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## New DDEP recommendations for the radioactive decay scheme of $^{137}\text{Cs}$

Sylvain Leblond

► **To cite this version:**

Sylvain Leblond. New DDEP recommendations for the radioactive decay scheme of  $^{137}\text{Cs}$ . ICRM 2023 - 23rd International Conference on Radionuclide Metrology and its Applications, Mar 2023, Bucarest, Romania. cea-04256925

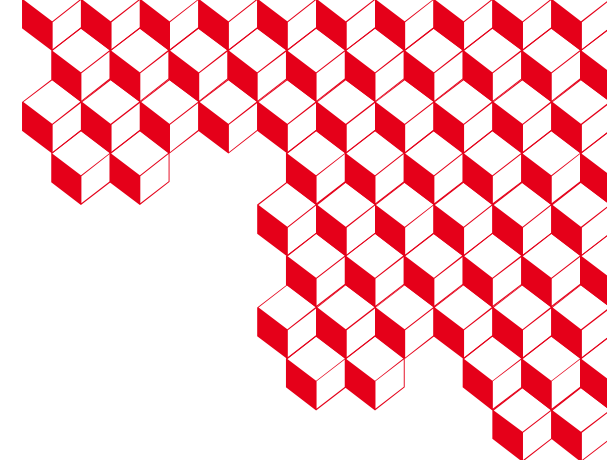
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Submitted on 24 Oct 2023

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# New DDEP recommendations for the radioactive decay scheme of $^{137}\text{Cs}$

S. Leblond



# Discovery of an iconic radionuclide

- In **1941** at **Berkeley**
  - Margaret Melhase and Glenn Seaborg
  - First chemical extraction of radioactive Caesium
  - Produced by thermal neutron induced fission on  $^{235}\text{U}$
- Since then...



(1941)



(1950)

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  - More than 80 000 publications in a wide range of applications



(1941)

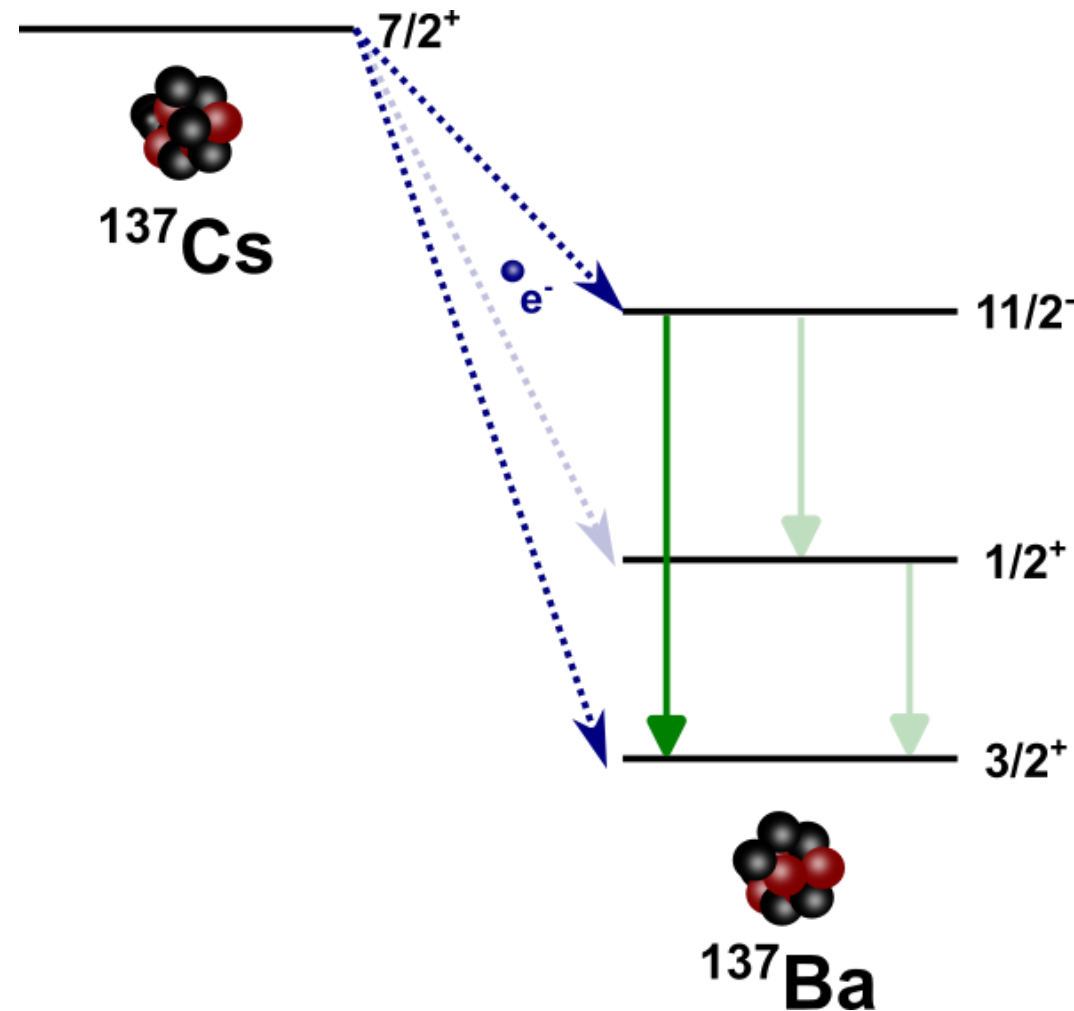


(1950)



# Decay overview of $^{137}\text{Cs}$

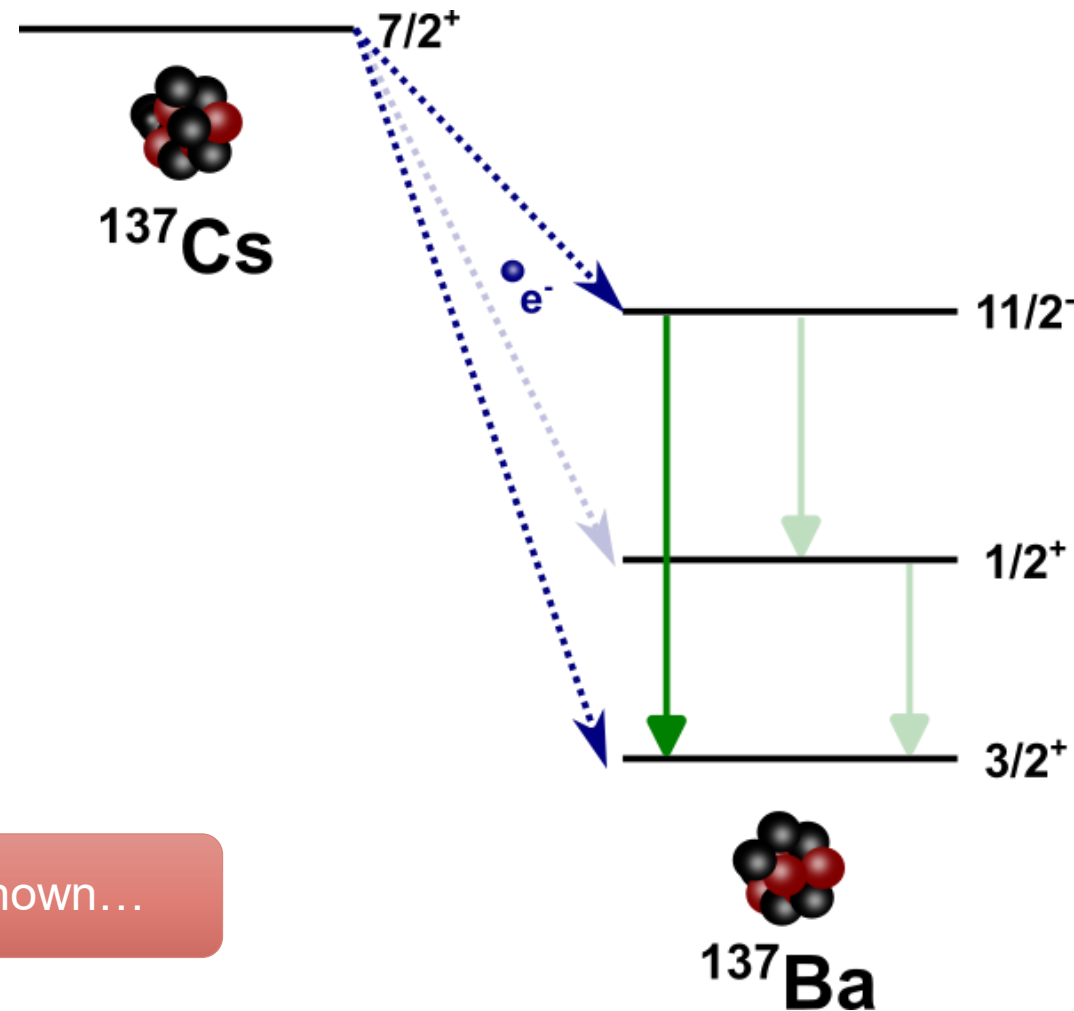
- A  $\beta^-$  decay to  $^{137}\text{Ba}$ 
  - Populates ground & excited states
  - Half-life around 30 years
  - Total Q-value close to 1176 keV
- Emissions
  - Two main  $\beta^-$  branches
    - Ground state ( $E_{\text{max}} \sim 1176 \text{ keV}$ )
    - Second excited state ( $E_{\text{max}} \sim 514 \text{ keV}$ )
  - Iconic 662 keV gamma



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Surely decay data are well known...



