Measurement of the ^{93}Zr capture cross-section at n_TOF facility at CERN

G.Tagliente

INFN – Bari (Italy)

n_TOF Collaboration

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40 Research Institutions

120 researchers

Outline

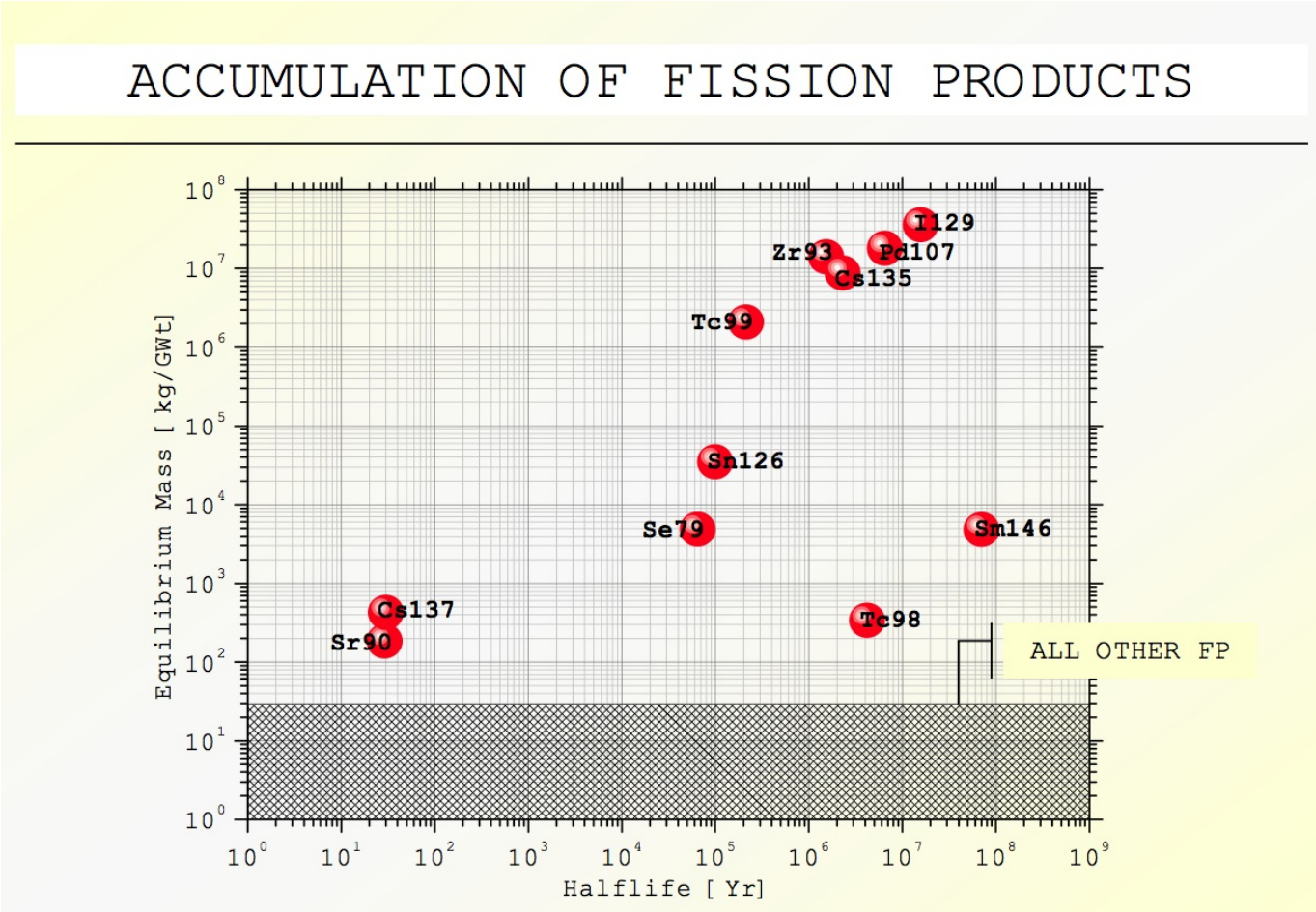
- **Scientific Motivations**
- **n_TOF facility**
- **Measurement**
- **Data Analysis and Results**
- **Astrophysical implication**
- **Conclusions**

