Ravi¹, E Månsson¹, L Bocklage^{1,2}, S Veltén¹, M Seitz¹, S Rockenstein¹, D Schmitt³, M Kovacev^{3,4}, F Calegari^{1,2,5}, R Röhlberger^{1,6}, and A Trabatt^{1,3,4}

¹ Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany

² The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, 22607 Hamburg, Germany

³ Leibniz University Hannover, Institute of Quantum Optics, Welfengarten 1, 30167 Hannover, Germany

⁴ Cluster of Excellence PhoenixD (Photonics, Optics, and Engineering-Innovation Across Disciplines), 30167 Hannover, Germany

⁵ Department of Physics, Universität Hamburg, 22761 Hamburg, Germany

⁶ Institute of Optics and Quantum Electronics, Friedrich-Schiller-Universität Jena, 07743 Jena, Germany



Motivation

- Nuclear states can exchange energy with surrounding electrons, enabling key decay processes [1].
- Internal conversion (IC) and Auger-Meitner electrons constitute the primary non-radiative nuclear decay channels [2-3].
- Due to their limited escape depth, IC electrons are particularly well-suited as surface-sensitive probes of nuclear-phonon interactions [2,4].
- The Mössbauer effect enables precise investigation of these interactions [1-3].
- Moreover, electron emission detection provides a powerful alternative to fluorescence-based measurements of the phonon density of states, allowing investigation of electron-nuclear energy exchange at surfaces, thin films, and interfaces [4].
- Understanding nucleus-phonon coupling and controlling energy exchange between electrons and nuclei could open new pathways for quantum technologies.

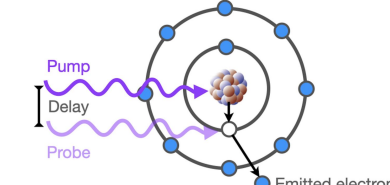


Figure 1: A prototype of time-resolved pump-probe laser spectroscopy of electron dynamics in nuclear transitions [5].

Experimental Setup

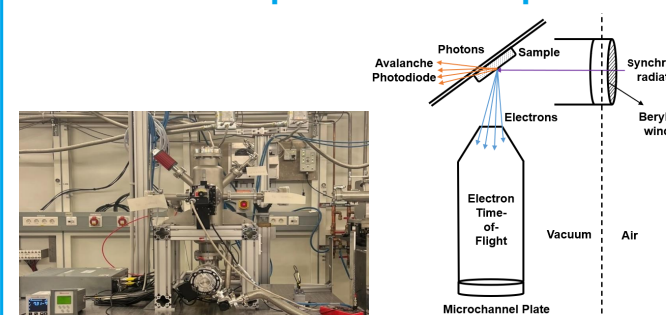


Figure 2: Sketch of the experimental setup

- Experiments performed at PETRA III (P01 High Resolution Dynamics Beamline), DESY
- Incident photon energy: 14.4 keV (resonant with ^{57}Fe)
- Repetition rate: 5.2 MHz
- ^{57}Fe nuclear lifetime: 141 ns
- Detection Techniques: Electron Time-of-Flight Spectrometer (ETOF)

Discussion

- Tuning the Synchrotron Mössbauer Source photon energy yielded high-resolution absorption spectra of ^{57}Fe foil (see figure 3) and thin films, resolving the expected hyperfine sextet structure.
- Detuning the photon energy off-resonance via the high-resolution monochromator enabled the extraction of a phonon spectrum by detecting the photoelectrons (see figure 4a).
- Plotting the photon energy detuning of the high-resolution monochromator against the decay times of IC electrons revealed a persistent dip in the curve at ~ 25 meV (see figure 4b), suggesting the presence of an additional pathway for IC decay.

Outlook

- Building up the new stopping potential spectrometer to perform ER-CENIS (energy resolved-conversion electron nuclear inelastic scattering) spectroscopy of ^{57}Fe .
- Possible time-resolved ER-CENIS spectroscopy of nuclear isotopes like ^{57}Fe , ^{45}Sc , ^{229}Th at European XFEL and other facilities within a pump-probe framework.

Conversion Electron Mössbauer Spectrum

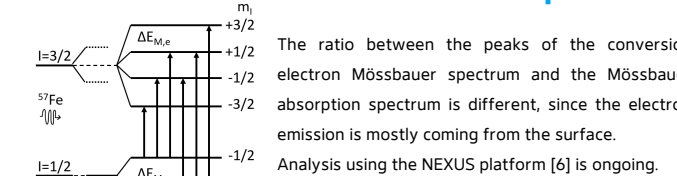


Figure 3: Conversion Electron Mössbauer Spectrum vs. Mössbauer Absorption Spectrum, ($E_0 = 14.4$ KeV)

Inelastic spectrum - Preliminary Results

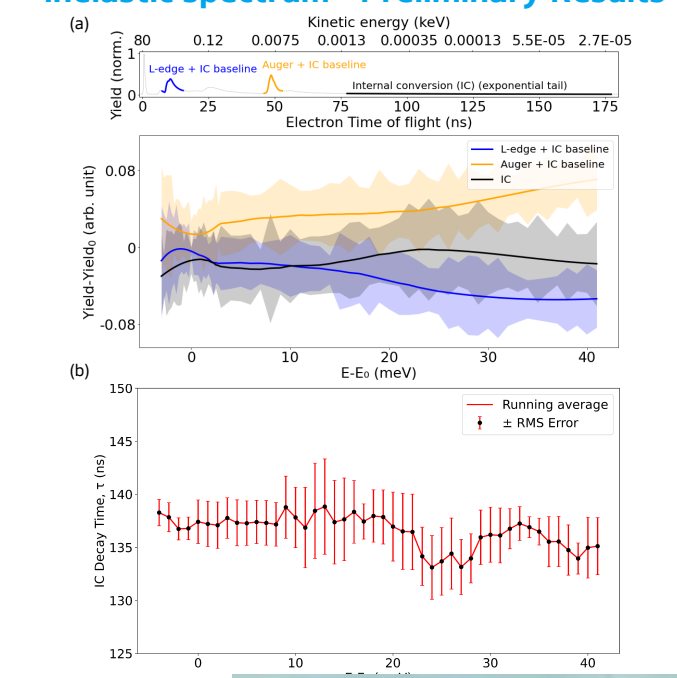


Figure 4: (a) Photoelectron-variation of IC decay time, (E_0 The half-life of IC electrons is value and is in good agree

- [1] R.L. Ingalls, "Application to Spectroscopy", Springer, Boston, MA, 1964.
- [2] W. Sturhahn, et al., "Electron emission from ^{57}Fe ", Phys. Rev. Lett. 73, 1771 (1994).
- [3] Yuri Shvyd'ko, R. Röhlberger, nuclear clock isomer ^{45}Sc , Nature 531, 201 (2016).
- [4] Taizo Kawauchi, Katsuyuki Fukui, inelastic nuclear resonant scattering, Phys. Rev. Lett. 115, 052501 (2015).
- [5] Seitz, M., Thirolf, P. G., Calegari, F., photo-induced electron dynamics in ^{57}Fe , Phys. Rev. Lett. 115, 052501 (2015).
- [6] Nuclear Elastic X-ray scattering, Phys. Rev. Lett. 115, 052501 (2015).
- [7] I. Ahmad, K. E. Rehm, E. P. Van der Sluis, Phys. Rev. Lett. 115, 052501 (2015).