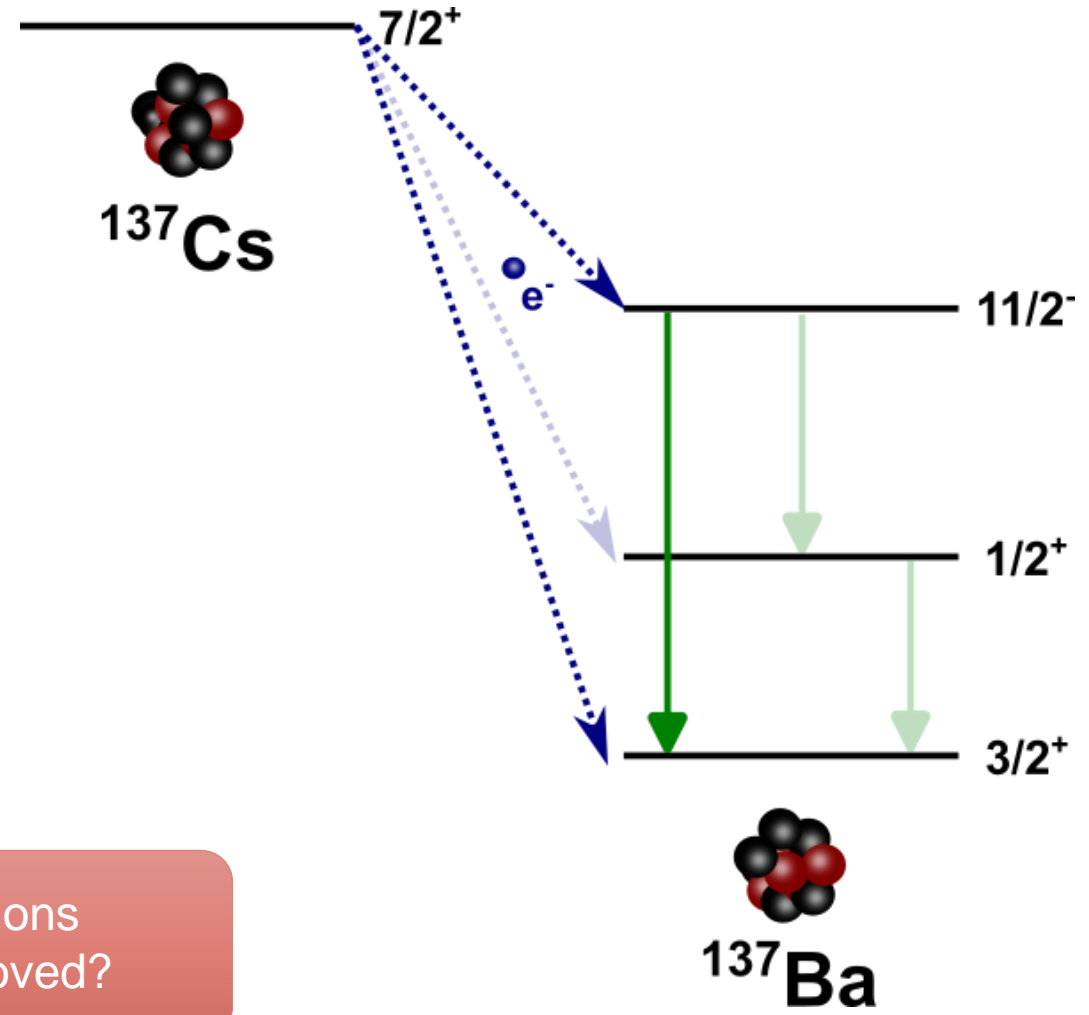


# Decay data status

- Latest  $^{137}\text{Cs}$  decay data evaluations
  - DDEP
    - R.G. Helmer and V.P. Chechev (2006)
  - ENSDF
    - E. Browne and J.K. Tuli (2007)

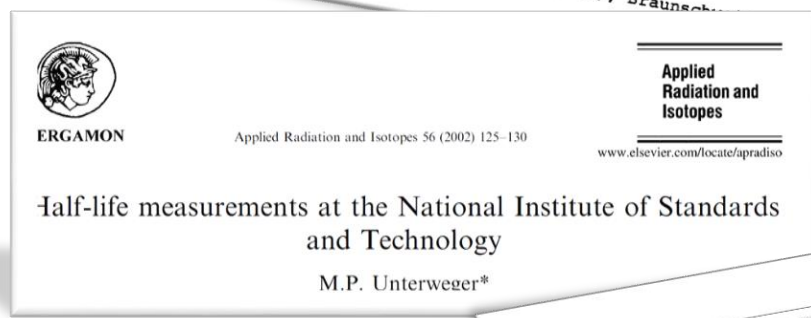
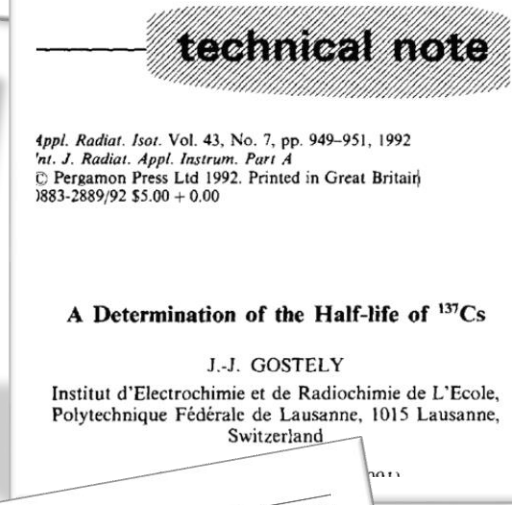
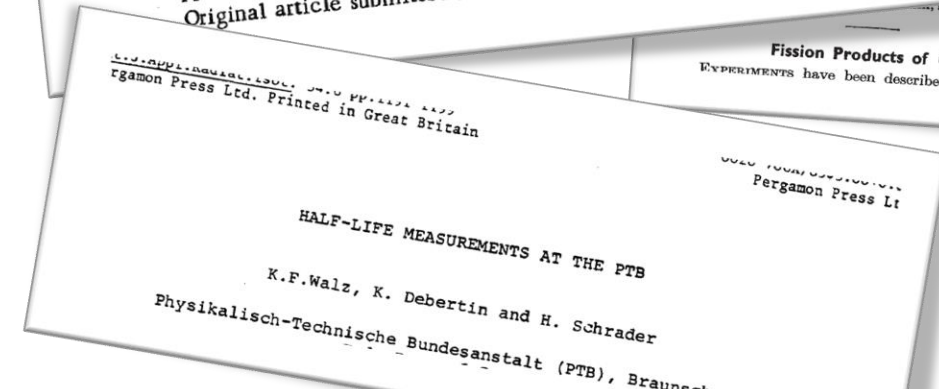
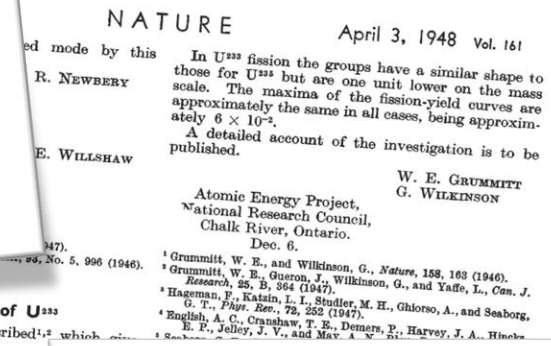
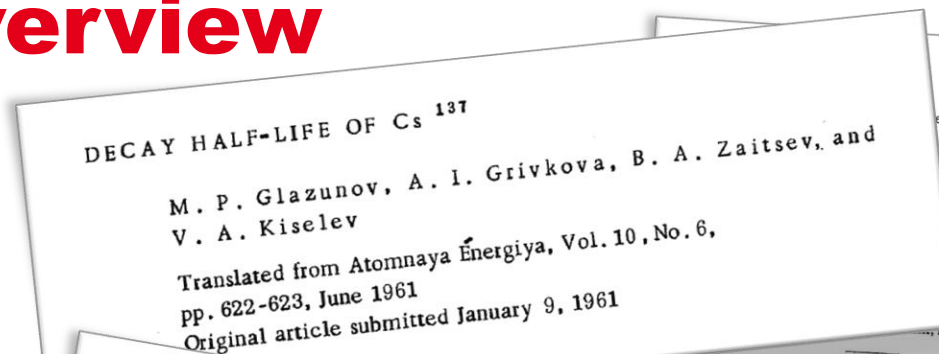
	DDEP	ENSDF
$T_{1/2}$ (y)	30.05 (8)	30.08 (9)
$I_{\beta}(11/2^-)$ (%)	94.36 (28)	94.7 (2)
$I_{\beta}(3/2^+)$ (%)	5.64 (28)	5.3 (2)
$I_{\gamma}$ (%)	84.99 (20)	85.1 (2)

New publications  
 Can it be improved?