Scientific Motivations

- Nuclear Technology

- Nuclear Astrophysics

Nuclear Technology



PWR 33GWtd THM^{-1} burnup

3 yr cooling spent fuel

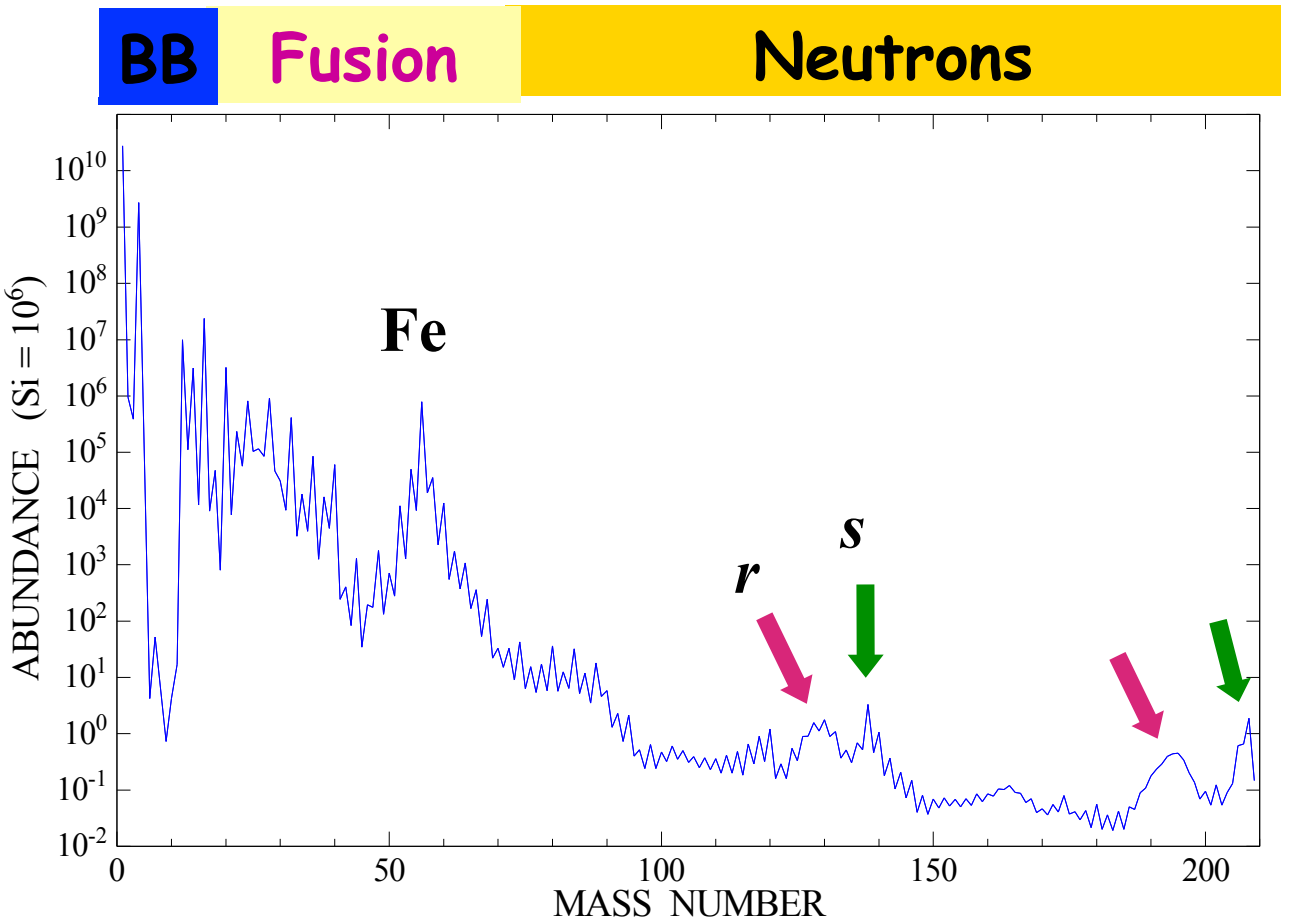
Nuclear Technology

- Partitioning and transmutation of nuclear wastes should make possible to reduce both the size of the repository for nuclear wastes and the long term risk. Fission products, such as ^{93}Zr , are considered as candidates for transmutation otherwise