# THE EUROPEAN PHYSICAL JOURNAL A



Regular Article - Experimental Physics

# Franz Käppeler and evolutions in nuclear physics

Ani Aprahamian<sup>1,2,a</sup>, Kevin Lee<sup>1</sup>, Shelly R. Lesher<sup>3</sup>

- <sup>1</sup> Department of Physics & Astronomy, University of Notre Dame, Notre Dame, IN 46556, USA
- <sup>2</sup> A. Alikhanyan National Science Laboratory of Armenia, 0036 Yerevan, Armenia
- <sup>3</sup> Department of Physics, University of Wisconsin La Crosse, La Crosse, WI 54601, USA

Received: 9 October 2023 / Accepted: 5 November 2023 / Published online: 20 November 2023

© The Author(s), under exclusive licence to Società Italiana di Fisica and Springer-Verlag GmbH Germany, part of Springer Nature 2023

Communicated by Nicolas Alamanos

**Abstract** This is a paper on the remarkable evolution in the career of Franz Käppeler that started with the measurements of neutron cross sections of various materials, followed by studies of key nuclei for the slow neutron capture process (s-process) in search of the origins of half of the heavy elements, and eventually to the measurements of the lifetimes of long-lived isomers identified in nuclei along the branching points of the s-process. This evolution of scientific interest took place over 4–5 decades and included the development of new tools and techniques that were implemented into the construction of new facilities for nuclear physics coupled with advances in both astronomical observations and theoretical approaches in modeling. Franz Käppeler led an exemplary life in his love of Rosel, good friends, great champagne, and those ever-elusive neutrons and their interactions with nuclei resulting in the various wonders of the universe.

## 1 Introduction

In physics, in art, in medicine, and in every human endeavor, we approach new concepts as revolutions in thought but, they are the results of steadfast and committed work that might be better defined as "evolutions in thought" that lead our lives of discovery and our quest for understanding the universe we live in. Franz Käppeler's career in nuclear physics started with the measurements of neutron capture cross-sections on various materials of interest to the nuclear energy community including neutron cross-sections for U, Am, Pu, and other actinides, their fission distributions, and the data for the design of fast reactors and their fuel cycle [1,2].

Nuclear cross-section measurements, data evaluation, and the generation of data for reactor calculations included the SNEAK 9B assembly. This SNEAK facility was to be sodium-free to avoid reactions of the neutrons with various materials of the reactor, such as the control rods. The measurements were focused on the potential for fast reactors. Research and design of fast reactors was ongoing among the nuclear "club" countries including the USA, Germany, France, Russia, and others in the 1970s and 1980s but neither the USA nor Germany continued towards implementation of fast nuclear reactors. These were meant to create less radioactive waste but research in this direction was abandoned in Germany and the Karlsruhe fast reactor research program [2] was shut down in the mid-1980s. On the brink of severe environmental impacts of climate change, the global effort has returned to more efficient and advanced reactor development and design according to the World Nuclear Association. It is somewhat ironic that some four decades later, we return to questions about fission distributions in neutronrich environments and neutron cross-sections with various coolants including molten salts. Fission has emerged as one of the frontier open questions in nuclear physics [3] with the observation of gravitational waves along with 70 electromagnetic transients that could observe the light from the merging of two neutron stars that happened 132 million years ago (GW170817). The nucleosynthesis process and the associated light observed in the kilonova event re-prioritized questions about the creation of specifically, the superheavies in nature and fission distributions in general for neutron-rich environments.

# 2 Origins of the heavy elements

The elemental abundances of the universe from the big bang to the present are shown in Fig. 1 in terms of the relative abundances of the elements. Over 99.9% of the visible mass of all the universe comes from nuclei that are made of protons and neutrons that themselves formed a few microseconds after the big bang as the primordial liquid known as quark-gluon



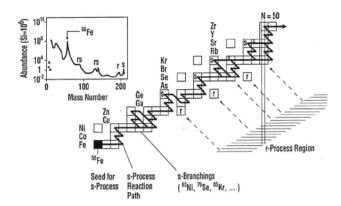
<sup>&</sup>lt;sup>a</sup> e-mail: aapraham@nd.edu (corresponding author)

**274** Page 2 of 5 Eur. Phys. J. A (2023) 59:274

plasma cooled and condensed. After the big bang, we find the very lightest elements of H, He, and Li. In the oldest stars, we observe the abundances of elements from Carbon to Iron produced in charged particle reactions where the solar abundances reflect the entire sum of the elements. Nuclear physics compels the explanation that all the elements higher than Fe have to be produced in neutron capture reactions. Figure 1c indicates the intertwined history of the astrophysical scenarios and the nuclear physics that operates in those conditions to yield the observed solar abundances.