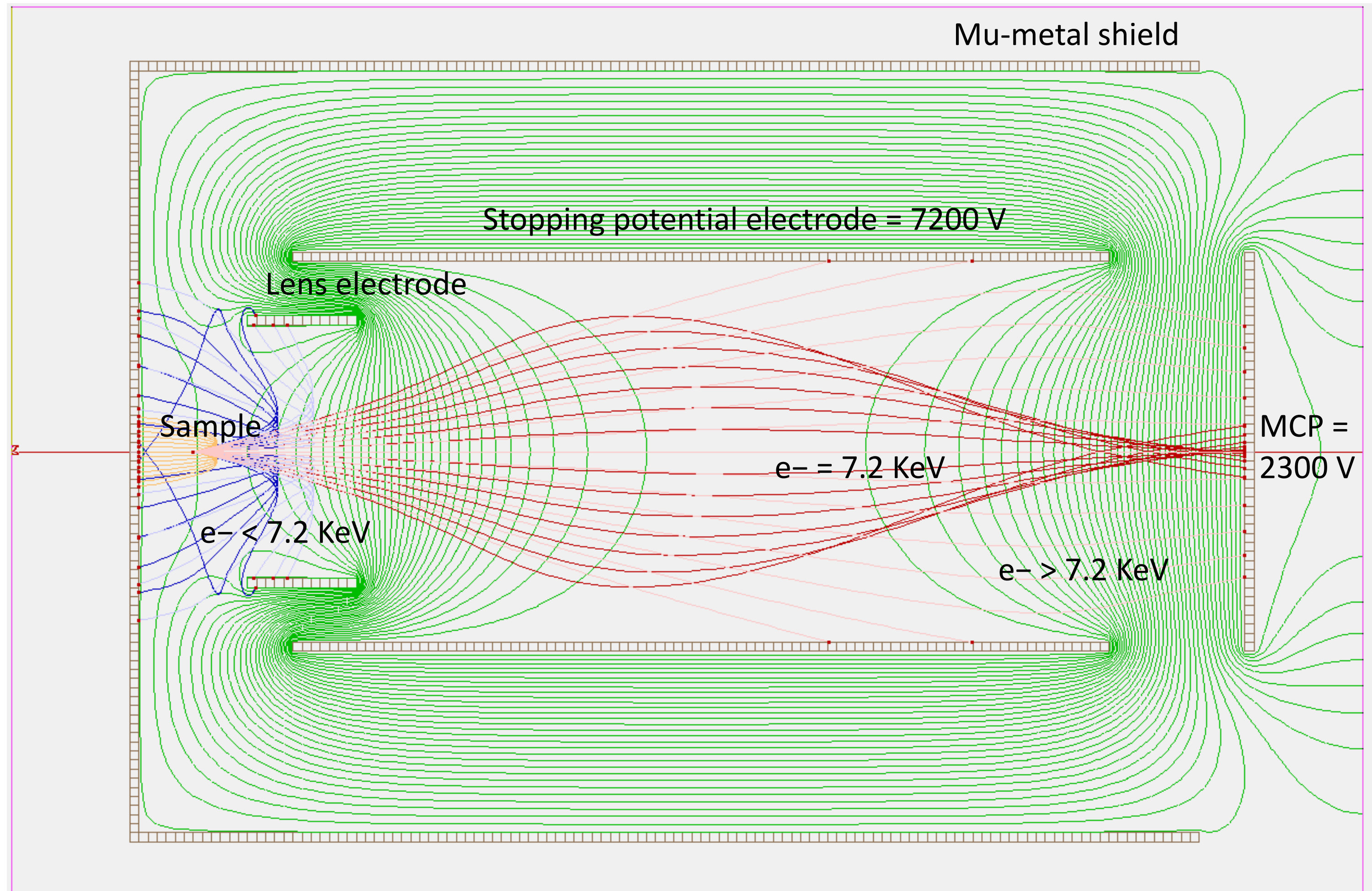


Keerthana Ravi

Development of a retarding-potential spectrometer

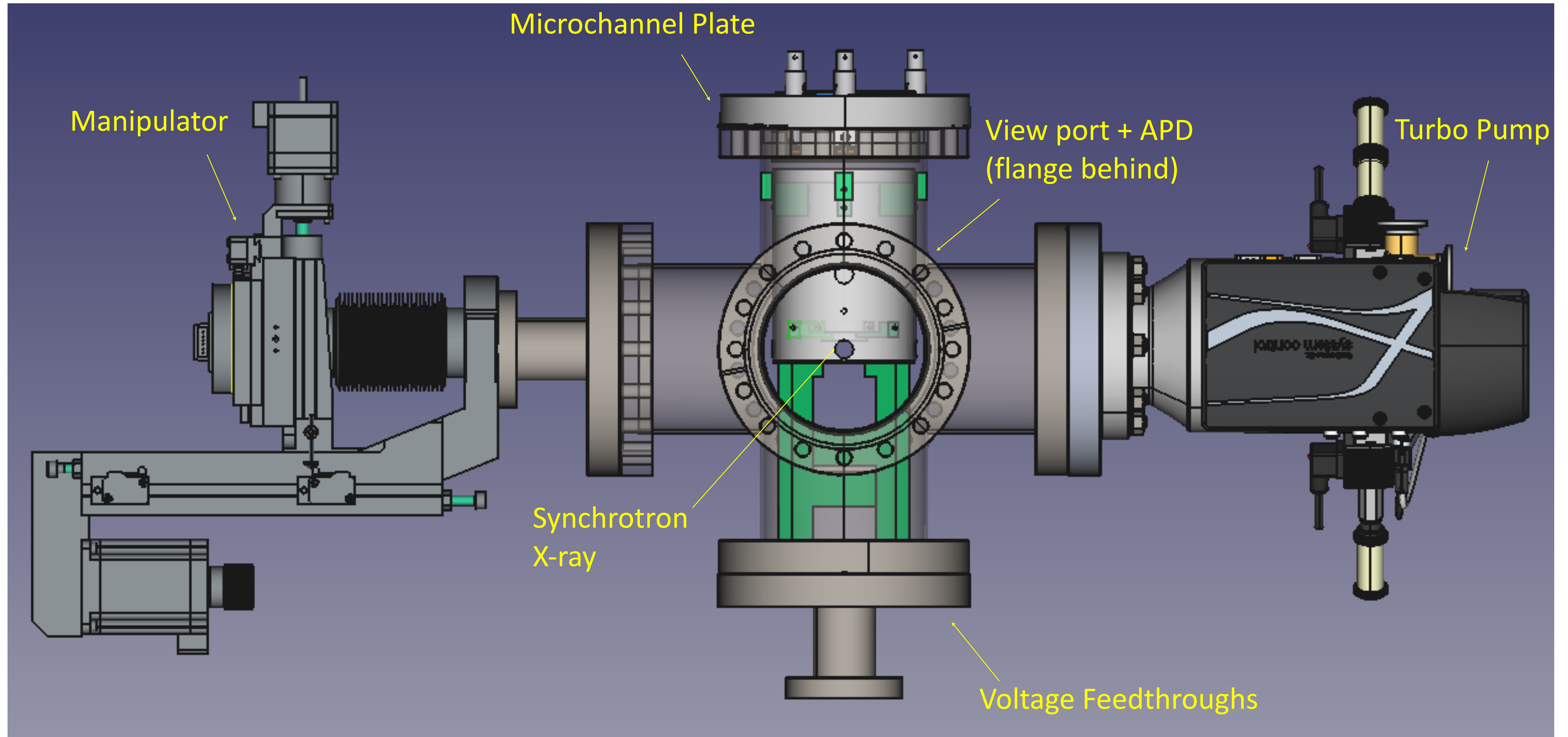
Energy-resolved conversion electron nuclear inelastic scattering

SIMION Simulation

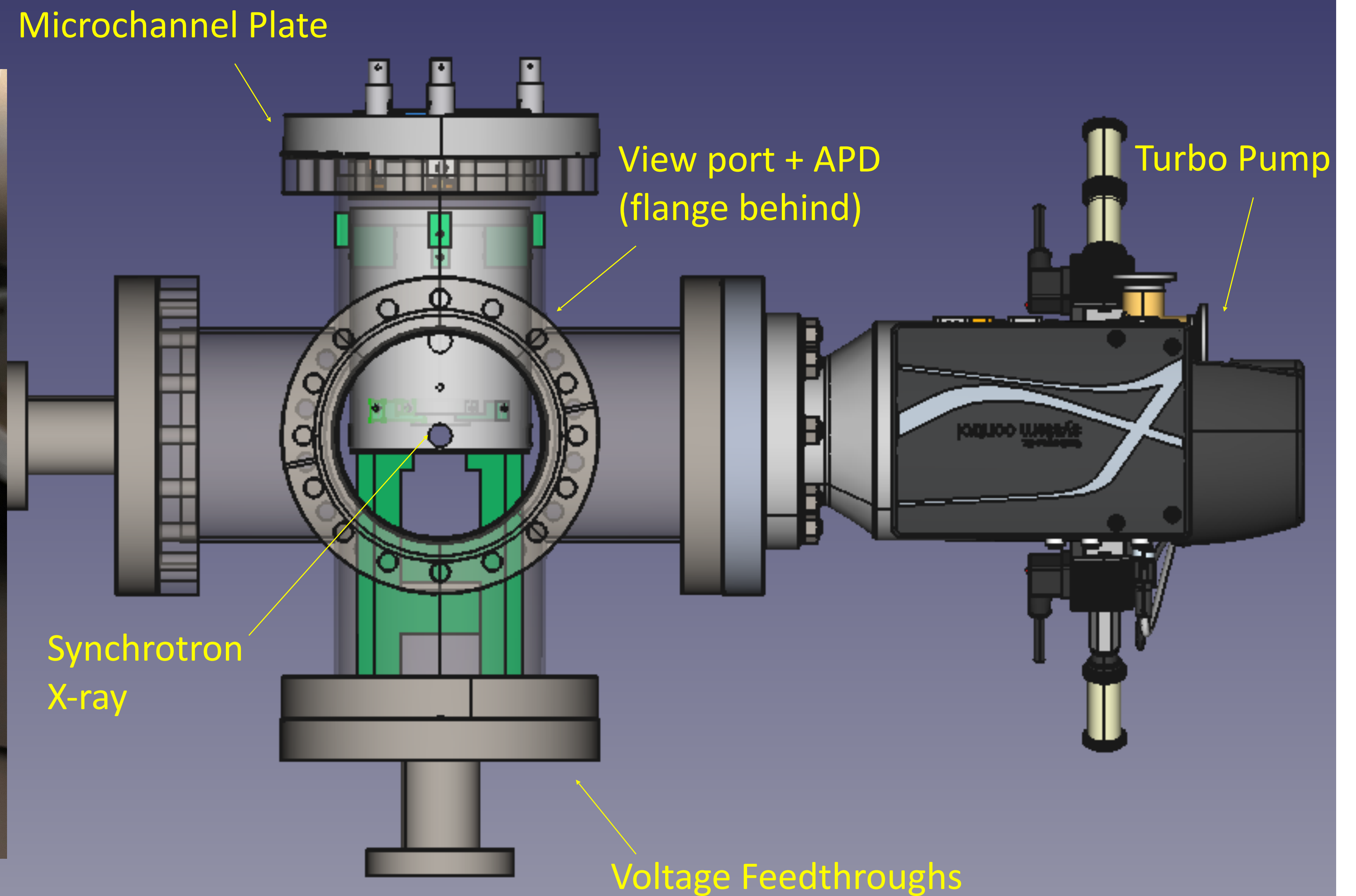
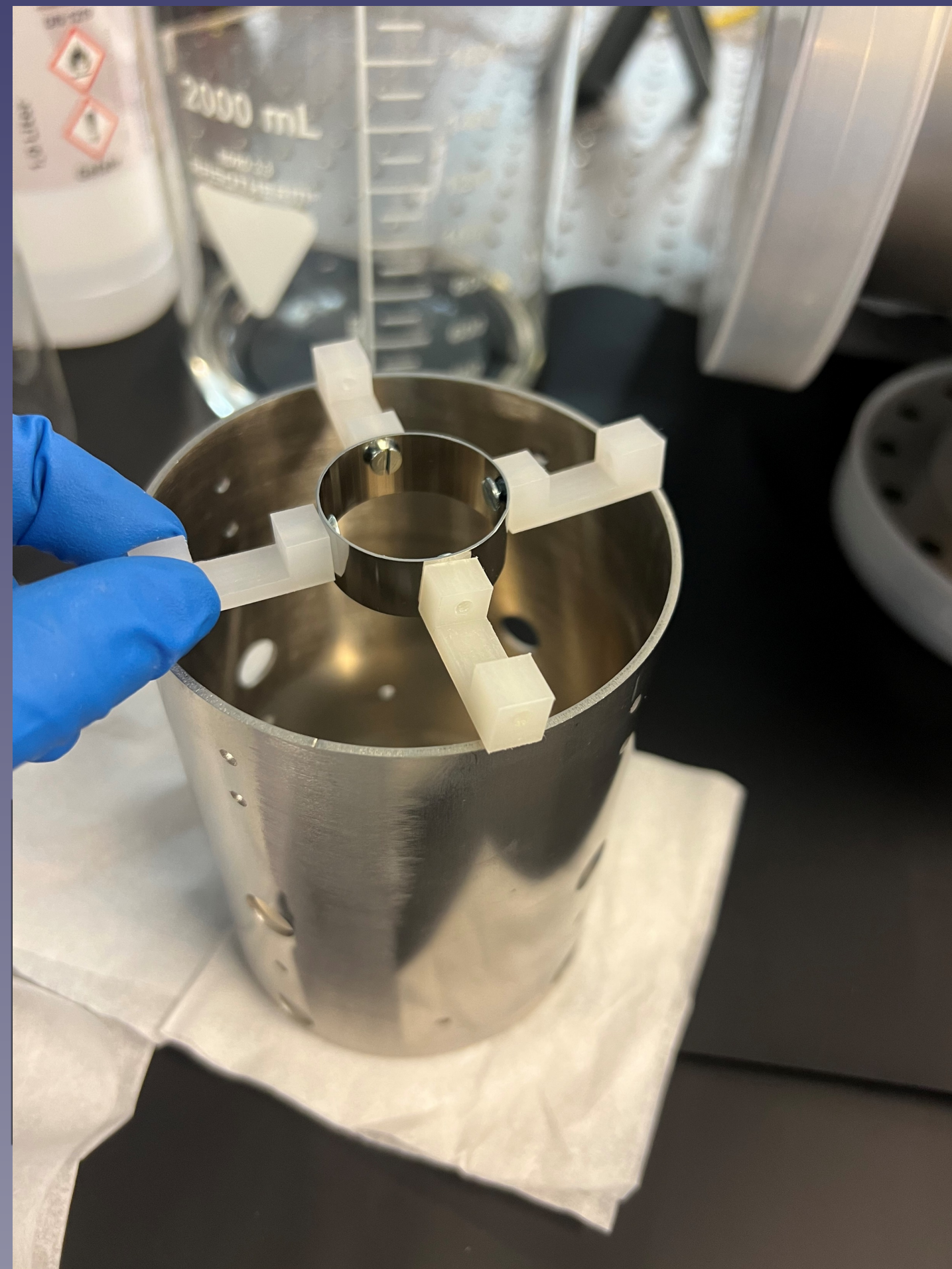