# Half-life of $^{137}\text{Cs}$ : overview

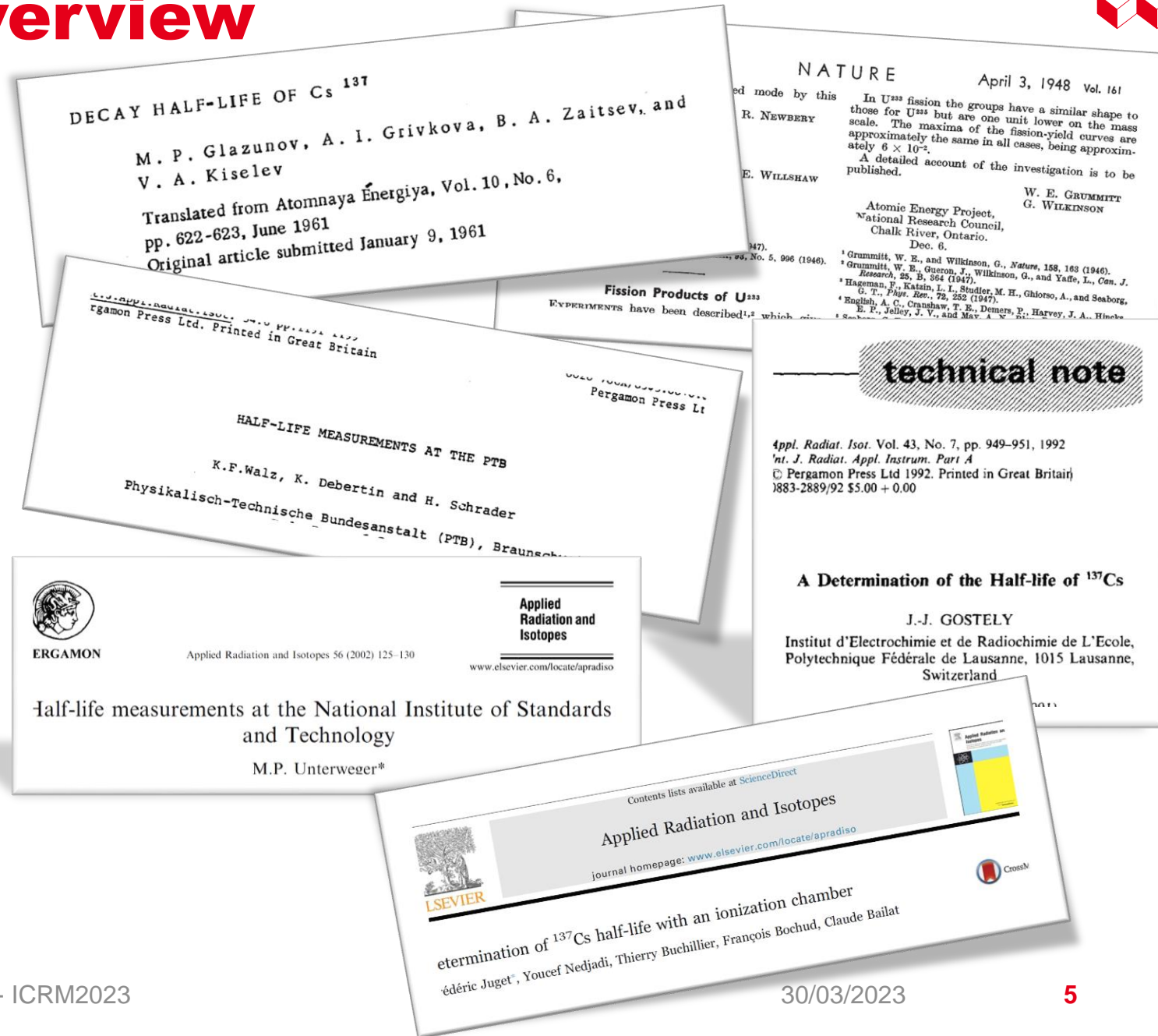
- Abundant bibliography
  - More than 75 publications
  - From 1948 to 2016
  - Different experimental methods
  - Many languages
- How to select a meaningful dataset?





# Half-life of $^{137}\text{Cs}$ : overview

- Abundant bibliography
  - More than 75 publications
  - From 1948 to 2016
  - Different experimental methods
  - Many languages
- How to select a meaningful dataset?
  - DDEP guidelines
    - Discard measurements without uncertainty estimation
    - One measurement per author / laboratory

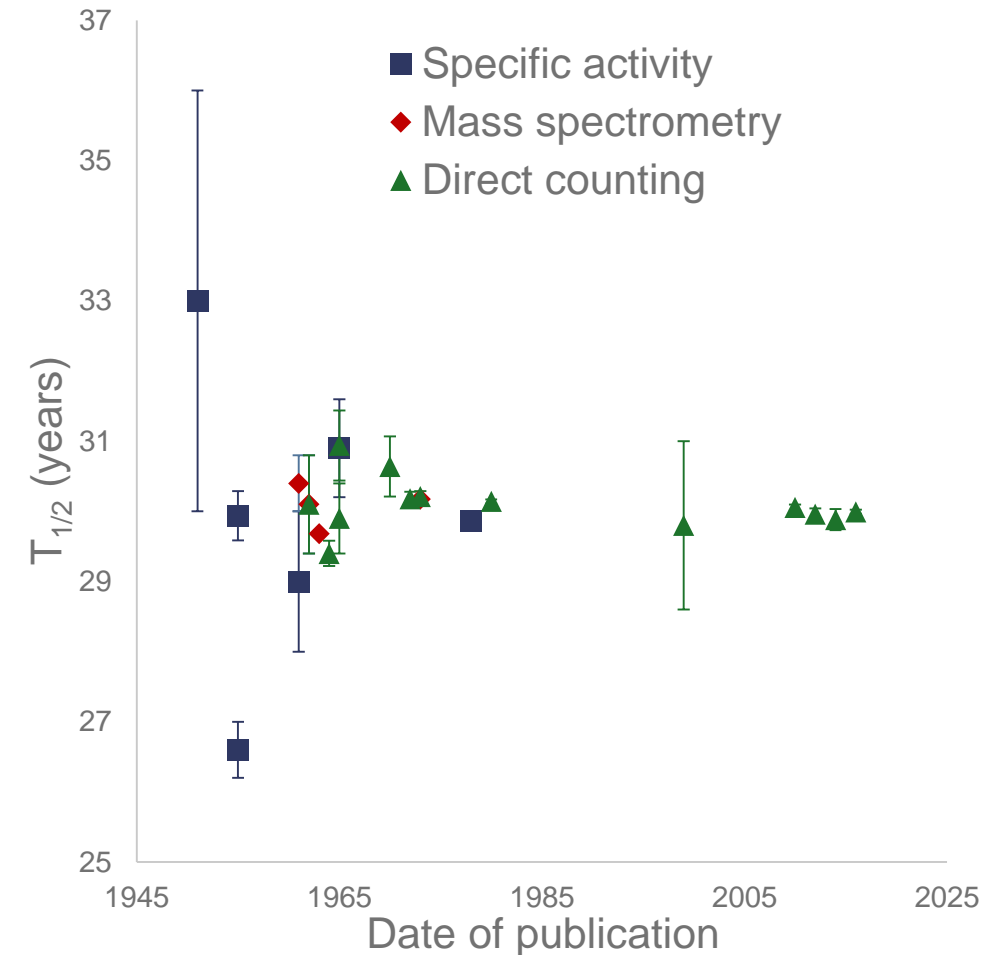


# Half-life of $^{137}\text{Cs}$ : a first dataset to work with

- Highly discrepant
  - 23 references considered
  - $T_{1/2}$  varying from 26 to 33 years
  - Uncertainties from 0.011 to 3 years
  - Statistical test of the distribution:

$$\chi^2 \sim 11 \gg \chi_{crit}^2 \sim 1.9$$

$$\chi^2 = (n - 1) \frac{\sigma_{ext}^2}{\sigma_{int}^2}$$



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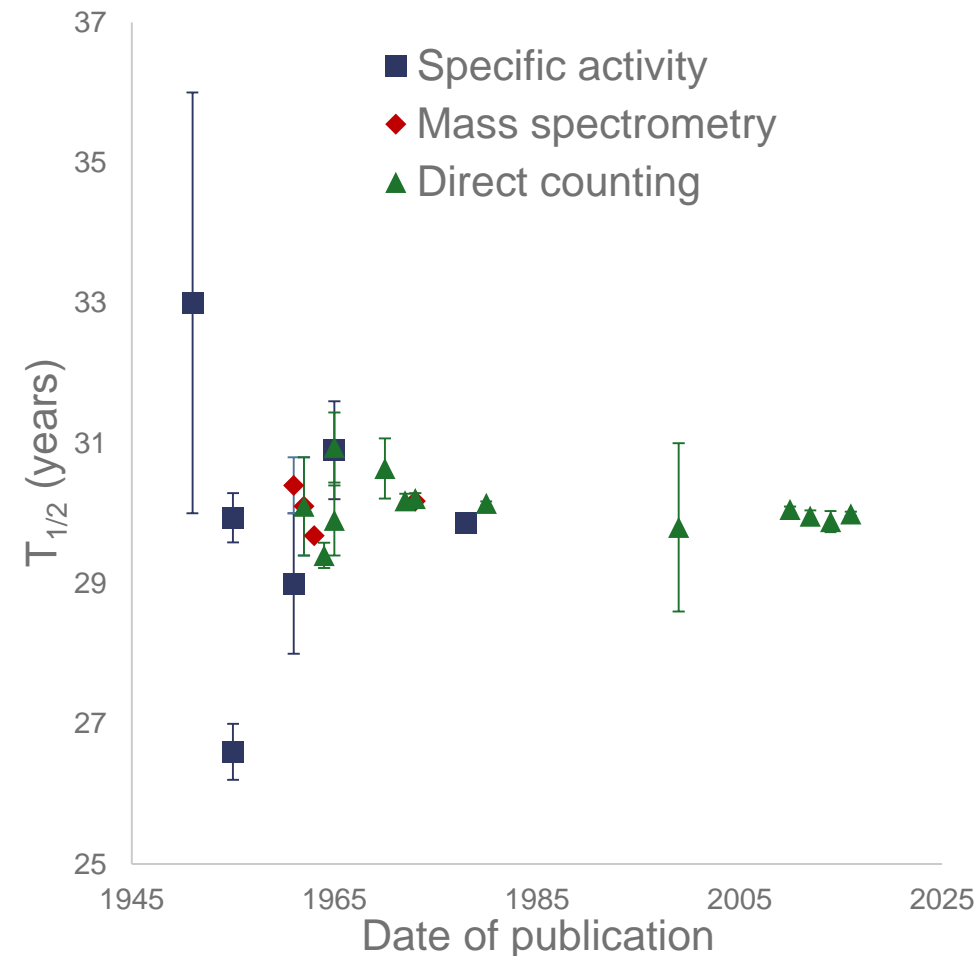
$$\chi^2 \sim 11 \gg \chi_{crit}^2 \sim 1.9$$