Neutron capture reactions were classified into slow (s-) and rapid (r-) neutron capture reactions by the 1950s by E. M. Burbidge et al. (B<sup>2</sup>FH) [5]. Details were explicitly developed by many others [6–8]. The s- and r- processes are each responsible for the existence of approximately 50% of all the heavy elements beyond Fe with the s-process though to occur in the He-burning layers of the low-mass asymptotic giant branch (AGB) stars [9,10]. The separation of the s- and r-process mainly came from the isotopic patterns where the s-process closely follows the line of stability since the capture of neutrons in this process is slower than  $\beta$ -decay.

Figure 2 shows the s- and r-process paths. The inset of the figure shows abundance peaks beyond Fe that are due to both r- and s-process nucleosynthesis. In the 1990s there were significant improvements in the precision of nuclear data, in the developments of stellar models to include temperature and neutron density effects, as well as observations of pre-solar grains and spectroscopy of astronomical objects resulted in a clear separation of the s- and r-process contributions to the solar abundances [13]. This clear identification of s-process abundances resulted in robust r-process abundance patterns that were identified in several metal poor stars [14]. The clear r-process pattern resulting after the subtraction of the s-process abundances from solar abundances were then used to disentangle the astrophysics from the nuclear physics. This was done with measurements of masses for neutron-rich nuclei produced in fission [15,16] and useful in reverse engi-



**Fig. 2** Path of the s- and r-processes in the nucleosynthesis of the heavy elements [9]. The inset of the figure includes the abundance peaks for both s- and r-process

neering high precision nuclear mass measurements of fission produced neutron-rich nuclei to finally identify neutron star mergers as a potentially important site for the r-process [17].

#### 3 Franz Käppeler

Franz Käppeler came into the detailed studies of the s-process following studies of fission cross sections of U, Am, and other heavy nuclei. His expertise in neutron capture cross sections of isomers was crucial to setting limits on astrophysical models. His precision work with isotopic branchings in the s-process revealed inconsistencies that could then be addressed. An example was the study of the "rarest isotope <sup>180</sup>Ta" [18] where the synthesis of <sup>180</sup>Ta is strongly dependent on the temperature of the astrophysical environment and inconsistent with measurements then available in the laboratory. It became clear that the s-process abundances could not be completely explained over the entire period of galactic evolution [11,19]. Questions remained about the impacts of the long- and short-lived isomers. An example is shown in Fig. 3 where some of the branchings beyond Sr are shown

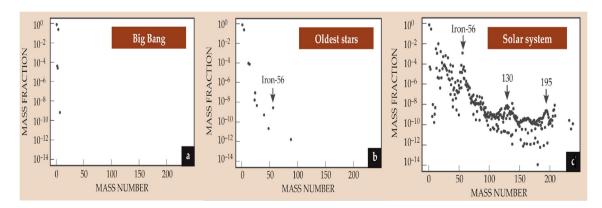


Fig. 1 Relative abundances of the elements [4]: a the relative mass fractions right after the big bang; b in the oldest stars; and c in the present solar system



Eur. Phys. J. A (2023) 59:274 Page 3 of 5 274

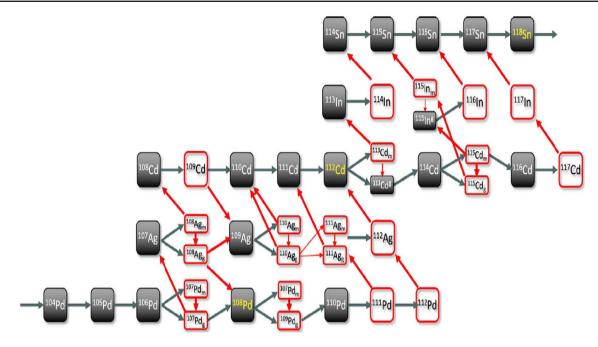


Fig. 3 Branchings from Pd to Sn along with the relevant isomeric states from Ref. [11]. These are the exact branchings where there are discrepancies and it is intriguing that the same elements correlate with r-process enhanced metal-poor stars [12]

[11]. Figure 4 shows the <sup>176</sup>Lu, <sup>176</sup>Hf branching pathways. The former case as shown in Fig. 3 is one of a complex r-process and s-process admixture. The lack of experimental data is absent in this region of nuclei. The latter case illustrated in Fig. 4 is very different since the neutron capture takes place around the isomeric state with no thermalization of the isomer and the ground state.