their very long half-lives necessitate storage in a repository for extremely long times.

- Neutron capture cross section of ^{93}Zr, as well as other long-lived fission products, is required for transmutation studies, as well as for nuclear reactor design purposes.

- Formal requests have been made for 5% uncertainties on capture cross sections of ^{93}Zr.

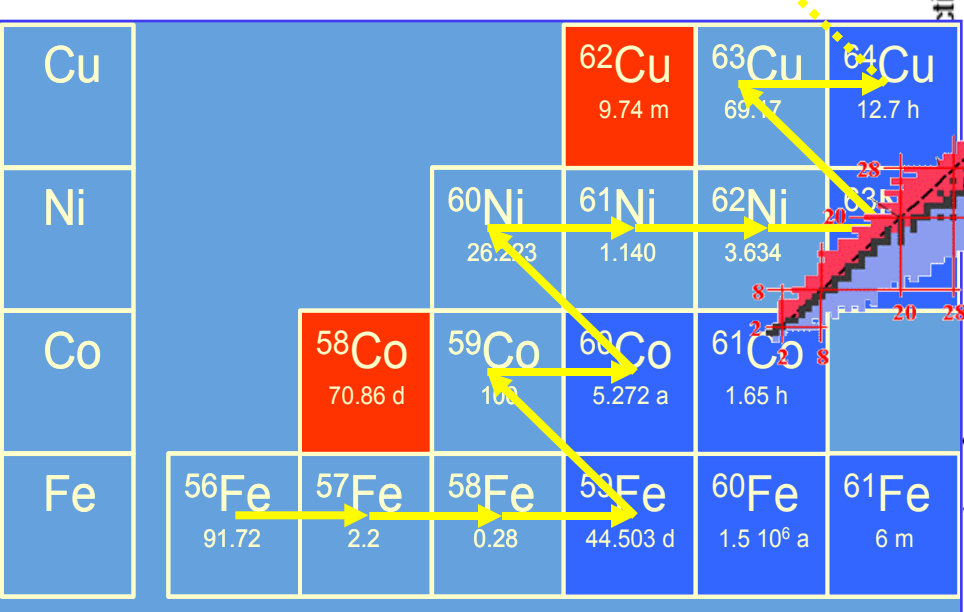
Abundances beyond Fe–ashes of stellar burning