$$\chi^2 = (n - 1) \frac{\sigma_{ext}^2}{\sigma_{int}^2}$$

- Unweighted average  $T_{1/2} = 30.09$  (8) a

Reminder:		DDEP	ENSDF
$T_{1/2}$ (a)		30.05 (8)	30.08 (9)

How to proceed further?



# Cs-137 Half-life: final refinement

- Critical analysis of each publication
  - Evaluate possible experimental bias
  - Evaluate uncertainty budget assessment

- Support from previous works

M.J. Woods, *The Half-life of  $^{137}\text{Cs}$  – A critical Review*, NIM A286 (1990)

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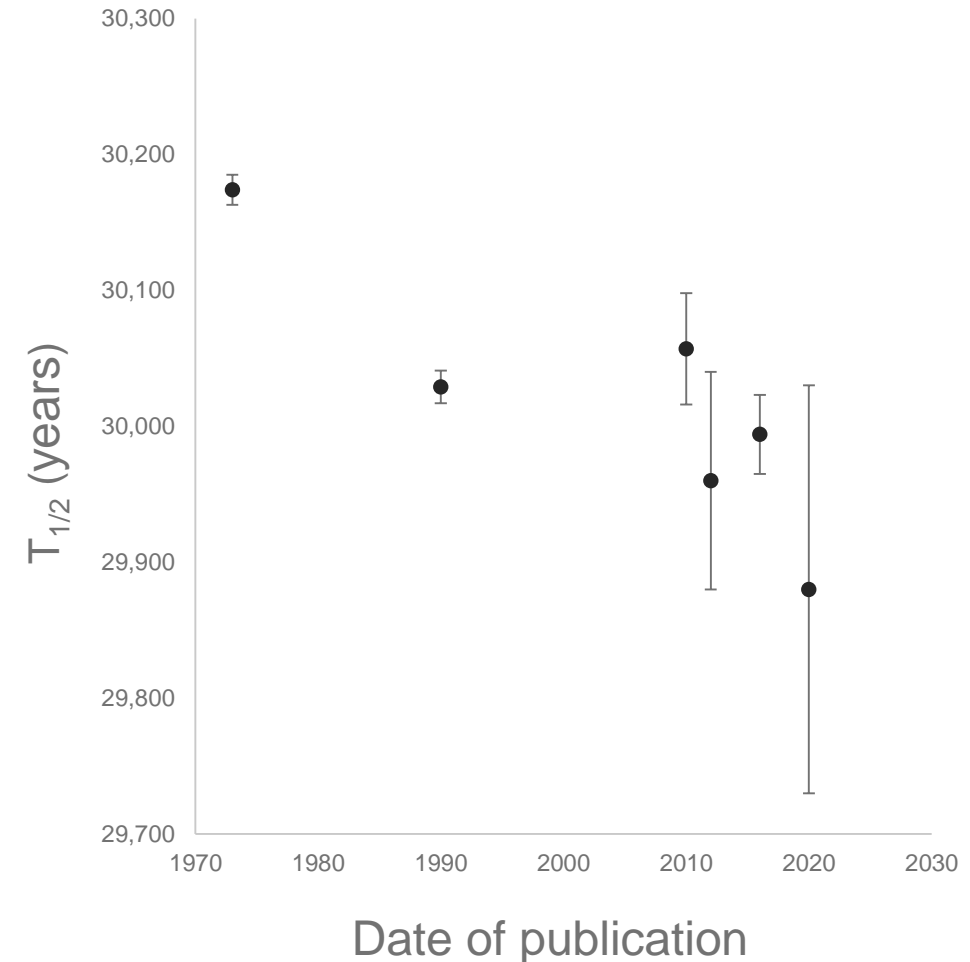
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- Selected publications

■ <i>Dietz et al.</i> , (1973)	GEC	$T_{1/2} = 30.174 (11) \text{ a}$
■ <i>Martin et al.</i> , (1990)	CRNL	$T_{1/2} = 30.029 (12) \text{ a}$
■ <i>Schrader et al.</i> , (2010)	PTB	$T_{1/2} = 30.06 (4) \text{ a}$
■ <i>Bellotti et al.</i> , (2012)	INFN	$T_{1/2} = 29.96 (8) \text{ a}$
■ <i>Juget et al.</i> , (2016),	IRP	$T_{1/2} = 29.994 (29) \text{ a}$
■ <i>Unterweger et al.</i> , (2020)	NIST	$T_{1/2} = 29.88 (15) \text{ a}$



# Cs-137 Half-life: final refinement

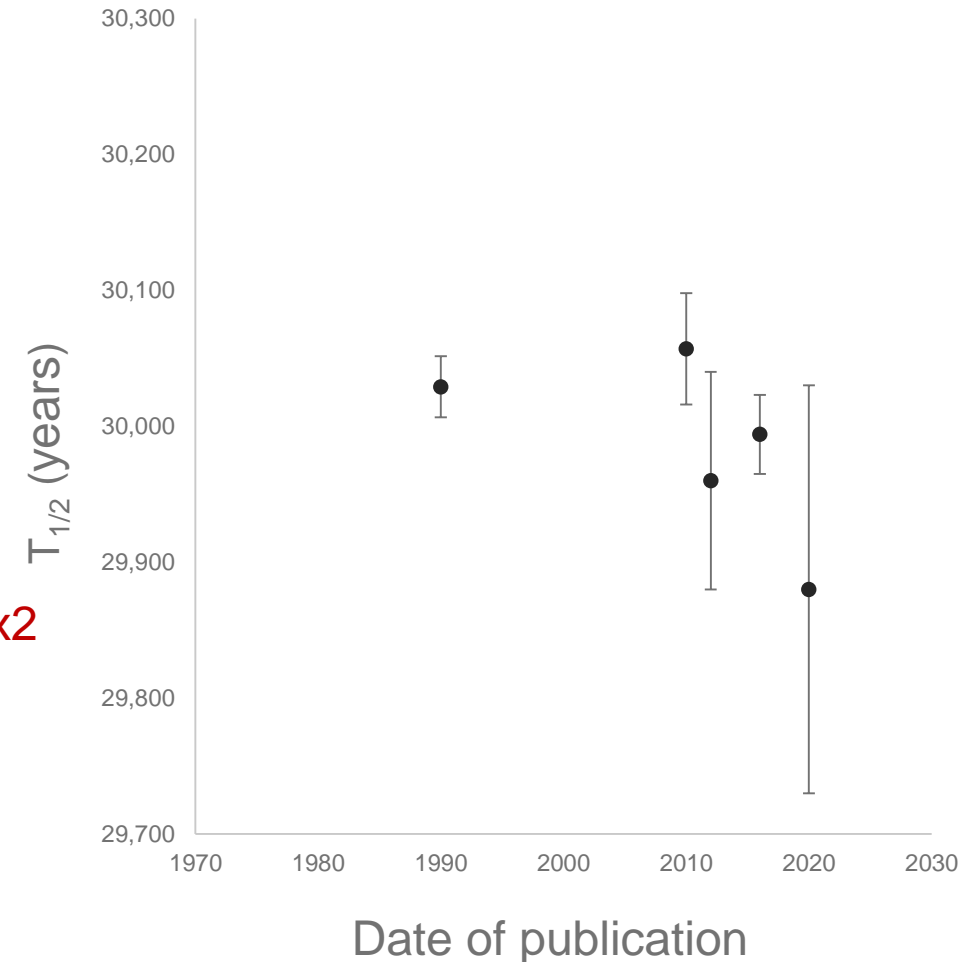
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# Cs-137 Half-life: new DDEP recommendation

- A final dataset of 5 measurements
  - Consistent:  $\chi^2 \sim 0,8 < \chi_{crit}^2 \sim 3.3$
  - Unweighted mean  $T_{1/2} = 29.984 (30) \text{ a}$
  - Weighted mean  $T_{1/2} = 30.018 (16)_{int} (14)_{ext} \text{ a}$
  - Recommended uncertainty extended to match *Martin et al.* (1990)

$$T_{1/2} = 30.018 (22) \text{ a}$$

	DDEP	ENSDF	This work
$T_{1/2} \text{ (a)}$	30.05 (8)	30.08 (9)	<b>30.018 (22)</b>