I met Franz Käppeler for the first time in 1993 at the International Conference on Capture Gamma-rays in Fribourg, Switzerland. At this time, he was already deeply engaged in nuclear astrophysics and a star for his work on <sup>56</sup>Fe. This paper takes a daring and audacious direction predicting what might have been the next direction of Franz Käp-

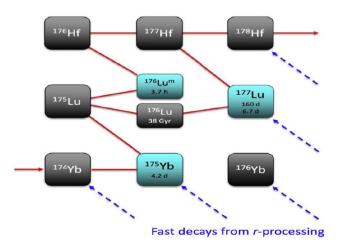


Fig. 4 The branching of <sup>176</sup>Lu and <sup>176</sup>Hf from Ref [23]

peler's research starting with nuclear reaction cross sections on materials to solving open challenges [20] in nuclear astrophysics with the neutron capture reaction rates above  $^{56}$ Fe. The high-resolution measurements of the neutron capture cross-sections of  $^{56,58}$ Fe and the higher masses allowed the sprocess calculations for abundances of elements A=56-75 to be consistent with the abundances of rare isotopes with  $A{\le}56$ . These results were significant steps in understanding the role of the s-process contribution to the solar abundances. In other words, it was these neutron capture cross sections that allowed the clear separation of s-process abundances from the entire observed solar abundances yielding a distinctive pattern for the r-process.

# 4 The s-process and Isomers

The existence of isomers and the branching paths of nuclei along the s-process raise questions about the astrophysical scenarios that can excite the long-lived isomers and change the resulting abundances. What if we consider even shorter-lived elements? Isomer lifetimes span the entire range of lifetimes [21] from the very long like the isomer of  $^{180m}$ Ta with a  $10^{15}$  year half-life to those that are in the nanoseconds range. The isomers and the potential manipulation of the isomer half-lives so that we can excite and de-excite isomers at will have unlimited application potential. The concept of induced gamma-ray emissions was extensively explored [22] in terms of the potential development of a gamma-laser for several decades but it is only recent advances that allow the potential of controlled emission to be reconsidered. The inhibition



274 Page 4 of 5 Eur. Phys. J. A (2023) 59:274

of the decay out of the isomers is viewed as a tremendous energy storage device.

### 5 Can we engineer nuclear lifetimes?

Can we manipulate isomer half-lives? Studies of isomer half-lives give us insights into nuclear wavefunctions. The ability to manipulate the nuclear wavefunction [24–26] is yet unproven and remains as an open question in nuclear physics. The potential applications of the ability to manipulate nuclear wavefunctions in order to affect half-lives of nuclear isomers are very broad. The potential of controlling the release of the energy stored in isomers can of course lead to the much sought after gamma-ray laser and to any more societal applications that can only be dreamed of in the present.

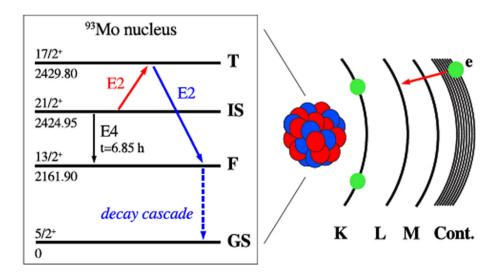
There is some evidence of isomer depletion by electron capture [27]. In electron capture, the nucleus releases an atomic electron instead of emitting a  $\gamma$ -ray. This "internal conversion" has been known for a long time and frequently used to study the nature of excited states in nuclear structure. In the inverse process, Nuclear Excitation by Electron Capture (NEEC), a free electron is captured into a bound atomic orbital and transfers the excess energy to the nucleus. This NEEC process was predicted in the mid-1970's by Goldanski and Namiot [28] and has yet to be observed [29]. The implication to astrophysics is that stellar environments potentially excite the ground states as well as the isomers which can then depopulate via shorter lived nuclear states by electromagnetic transitions, thereby contributing to the observed elemental abundances by changing the isotopic abundances after they have been produced in the s-process.