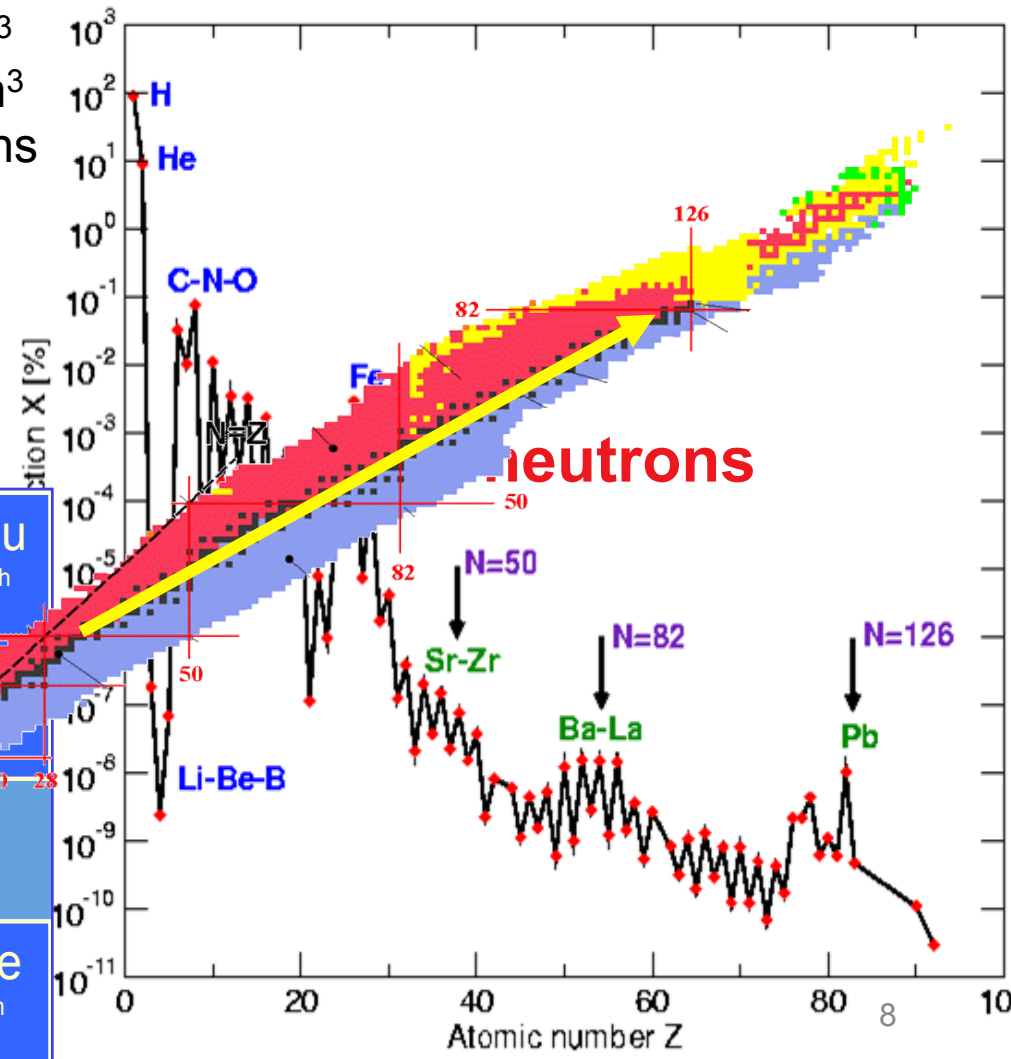
The s-process

s-process lifetime 10^4y $n_n \approx 10^8$neutron/cm^3
r-process lifetime μs $n_n \approx 10^{22}$ neutron/cm^3
β-decay lifetime: few hours to some months