- Compact electron spectrometer equipped with a high-voltage stopping potential
- Wide electron acceptance angle (30° vs. 0.48° in the previous spectrometer)
- 3 orders of magnitude larger statistics than in the previous experiment.

Development of a retarding-potential spectrometer

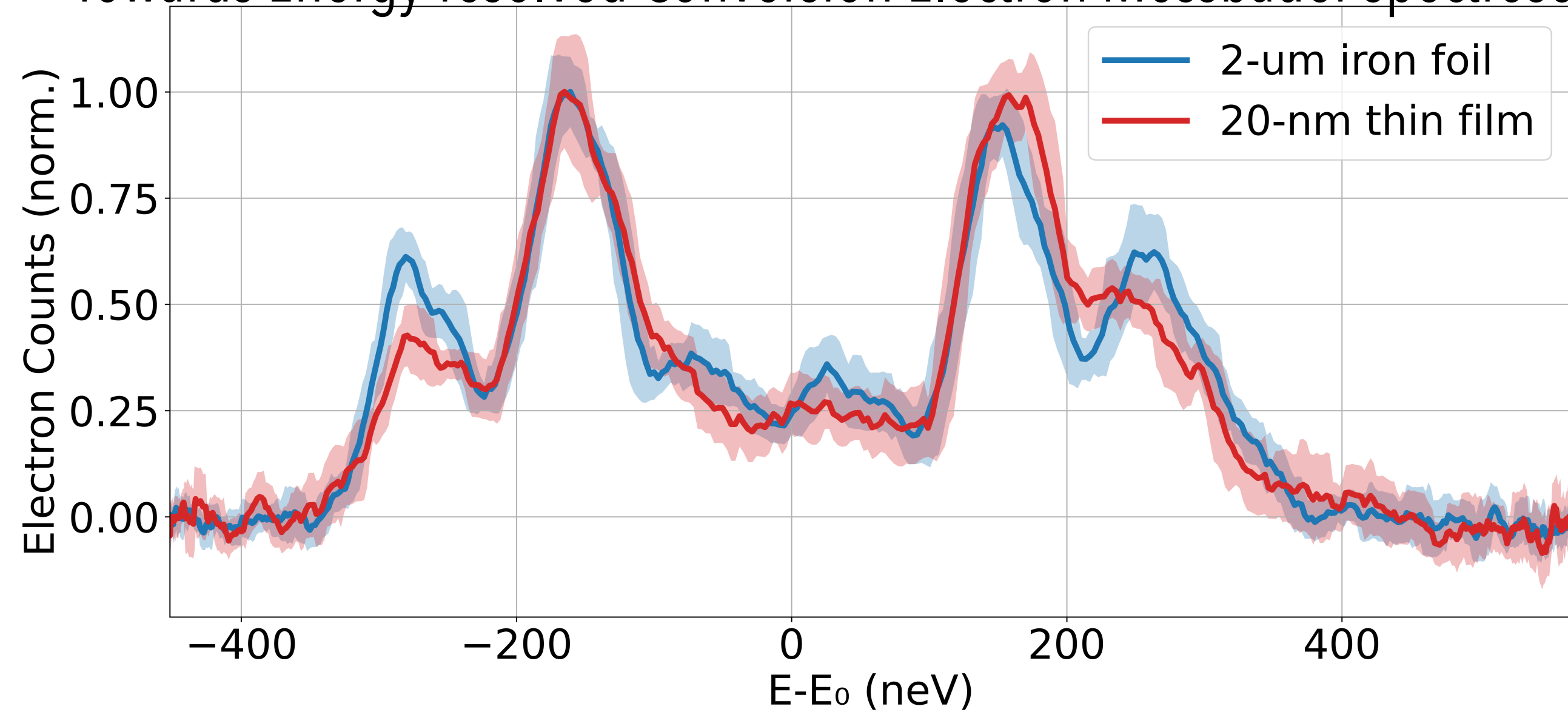


Development of a retarding-potential spectrometer