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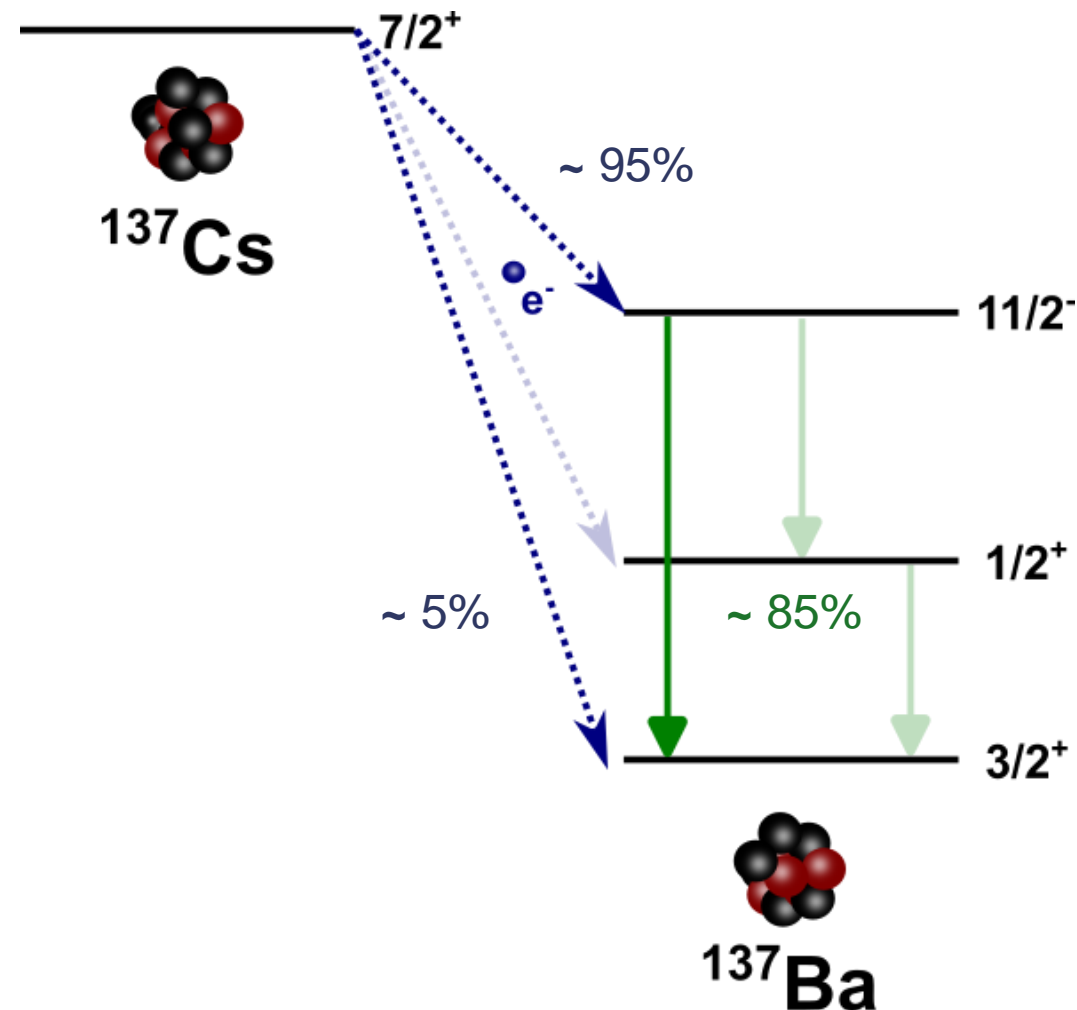
$$T_{1/2} = \mathbf{30.018 (22) \text{ a}}$$

- Outlooks
  - Lower dependency to *Martin et al.*
    - 50% of the weight
  - Need for additional measurements
    - Unpublished results ?
      - PTB (continuation of Schrader works)
      - NRC (IC from 1995 2009)

	DDEP	ENSDF	This work
$T_{1/2} \text{ (a)}$	30.05 (8)	30.08 (9)	<b>30.018 (22)</b>

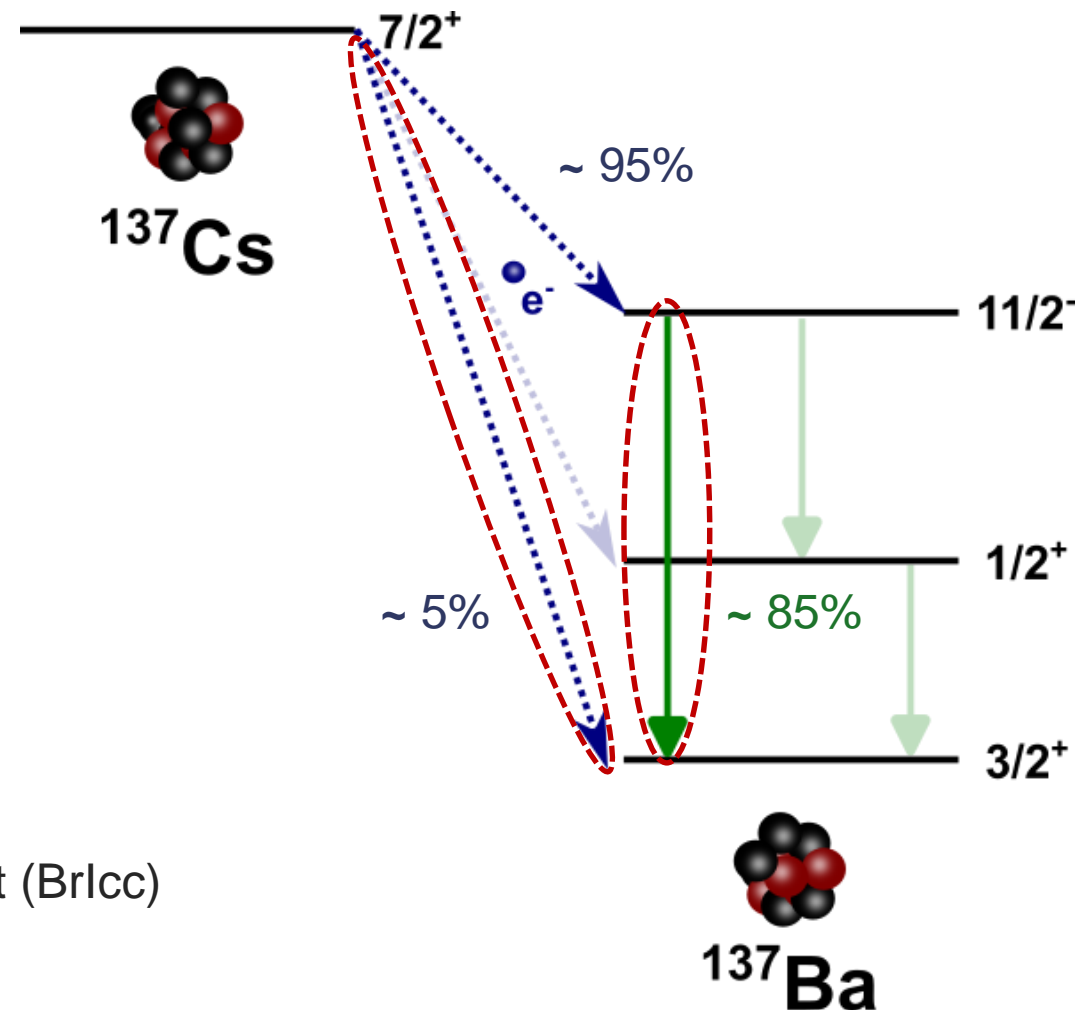
# Decay scheme of $^{137}\text{Cs}$ : overview

- A simple decay scheme
  - Three main transitions
    - $^{137}\text{Cs} \rightarrow ^{137}\text{Ba} (11/2^-)$   $\sim 95\%$
    - $^{137}\text{Cs} \rightarrow ^{137}\text{Ba} (3/2^+)$   $\sim 5\%$
    - $^{137}\text{Ba} (11/2^-) \rightarrow ^{137}\text{Ba} (3/2^+)$   $\sim 85+10\%$  ( $\gamma + \text{EC}$ )
  - Three negligible transitions
    - $^{137}\text{Cs} \rightarrow ^{137}\text{Ba} (1/2^+)$   $\sim 6 \cdot 10^{-4} \%$
    - $^{137}\text{Ba} (11/2^-) \rightarrow ^{137}\text{Ba} (1/2^+)$   $\sim 1 \cdot 10^{-5} \%$
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    - $^{137}\text{Ba} (1/2^+) \rightarrow ^{137}\text{Ba} (3/2^+)$   $\sim 6 \cdot 10^{-4} \%$
- Knowledge of one of the main transitions is enough
  - Completeness of the decay scheme
  - Assuming knowledge of the  $\alpha_T$  conversion coefficient (BrIcc)