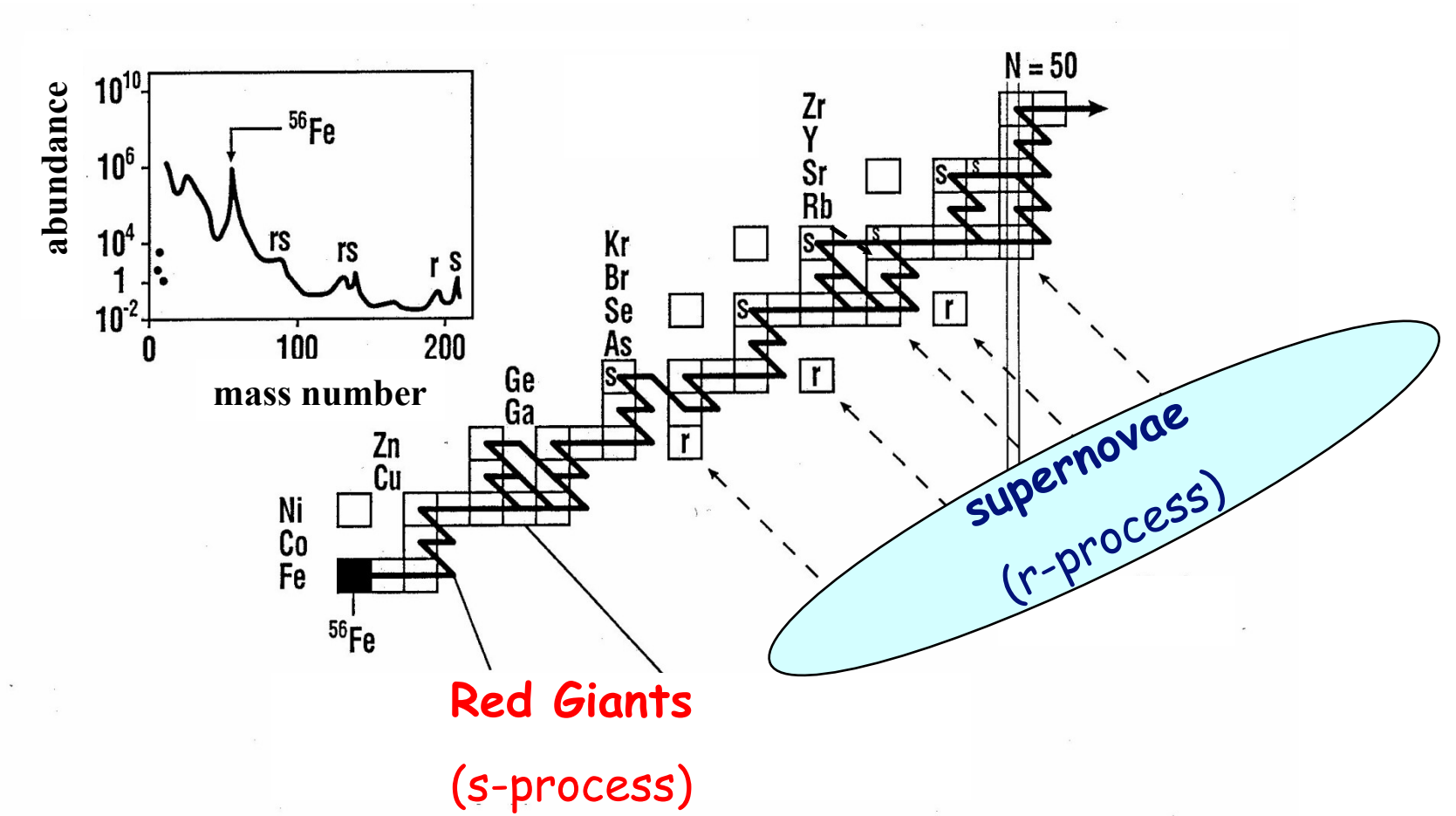
The canonical s-process



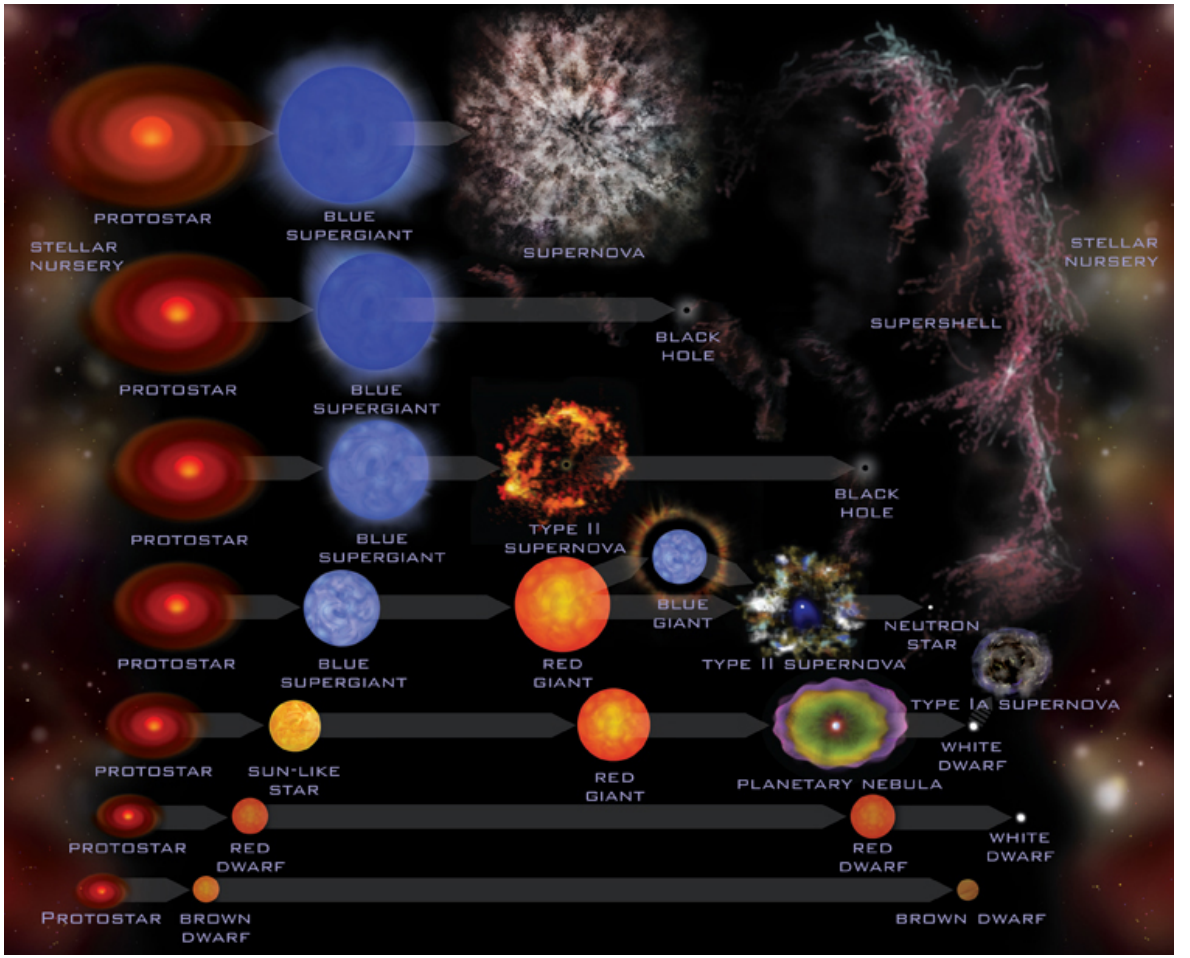
Solar system elemental abundances