Energy-resolved conversion electron spectroscopy

Towards Energy-resolved Conversion Electron Mössbauer spectroscopy



Towards Energy-resolved Conversion Electron Nuclear Inelastic scattering

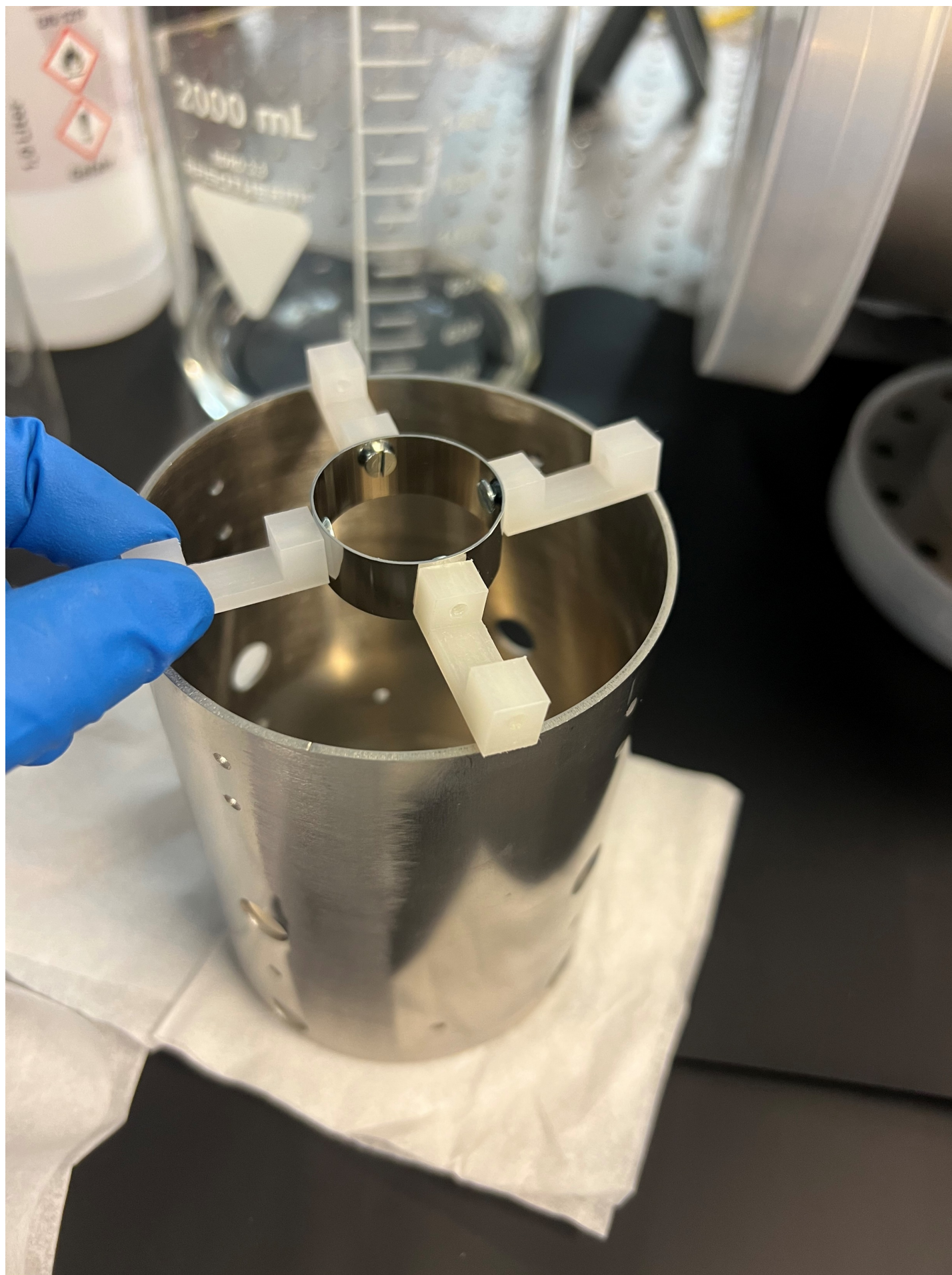
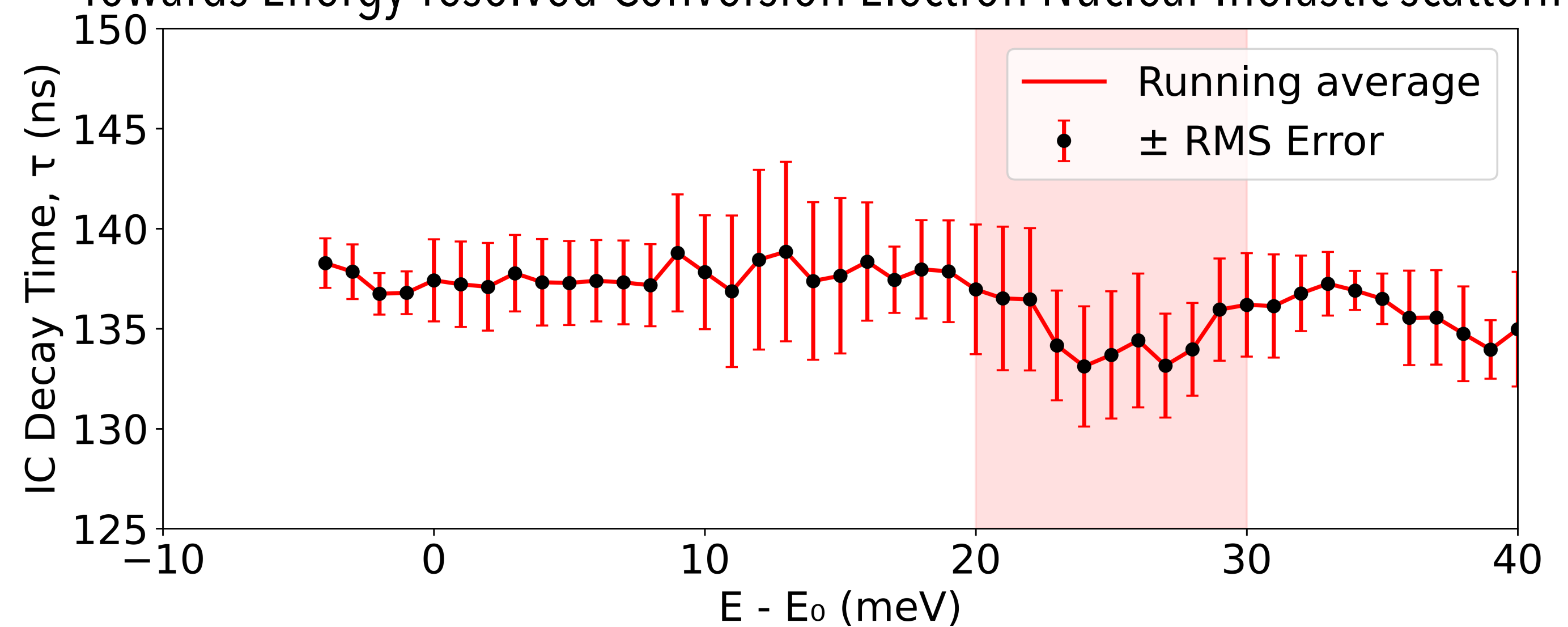
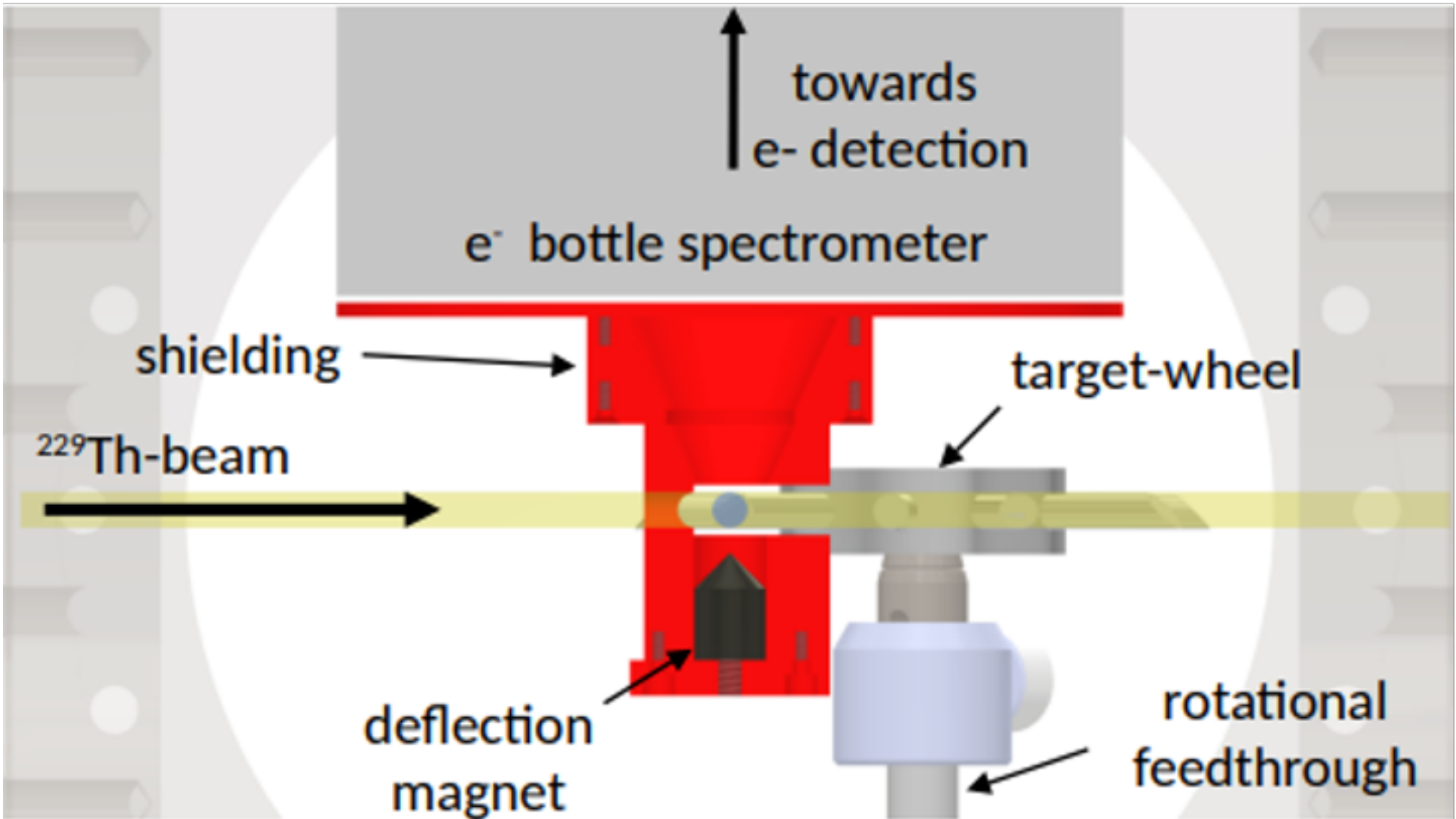
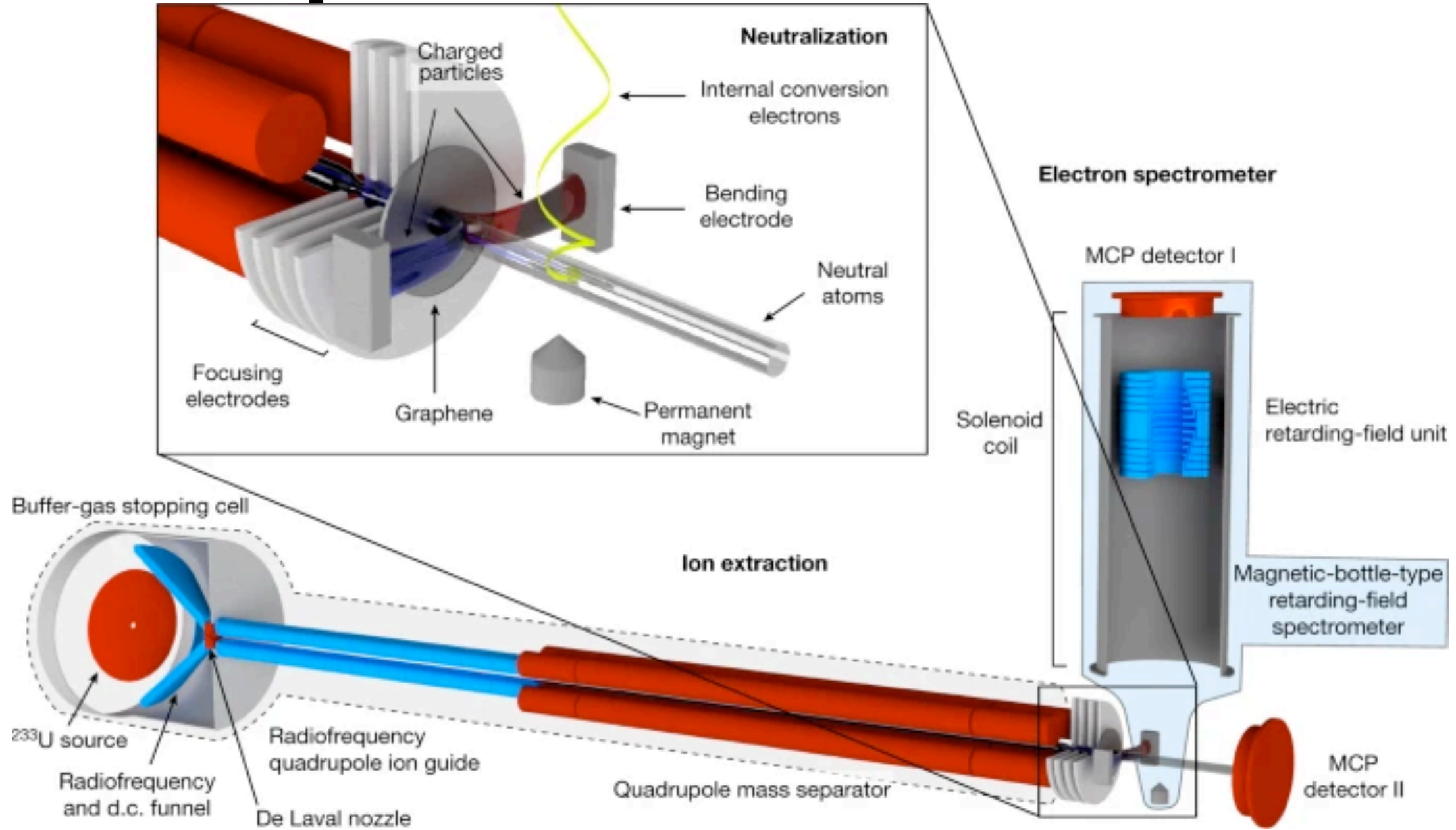