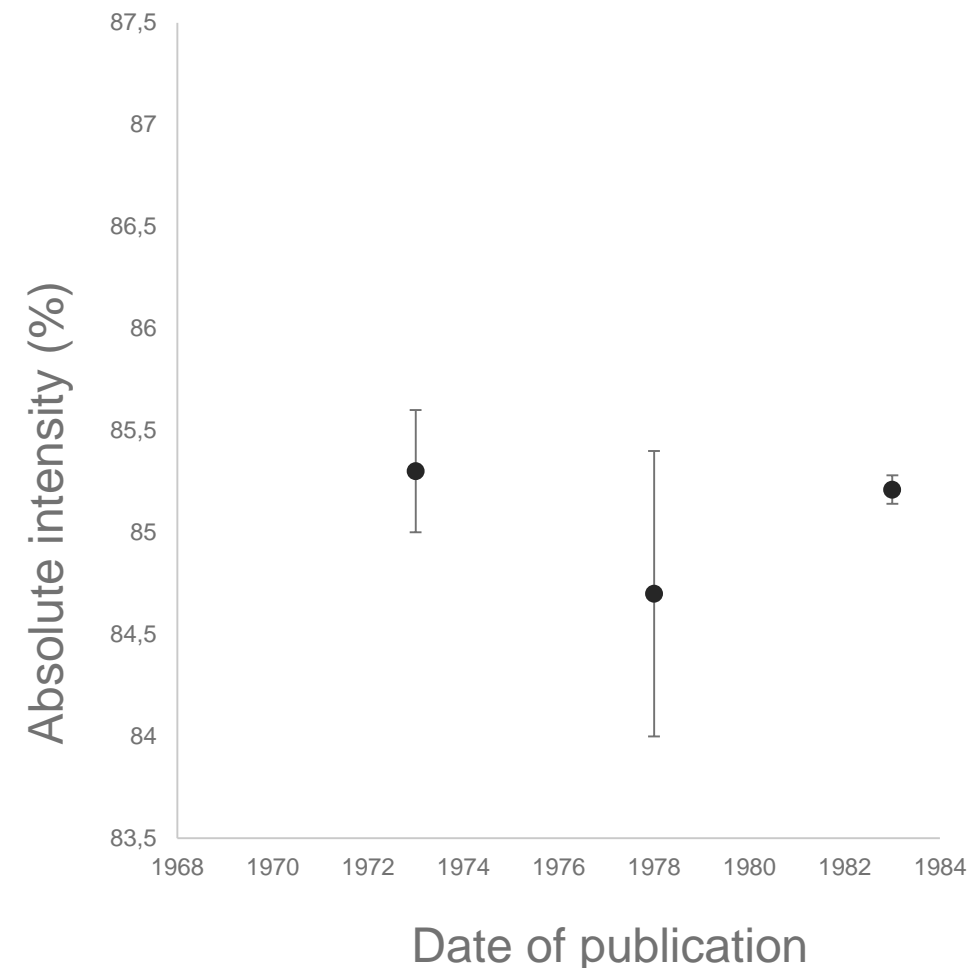
# Intensity of the 662 keV gamma emission

- A limited dataset

- Only six publication (from 1965 to 1983)

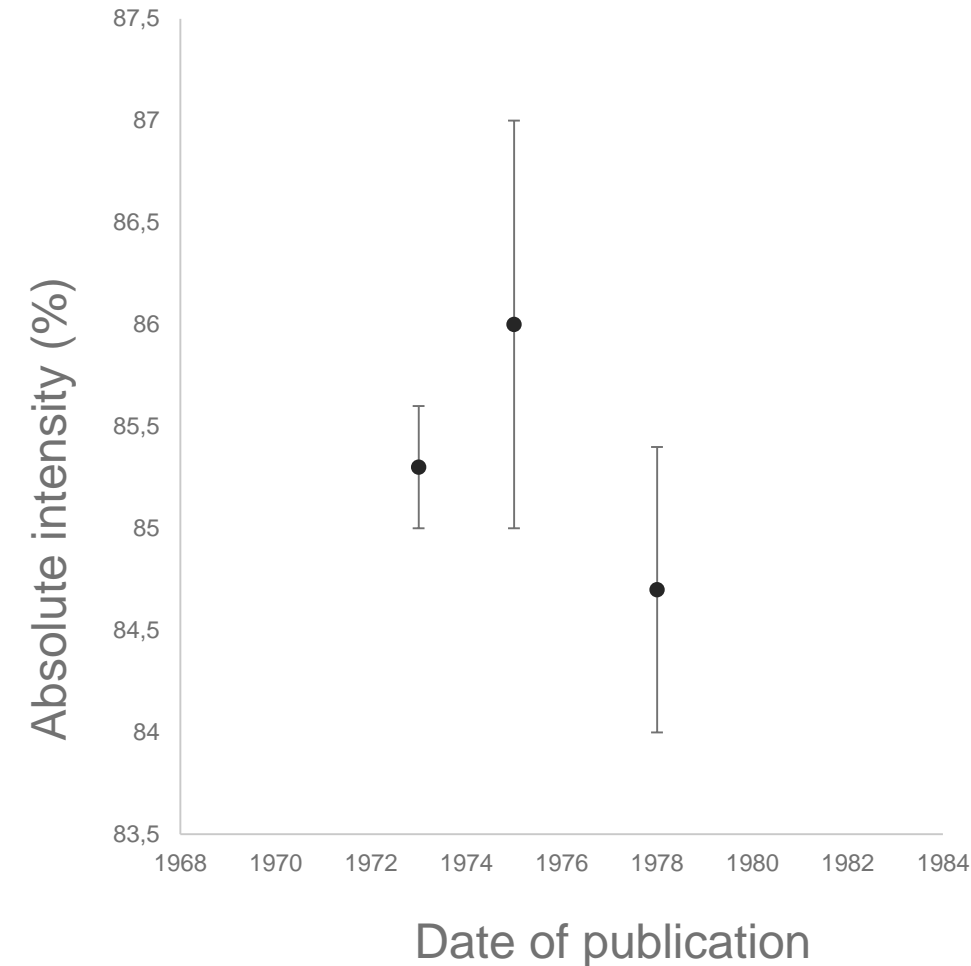
- Three remains after applying DDEP criteria

- *Legrand et al.* (1973)      CEA       $I_{\gamma} = 85.3 (3) \%$
- *Merritt et al.* (1978)      CRNL       $I_{\gamma} = 84.7 (7) \%$
- *Behrens et al.* (1983)      NPL       $I_{\gamma} = 85.21 (7) \%$



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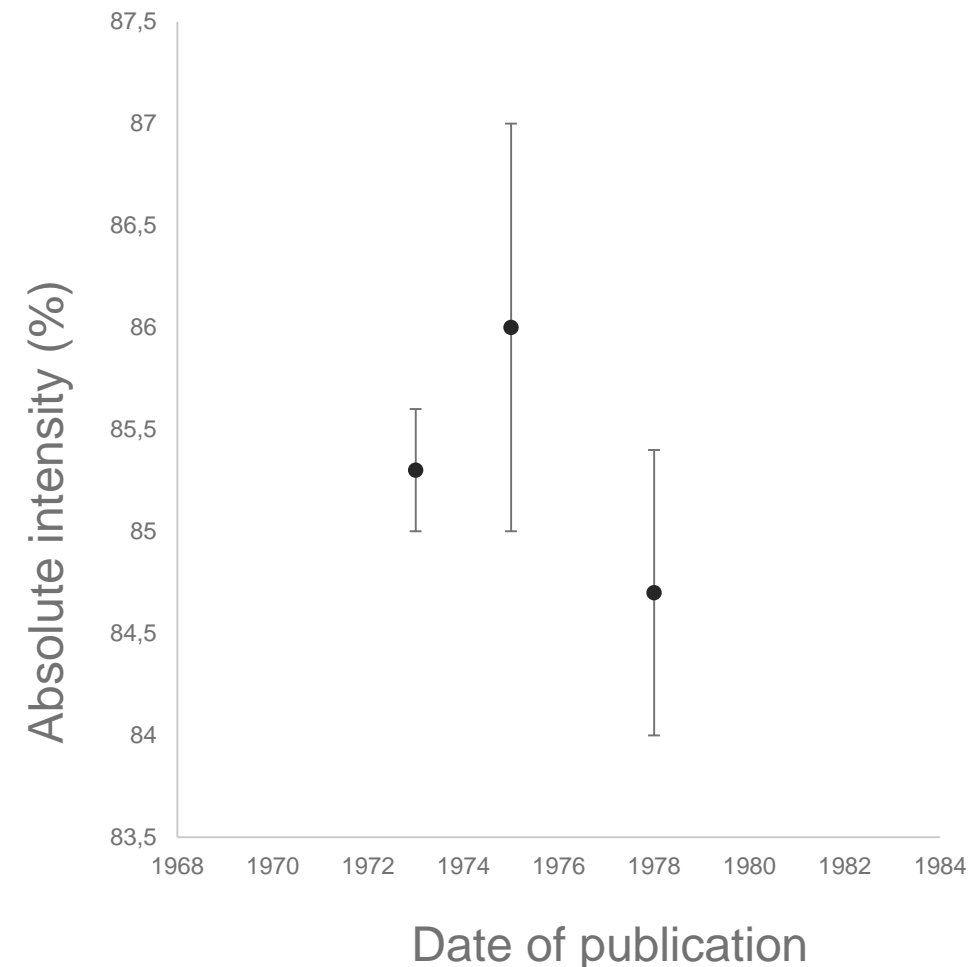
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- Consistent dataset but an issue with *Behrens et al.*
  - Estimated using magnetic spectrometer
  - Usage of discrepant  $\alpha_T$  conversion coefficient
  - Replaced by Goodier et al. (1975)  $I_{\gamma} = 86$  (1) %