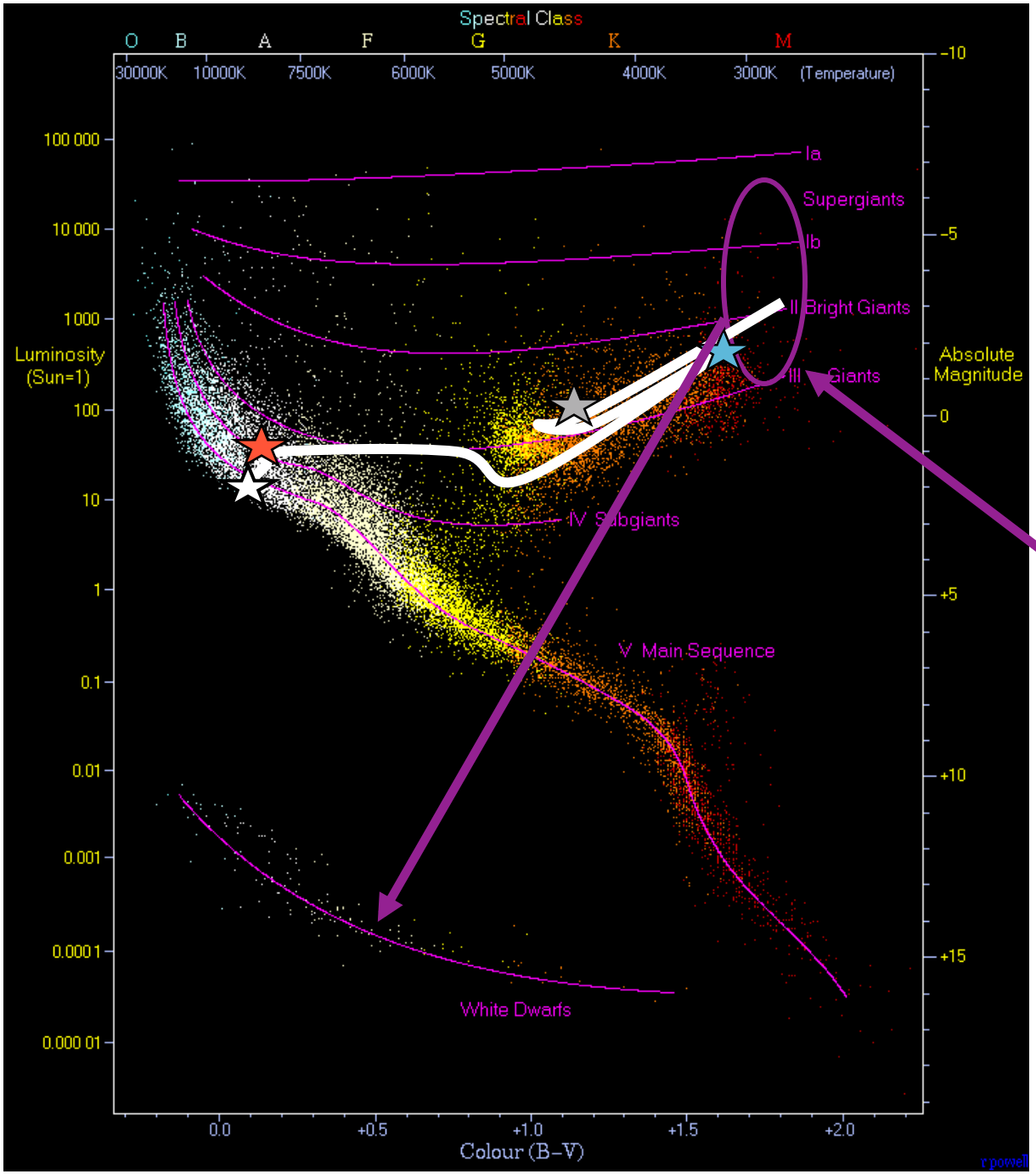
The r-process



Stellar evolution



Theoretical evolutionary track of a star of 2 $M_\odot$