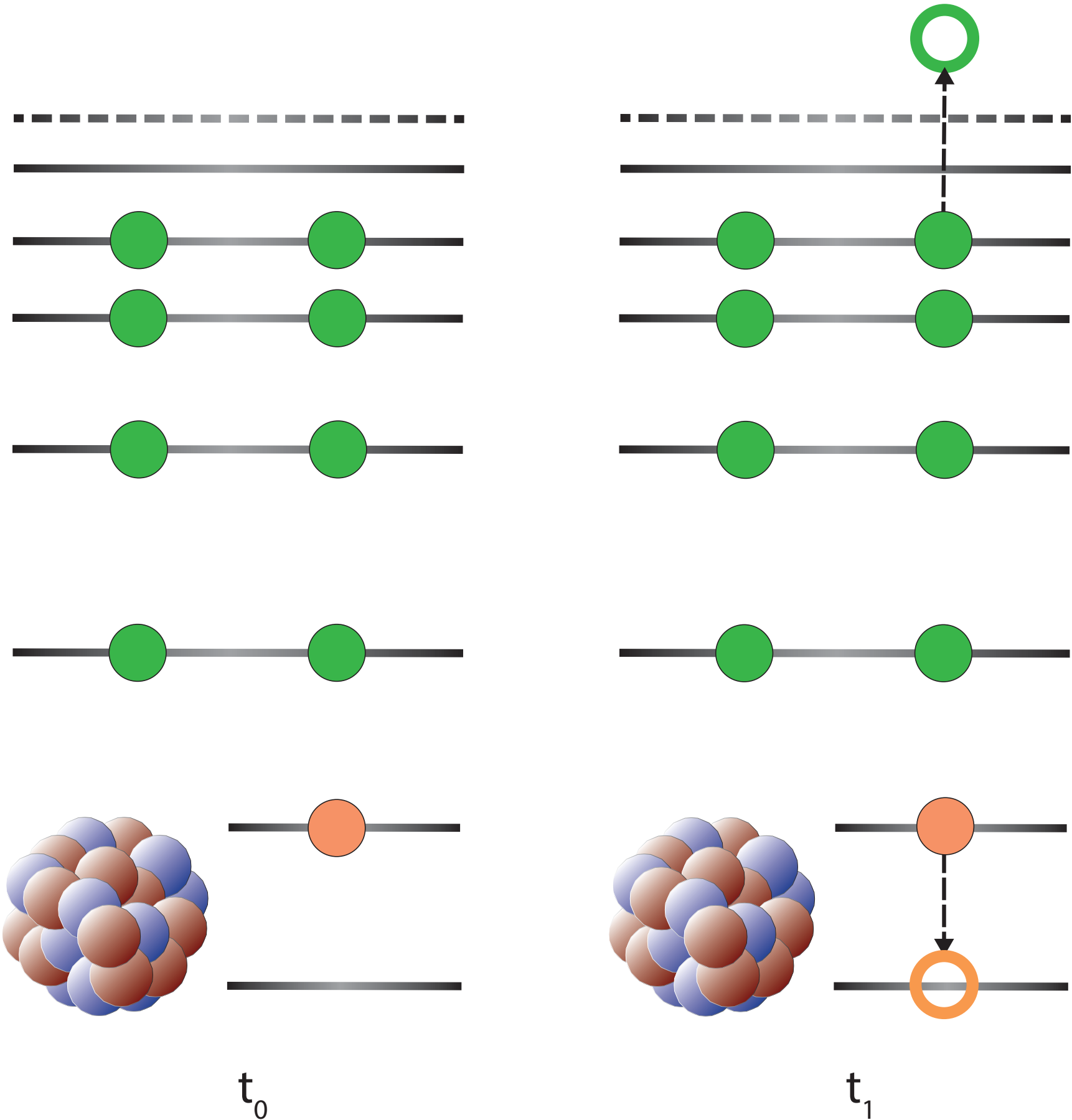
Photo-perturbation of IC in ^{229m}Th



Marc Seitz

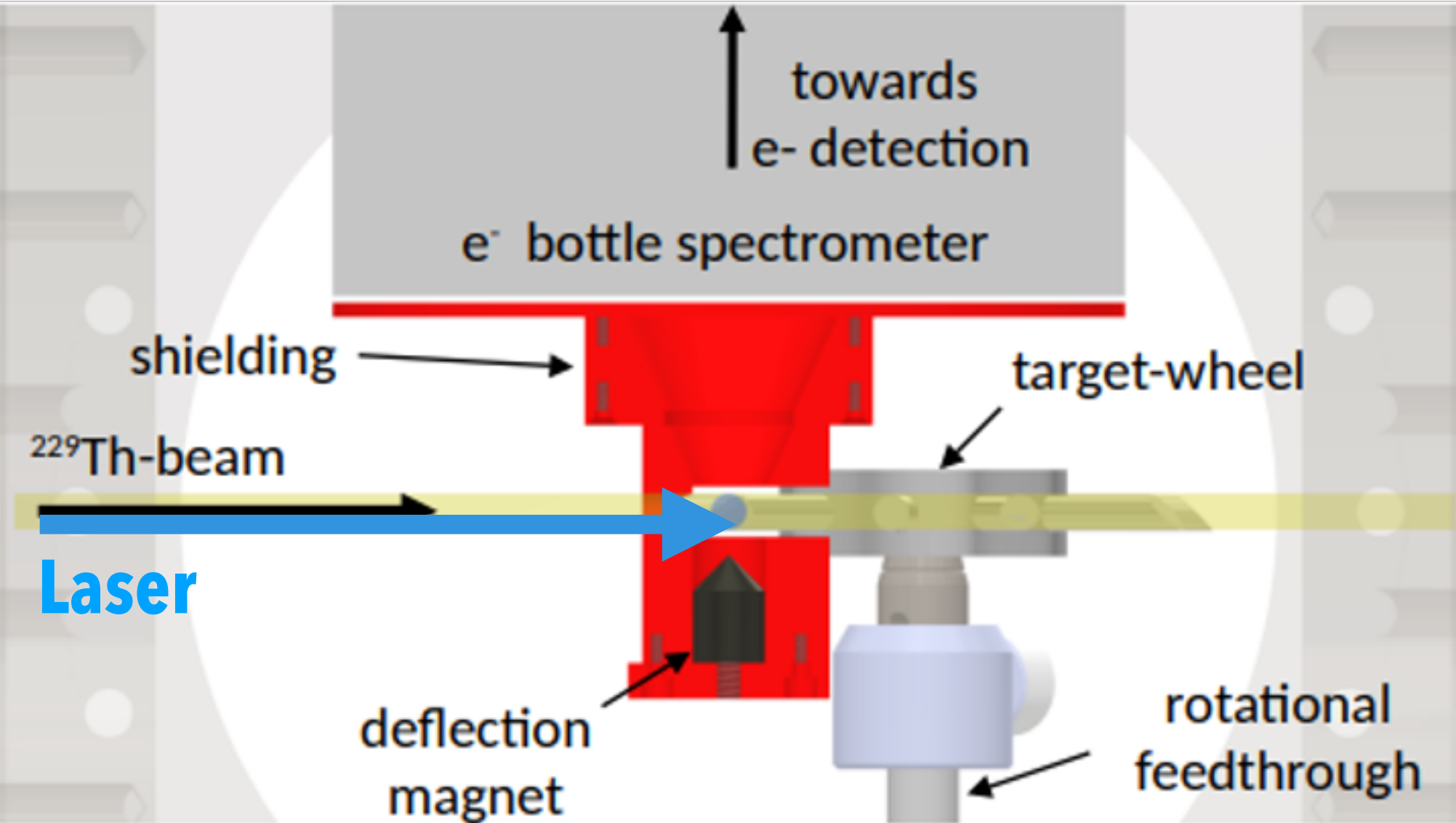
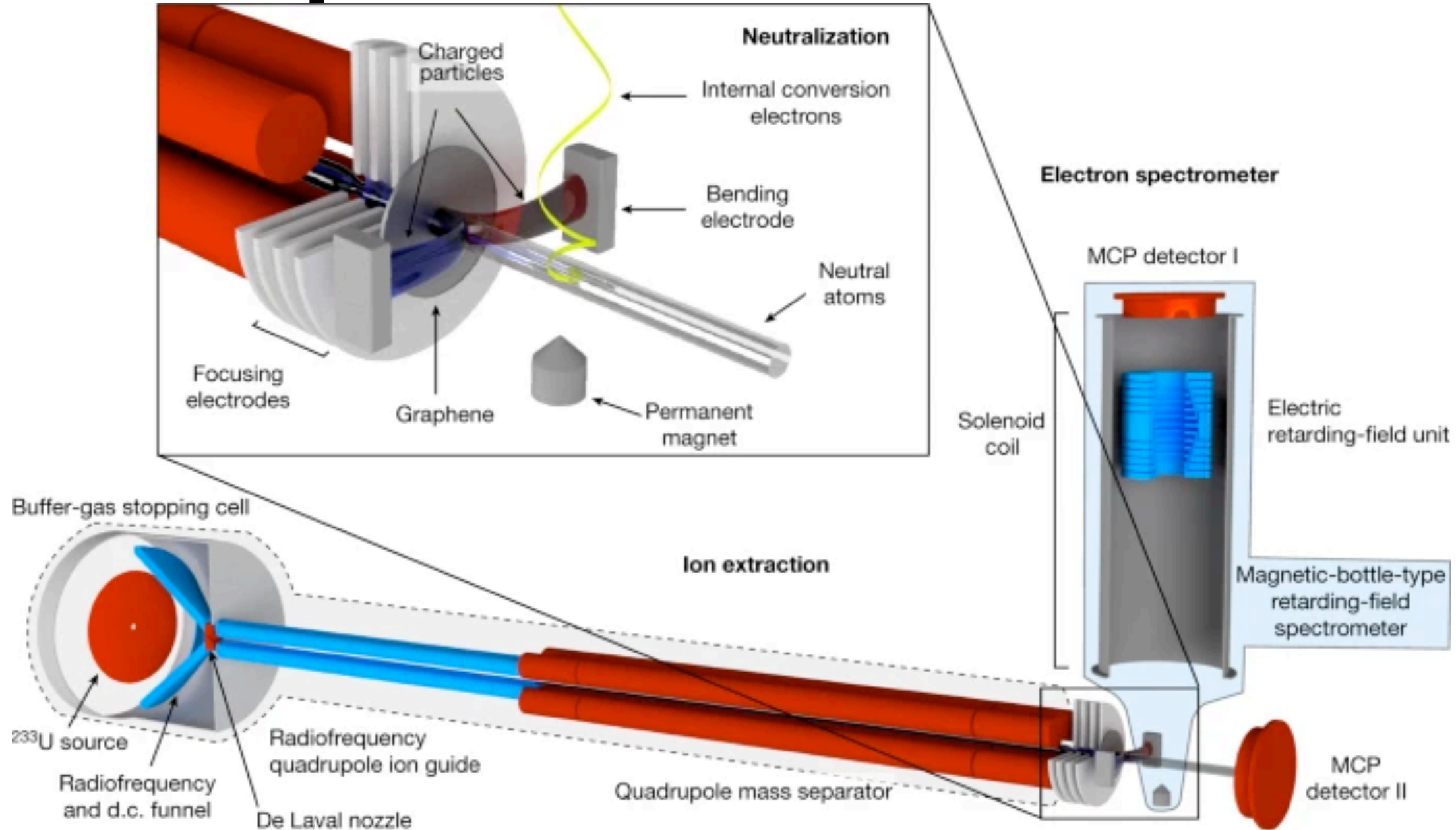