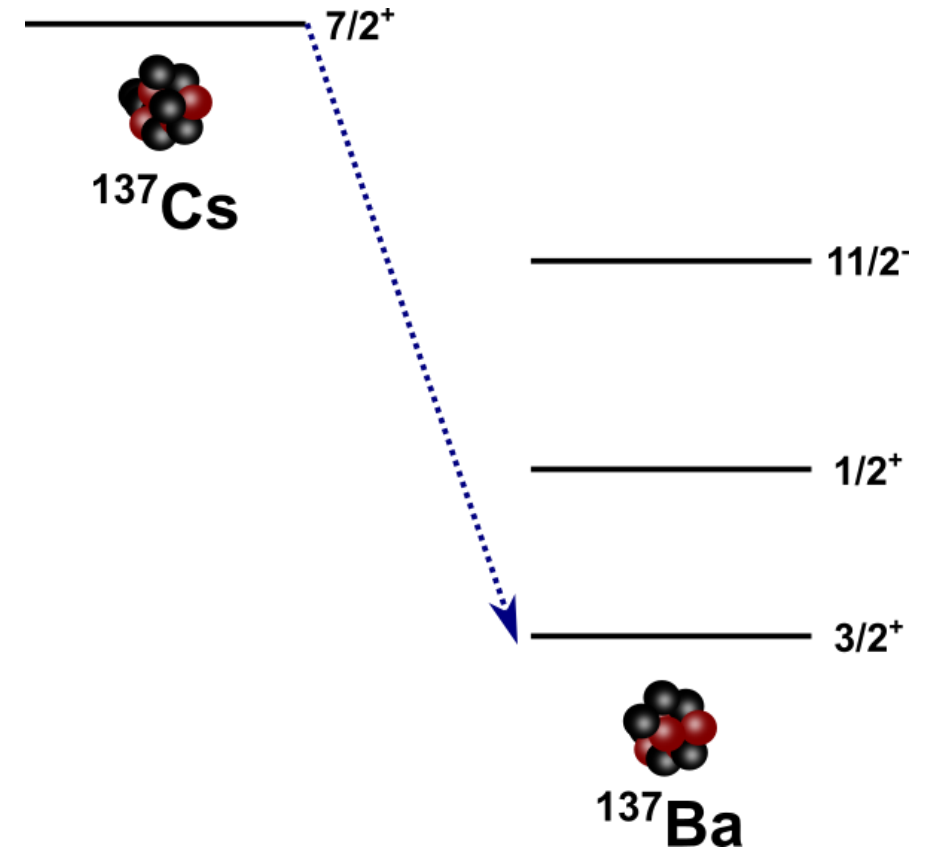
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  - Estimated using magnetic spectrometer
  - Usage of discrepant  $\alpha_T$  conversion coefficient
  - Replaced by Goodier et al. (1975)  $I_{\gamma} = 86 (1) \%$
- Weighted mean  $I_{\gamma} = 85.26(30) \%$ 
  - Weight of *Legrand et al.* 78 %



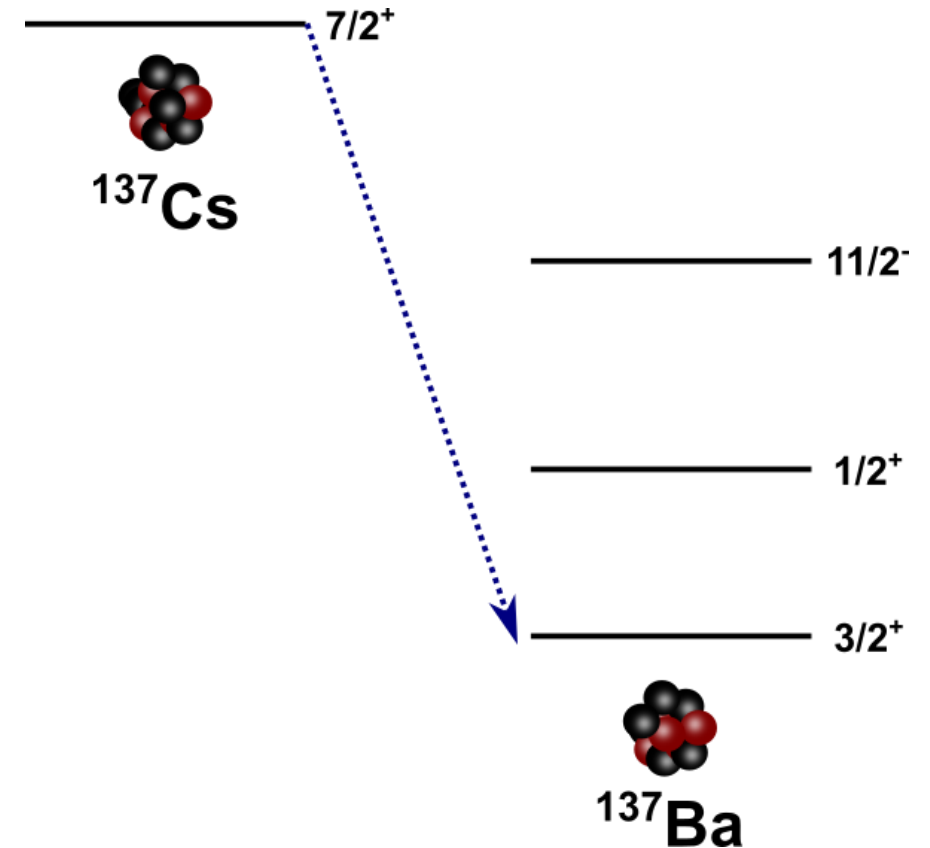
# Intensity of the beta to ground state transition

- Another limited dataset
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  - Four of them are discarded after analysis
  - Only two measurements available
    - *Hansen et al.* (1969)      CBNM       $I_{\beta} = 5.4 (3) \%$
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# Intensity of the beta to ground state transition

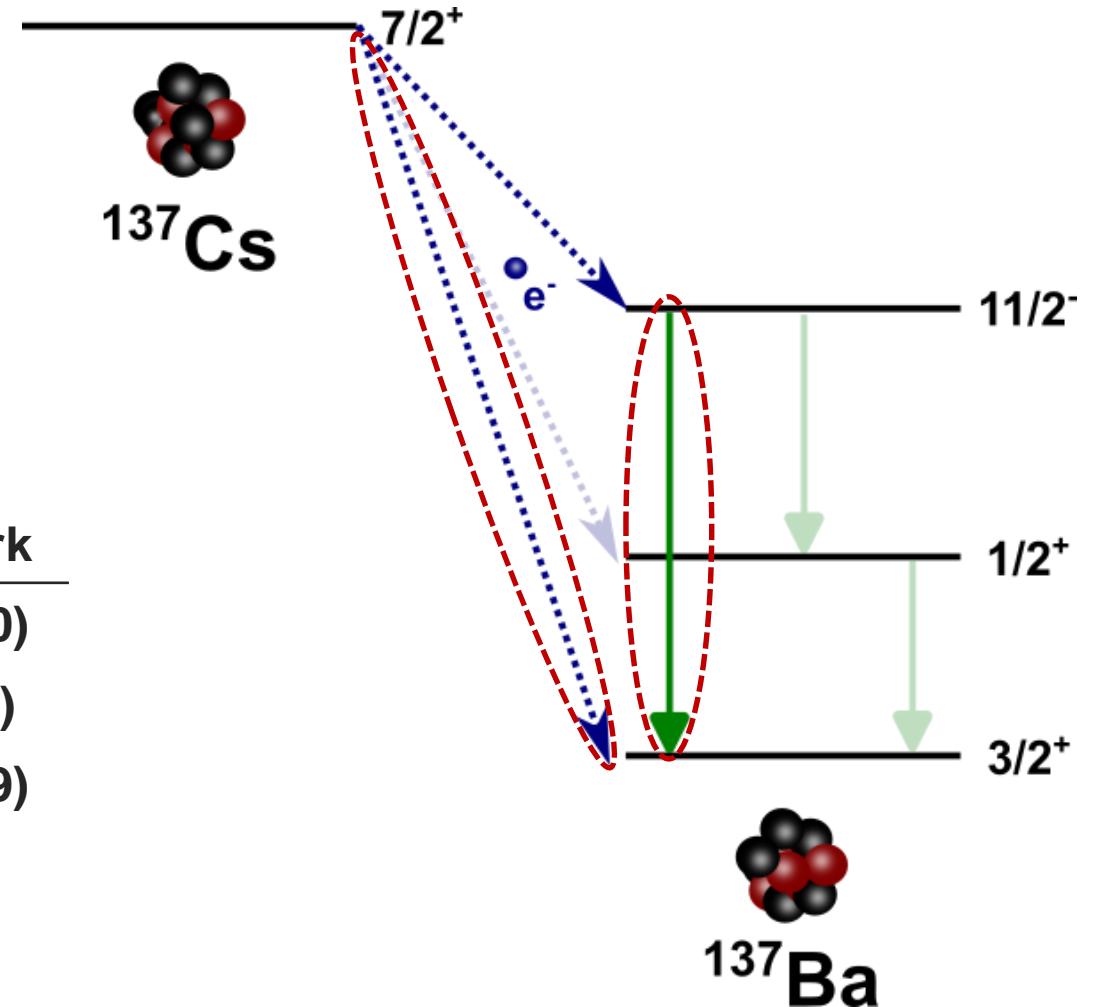
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- Question regarding the measurement of *Behrens et al.*
  - Inaccurate measurement of  $\alpha_T$  conversion coefficient
  - Limitation of weight of *Behrens et al.* to 50 %
    - *Behrens et al.* (1983)      NPL       $I_\beta = 5.57 (30) \%$
- Average       $I_\beta = 5.48 (30) \%$