Core H exhaustion

Core He burning starts

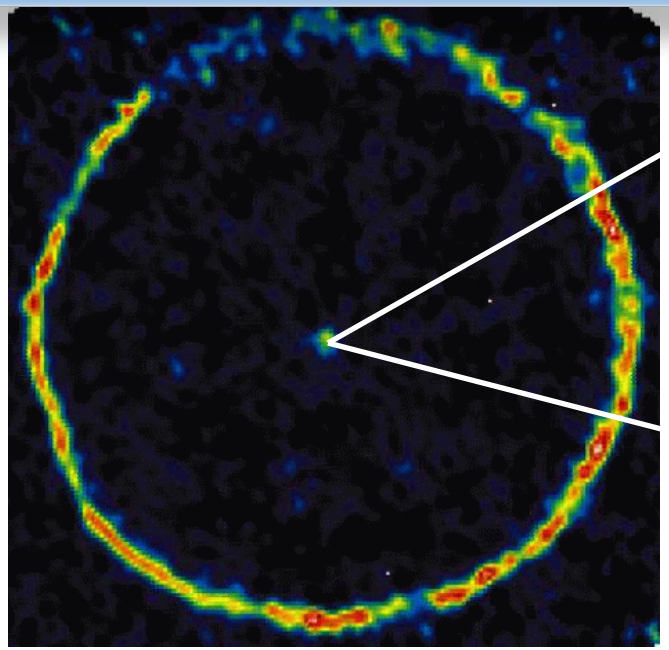
Core He exhaustion