Daniel Moritz,
Peter G. Thirolf



B. Seiferle, et al., Nature 573, 243 (2019)

Photo-perturbation of IC in ^{229m}Th



Marc Seitz



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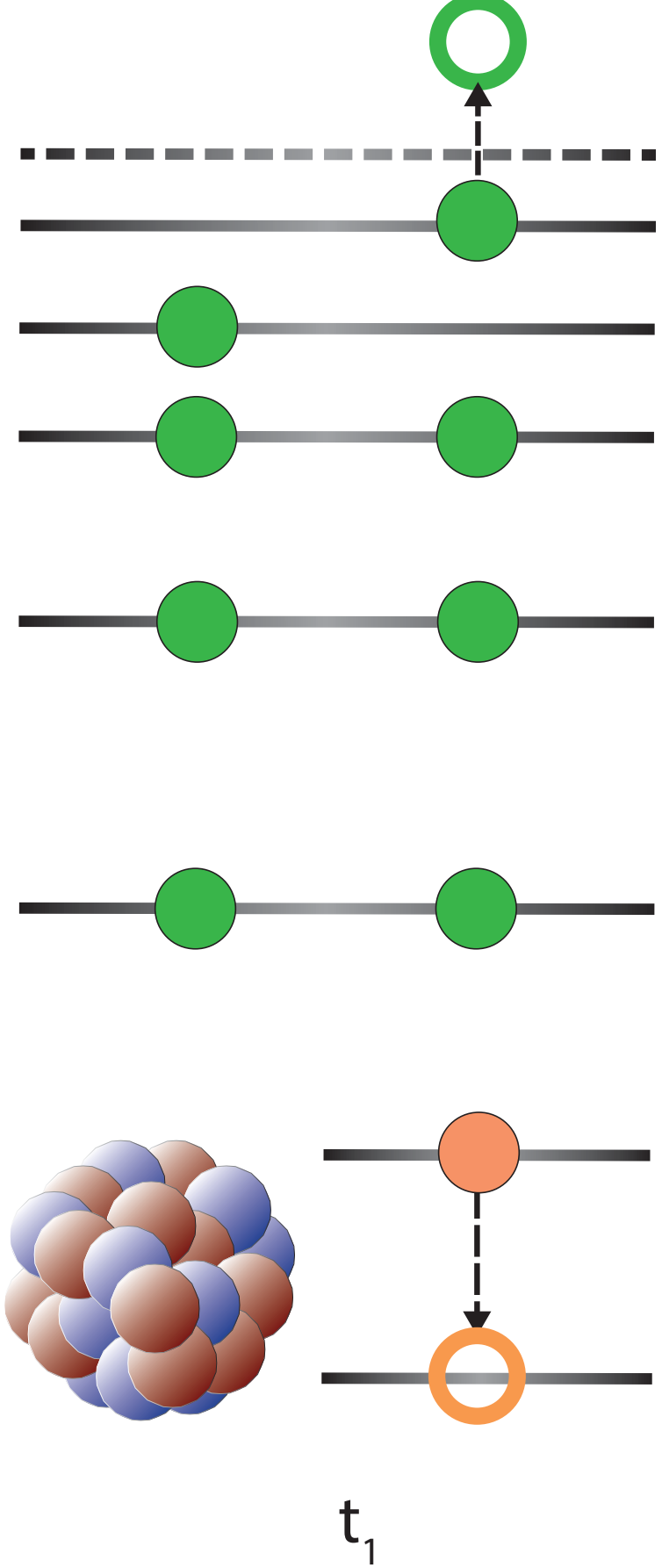
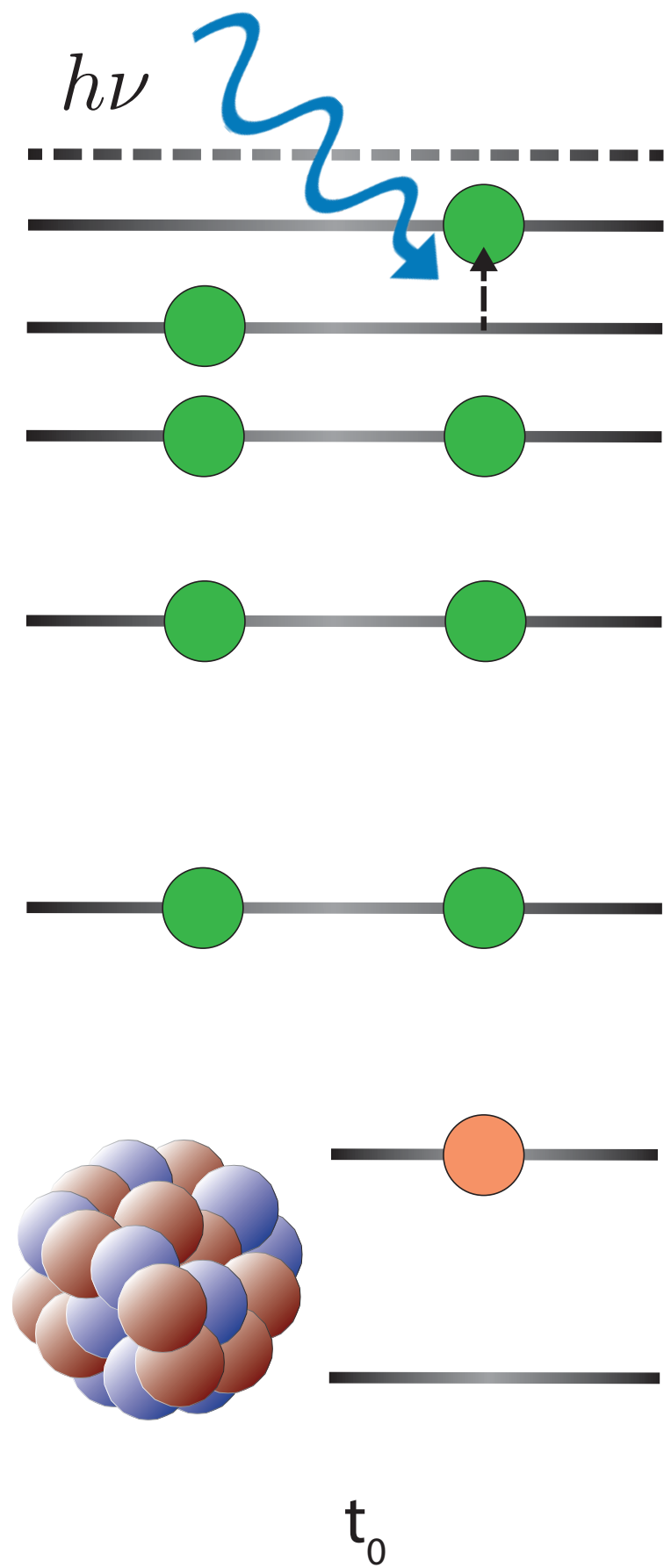
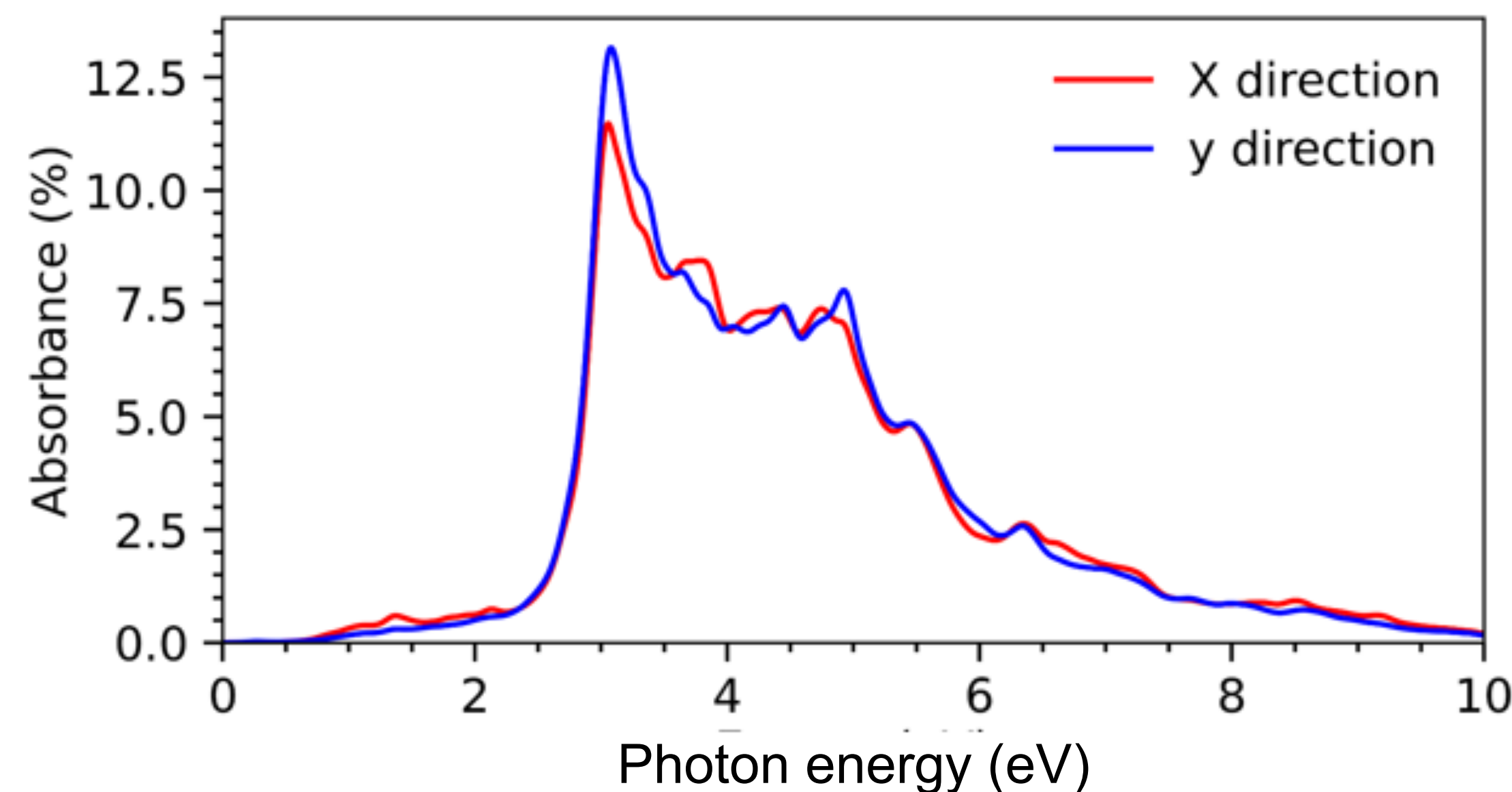