# Decay scheme reconstruction

- Two main possibilities
  - From 662 keV gamma intensity
    - $I_\gamma = 85.26$  (30) %
  - From the beta decay to the ground state of  $^{137}\text{Ba}$ 
    - $I_\beta = 5.48$  (30) % corresponds to  $I_\gamma = 84.97$  (29) %
  - Final recommendation: weighted average of both

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$I_\beta(11/2^-)$ (%)	94.36 (28)	94.7 (2)	<b>94.65 (30)</b>
$I_\beta(3/2^+)$ (%)	5.64 (28)	5.3 (2)	<b>5.35 (30)</b>
$I_\gamma$ (%)	84.99 (20)	85.1 (2)	<b>85.09 (29)</b>

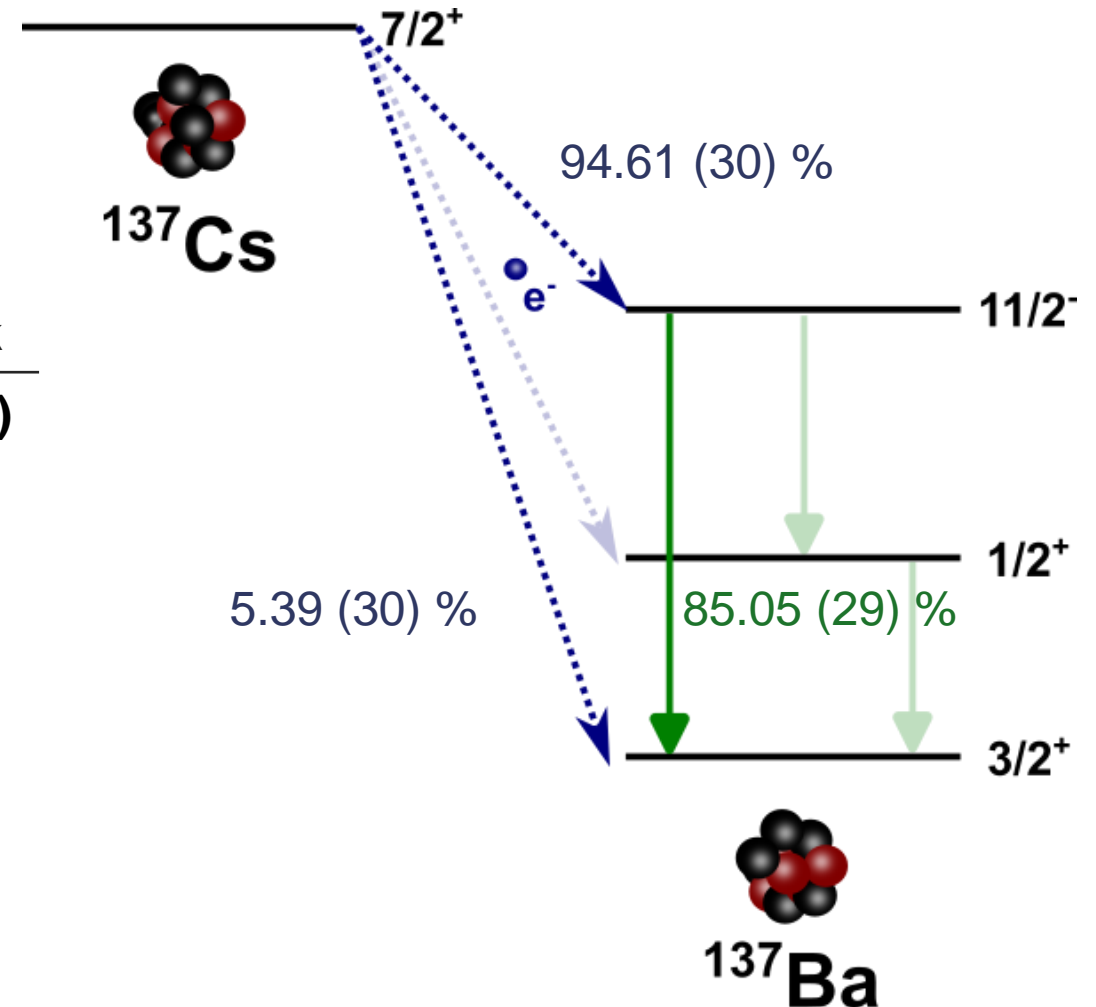


# Conclusions & outlooks

- A new evaluation of  $^{137}\text{Cs}$  has been performed
- Recommendations are consistent with previous evaluations

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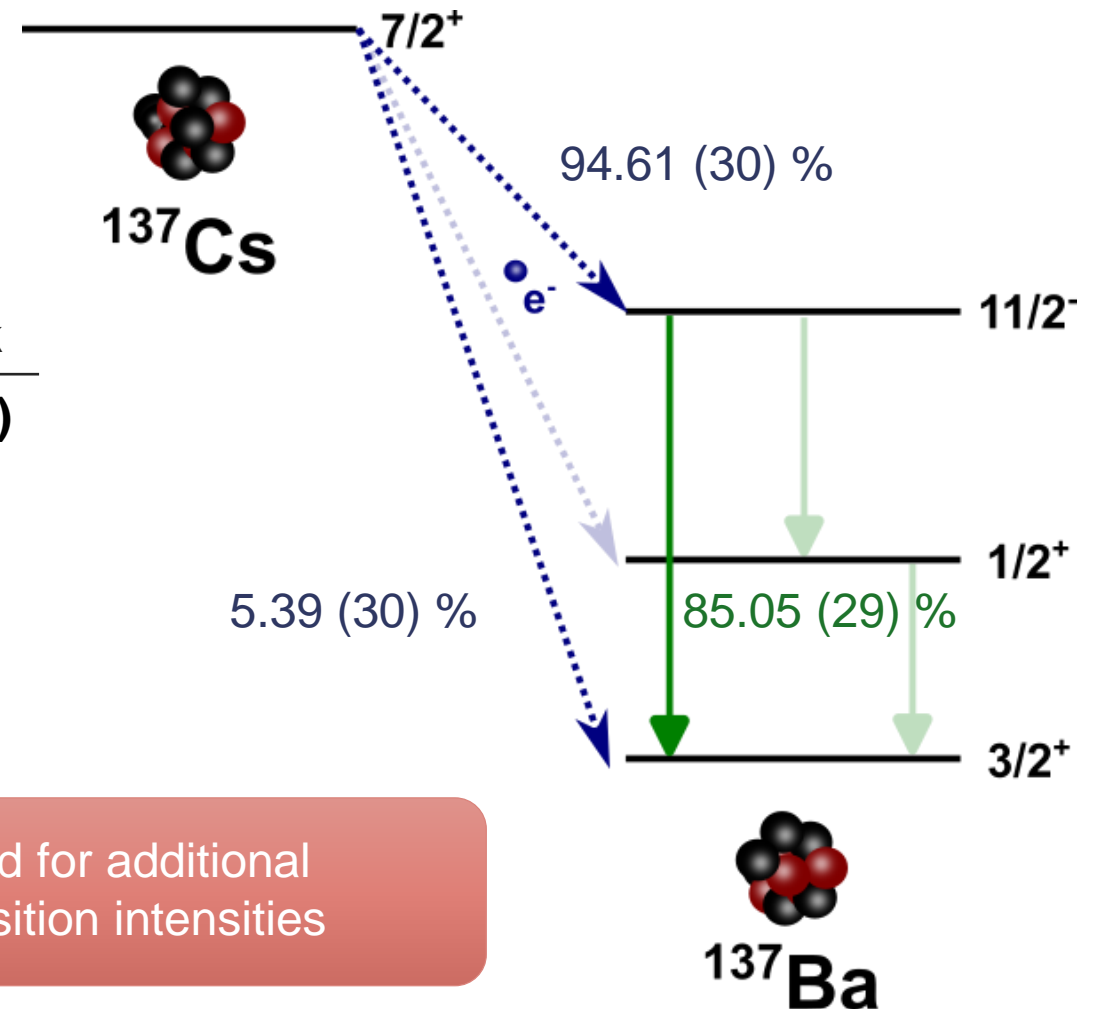
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Analysis of datasets has shown the need for additional measurements for both half-life and transition intensities

$$T_{1/2} = 30.018 (22) \text{ a}$$





# Acknowledgment

- DDEP collaboration
  - M.A. Kellett & X. Mougeot
  - My very patient reviewer: A. L. Nichols
- Previous evaluations
  - M.J. Woods (1990)
  - R.G. Helmer and V.P. Chechev (2006)
- *How Cesium-137 was discovered by an undergraduate student ?*  
(Dennis D. Patton, The journal of nuclear Medicine 40 1999)