A measurement using the NEEC technique [25,27] has shown results for the <sup>93</sup>Mo nucleus. Here the nucleus is in

the isomeric state at 2424.95 keV with a 6.85 hrs. half-life that could be excited by the capture of an electron (NEEC) to a higher excited level at 2429.80 keV and subsequently depopulate by a series of E2 transitions. The isomeric state has a spin of  $(21/2)^+$  and the likely depopulation to the  $(13/2)^+$  state is delayed since the transition is required to be of E4 multipolarity. This is illustrated in Fig. 5. The process includes photoexcitation by x-ray free-electron laser beams interacting with a target and a secondary process of electron capture. The results show a significant depletion of the <sup>93m</sup>Mo isomer. NEEC appears to be orders of magnitude more efficient than the direct laser-nucleus interaction. The authors [27] claim that the main process is the production of a plasma where the secondary process of electron capture may take place. The authors of Ref. [25] claim that the predicted cross-sections for such a process show the greatest gain for NEEC which typically spans many orders of magnitude.

There has been intensive work over the years to explore methods of changing the lifetime of an isomer or "nuclear engineering" the wave function of an isomer with x-ray freeelectron lasers [25] or via a process of Nuclear Excitation by Electronic Transition (NEET) [24,30]. There is concrete evidence of shortening the lifetime of the <sup>183m</sup> Ta isomer to an 8 h beta-decaying <sup>180</sup>Ta ground state with by photon bombardment [31-33]. Electron linacs have also been used to reduce the 438-year isomer of <sup>108</sup>Ag to its 2.38 min ground state using 6 MeV bremmstrahlung [34]. There are other examples but to date, none of them have shown the release of the promised energy from the isomers. There were other attempts to modify the lifetimes of isomers by directly exploiting the quantum nature of the isomer wave functions with specially designed schemes. This approach was realized for neutrons, electrons, and atoms [35]. The possibility to twist matter waves by imparting angular momentum on fundamental particles may deliver some promise [36].

Fig. 5 A schematic illustration [25] of the NEEC process for  $^{93m}$ Mo. The idea of isomer triggering is to connect the long-lived isomeric state with a higher level linked to freely radiating states as shown here. The isomeric state is excited by electron capture and decays by E2 transitions (blue)





Eur. Phys. J. A (2023) 59:274 Page 5 of 5 274

Preliminary NEEC theory calculations have shown that nuclear excitations by electron capture can be enhanced by several orders of magnitude if engineered electron beams are used [37,38]. The grand idea is to use a similar technique used to determine the proton spin shown in Ref. [37]. The use of twisted matter waves has been shown by using nanoshaped electromagnetic fields [37]. It is a way of altering the longitudinal part of the wavefunction and therefore affecting the wavefunction of a given isomeric state. The method is potentially very powerful with enormous societal impacts but it is yet to be shown to work.

There is now a new and ambitious proposal [39] of a Gamma Factory (GF) with photons up to  $\approx$ 400 MeV in energy and fluxes of photons, up to  $\approx$ 10<sup>17</sup> photons/s. It is expected that the high-energy (secondary) photons will be produced via resonant scattering of the primary laser photons by highly relativistic partially-stripped ions circulating in the accelerator. The secondary photons would then be emitted in a narrow cone and the energy of the beam can be monochromatized, down to  $10^{-3} \rightarrow 10^{-6}$  level, via collimation. Of course such a severe collimation would be at the expense of photon flux. The GF concept is being discussed within the Physics Beyond Colliders program at CERN.

#### 6 Conclusions

The proposed ideas mentioned here are perhaps the most daring schemes for engineering the nuclear wave functions of metastable or isomeric states. These ideas are possible theoretically but experimentally, they are yet to be implemented. The potential impacts of altering the nuclear structure of states and wave functions range over a broad spectrum from the release of stored energy in isomers to altering the abundances produced in various astrophysical processes. While the new ideas seem very bold and revolutionary, we have tried to show that progress has been evolutionary in nature. Franz Käppeler made similar evolutionary steps that led to a revolutionary understanding of the synthesis of heavy elements by neutron capture. The expertise of Franz Käppeler with neutrons took him from nuclear energy in reactor developments to nuclear astrophysics and our imagination (completely fictional) that he might have applied his expertise to change the lifetimes of metastable nuclear states.