Photo-perturbation of IC in ^{229m}Th

Absorption spectrum ^{229}Th @ hBN



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Hannes Huebener



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Adriana Pálffy

Unperturbed IC

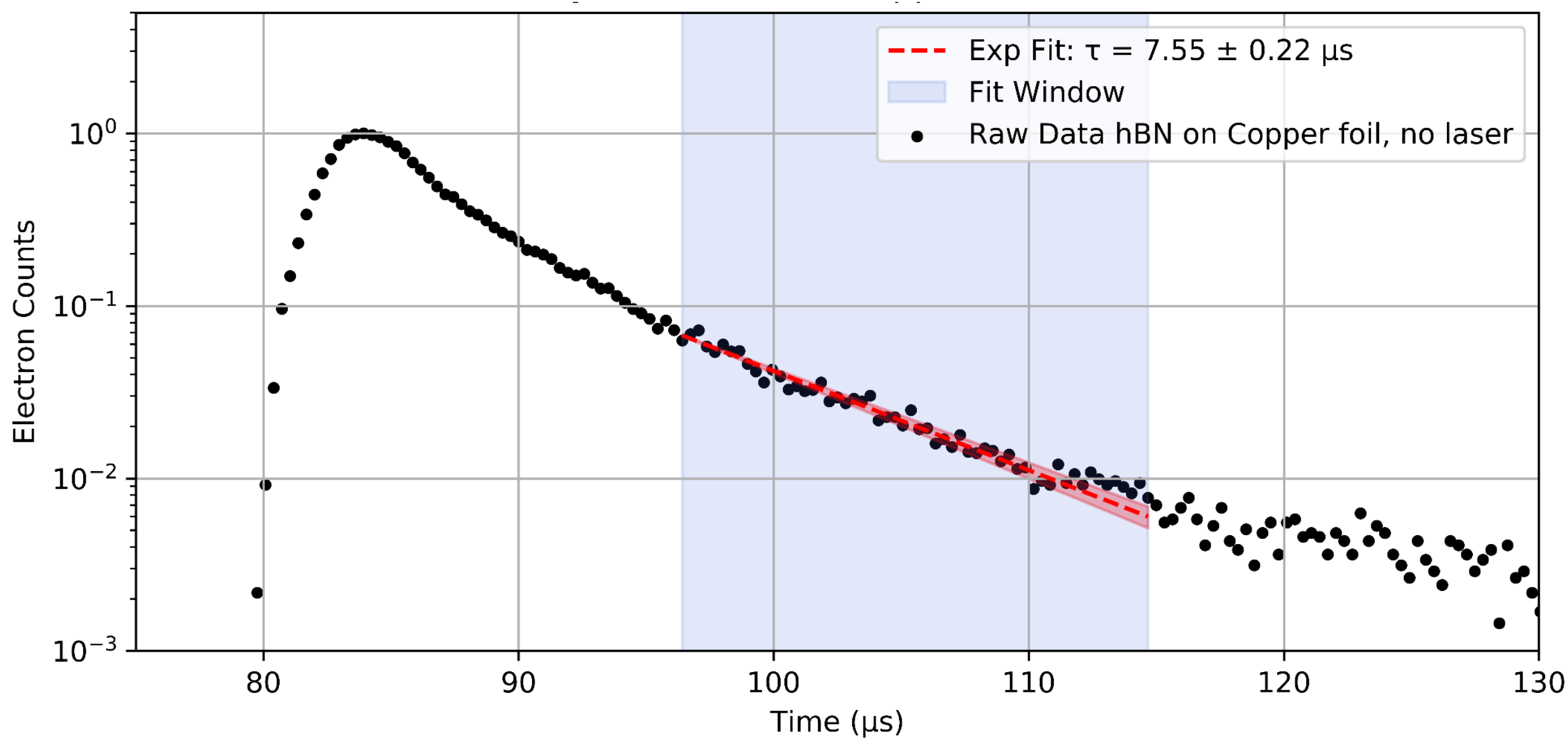
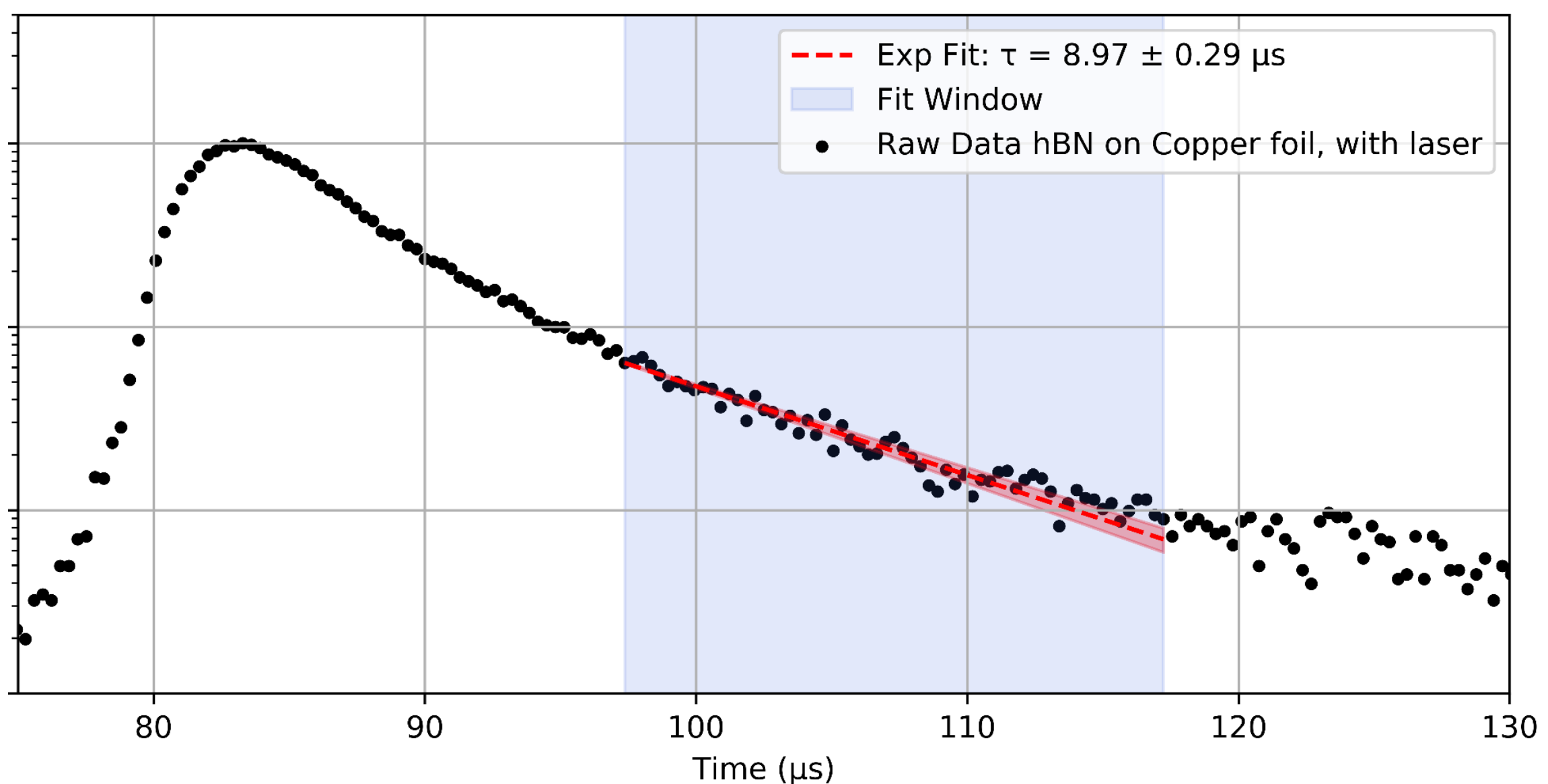


Photo-perturbed IC



Conclusions

Currently available light sources (table-top, synchrotrons, FELs) are opening the perspective to investigate a new family of electron dynamics with the tools of laser spectroscopy.