**Acknowledgements** This work was supported by the US National Science Foundation and the US Office of Naval Research.

**Data Availability Statement** Please add a comment to your current Data Availability Statement: "This manuscript has no associated data or the data will not be deposited. [Authors' comment: The data presented here is published in the literature and there is nothing proprietary.]

#### References

- S. Cierjacks, F. Kaeppeler, I. Broeders, F.H. Froehner, B. Goel, E. Kiefhaber, H.W. Wiese, KFK Nachrichten 10(3/4), 54–60 (1978)
- W. Marth, "The story of the European Fast Reactor Collaboration", Karlsruhe Kernforschungszentrum Report KFK5255, (1993)
- 3. A. Aprahamian, Nucl. Phys. News 31, 11 (2021)
- 4. H. Schatz, Phys. Today 61(11), 40–45 (2008)
- E. Margaret Burbidge, G..R. Burbidge, William A. Fowler, F. Hoyle, Rev. Mod. Phys. 29, 547 (1957)
- 6. D. Clayton, M. Rassbach, Astrophys. J. 148, 69 (1967)
- 7. D. Clayton et al., Ann. Phys. 12, 331 (1961)
- 8. D. Clayton, R. Ward, Astrophys. J. 193, 397 (1974)
- 9. F. Käppeler et al., Rev. Mod. Phys. 83, 157 (2011)
- 10. R. Reifarth et al., J. Phys. G Nucl. Part. Phys. 41, 053101 (2014)
- 11. M.M. Busso et al., Front. Astron. Sp. Sci. 9, 956633 (2022)
- 12. I.U. Roderer et al., ApJS 260, 27 (2022)
- 13. C. Arlandini et al., Astrophys. J. 525, 886 (1999)
- 14. John J. Cowan et al., Rev. Mod. Phys. 93, 015002 (2021)
- 15. M. Vilen et al., Phys. Rev. Lett. 120, 262701 (2018)
- 16. R. Orford et al., Phys. Rev. Lett. 120, 262702 (2018)
- 17. M. Mumpower et al., J. Phys. G 44, 034003 (2017)
- 18. P. Mohr, F. Käppeler, R. Gallino, Phys. Rev. C 75, 012802R (2007)
- S. Bizterso et al., Royal Astronomical Society. MNRAS 449, 506 (2015)
- 20. L. D. Hong et al., Kernforschungszentrum report KFK-2337 (1976)
- 21. A. Aprahamian, Y. Sun, Nat. Phys. 1, 81 (2005)
- 22. G.C. Baldwin et al., Rev. Mod. Phys. 53, 687 (1981)
- 23. M.M. Busso et al., Astrophys. J. 908, 55 (2021)
- 24. P.M. Walker, James J. Carroll, Phys. Today 58, 39 (2005)
- 25. J. Gunst et al., Phys. Plasma 22, 112706 (2015)
- P.M. Walker, Z. Podolyák, Chapter on nuclear isomers, in *Hand-book of Nuclear Physics*. ed. by I. Tanihata et al. (Springer Nature Physics) (2022)
- 27. C.J. Chiara et al., Nature 554, 216 (2018)
- 28. V.I. Goldanskii, V.A. Namiot, Phys. Lett. B 62, 393 (1976)
- 29. G. Gosselin, P. Morel, Phys. Rev. C 70, 064603 (2004)
- 30. P. Morel et al., AIP Conf. Proc. 769, 1085 (2005)
- 31. D. Belic et al., Phys. Rev. C 65, 35801 (2002)
- 32. P.M. Walker, G.D. Dracoulis, Hyperfine Interact. 135, 83 (2001)
- 33. Y. Litvinov et al., Phys. Lett. B 573, 80 (2003)
- 34. J.J. Carroll et al., AIP Conf. Proc. 1525, 586 (2013)
- 35. C.W. Clark et al., Nature 525, 504 (2015)
- 36. H. Larocque et al., Nat. Phys. 14, 1 (2018)
- 37. G.M. Vanacore et al., Nat. Mater. 18, 573 (2019)
- 38. G.M. Vanacore et al., Riv. Nuovo Cimento 43, 567-597 (2020)
- 39. D. Budker et al., Ann. Phys. 534, 2100284 (2022)

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