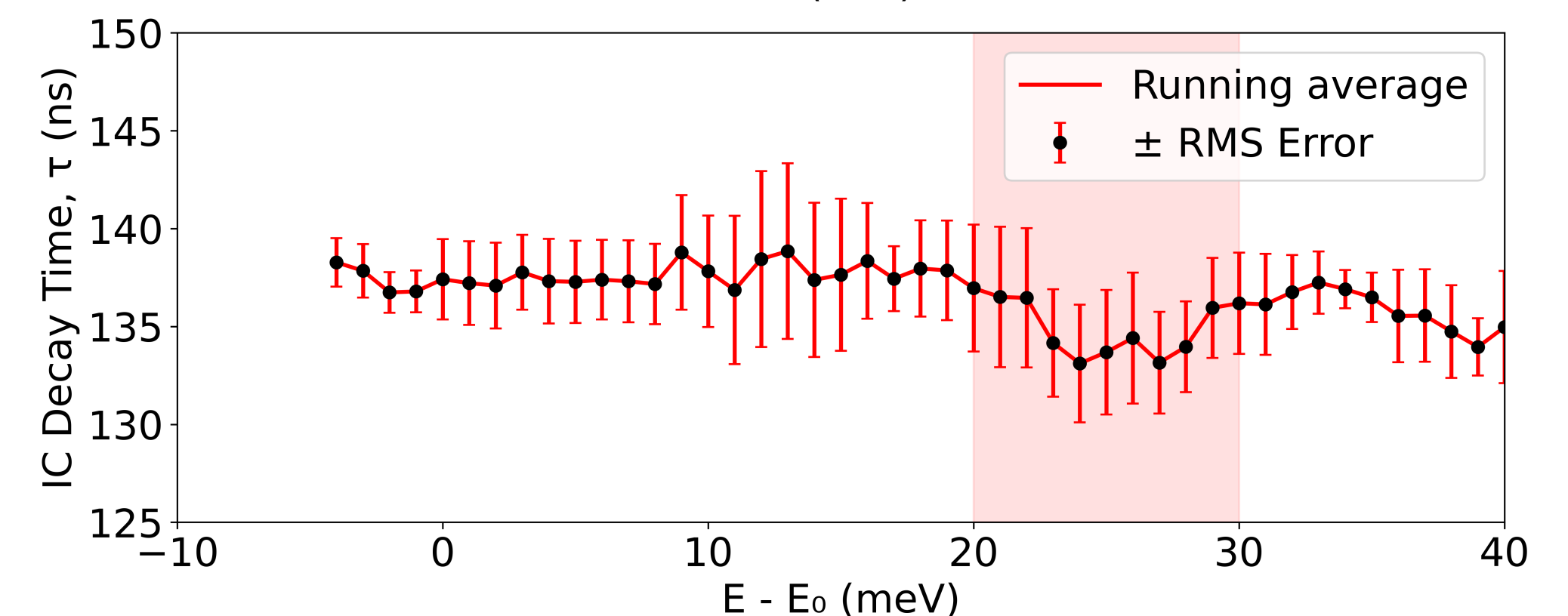
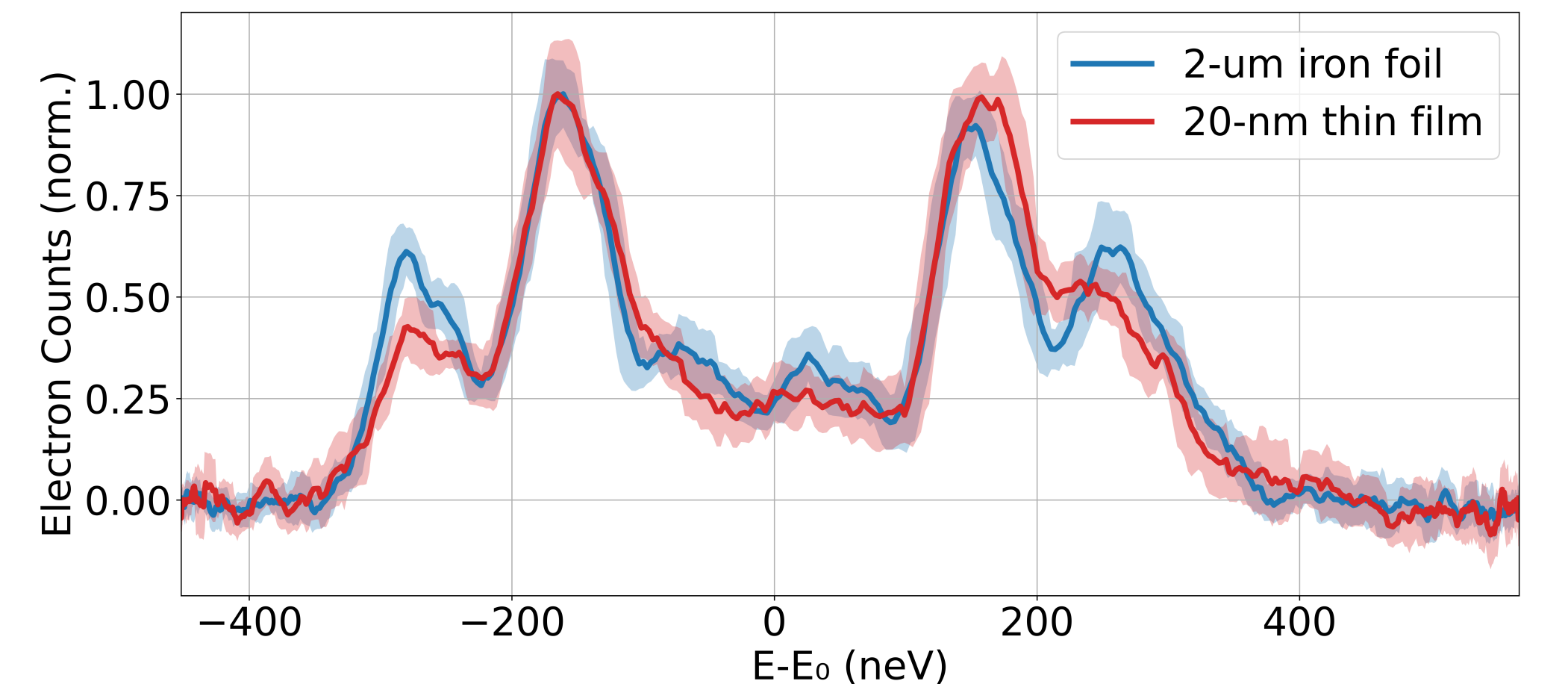
This research is leading to a new understanding of the coupling between nuclear and electronic states. This can enable interesting knobs of control over nuclear degrees of freedom through light.

This first experiment demonstrates that such an investigation is possible and already shows rich physics.

Next steps

Energy-resolved conversion electron nuclear inelastic scattering

Perturbation of the electronic structure with a laser: first examples of photo-control/pump-probe.



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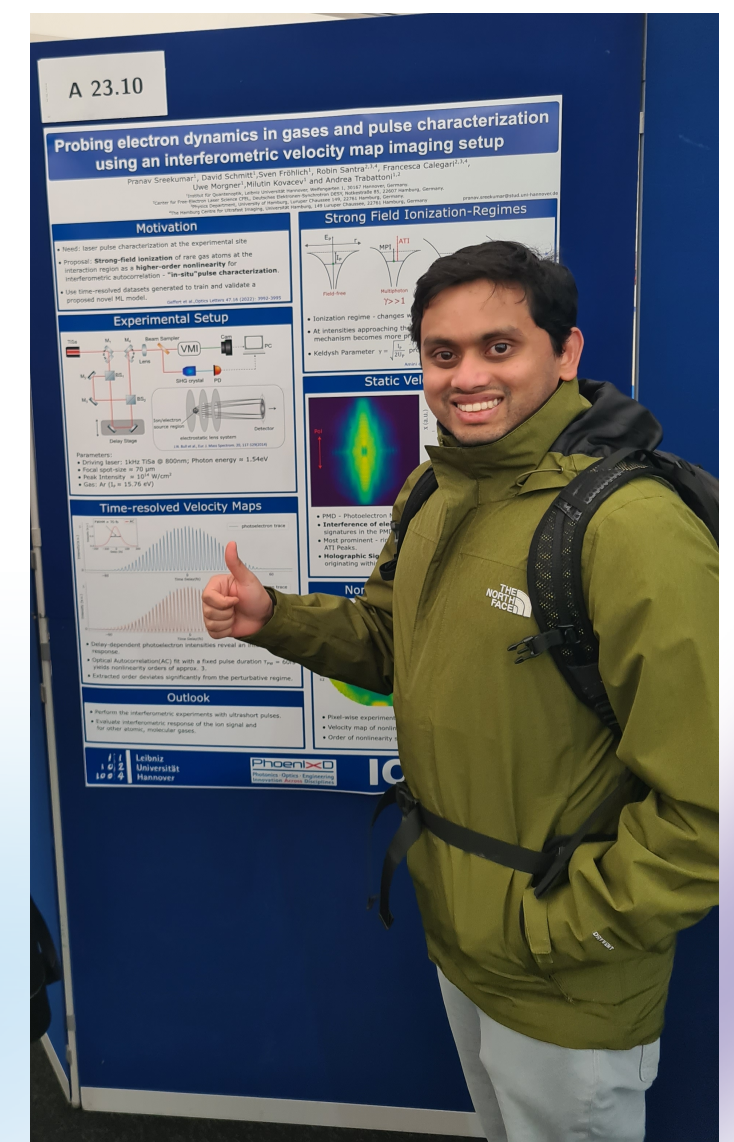
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<https://doi.org/10.1103/31qf-pfvj>