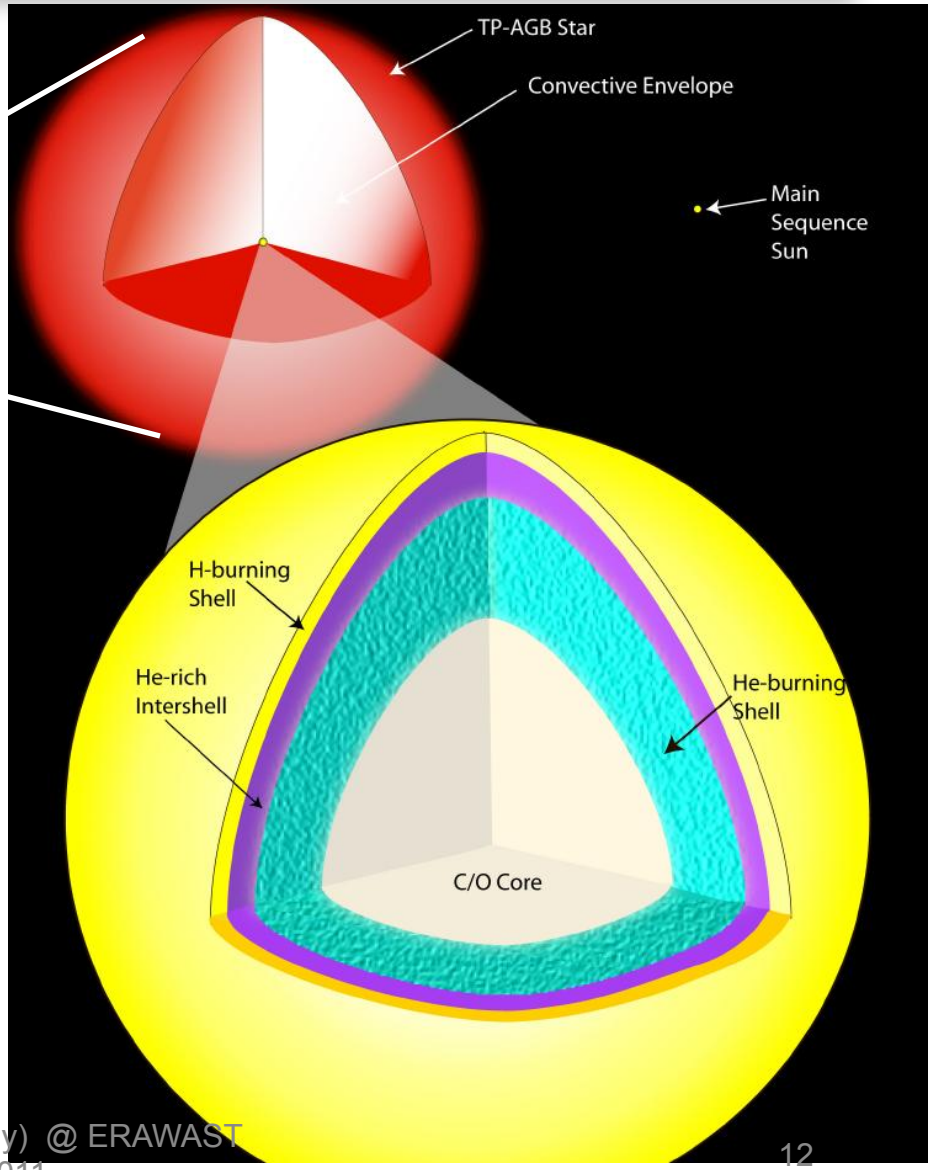
AGB stars

All stars with masses < 9 $M_\odot$ go through the AGB.

AGB stars: cool and luminous red giant stars

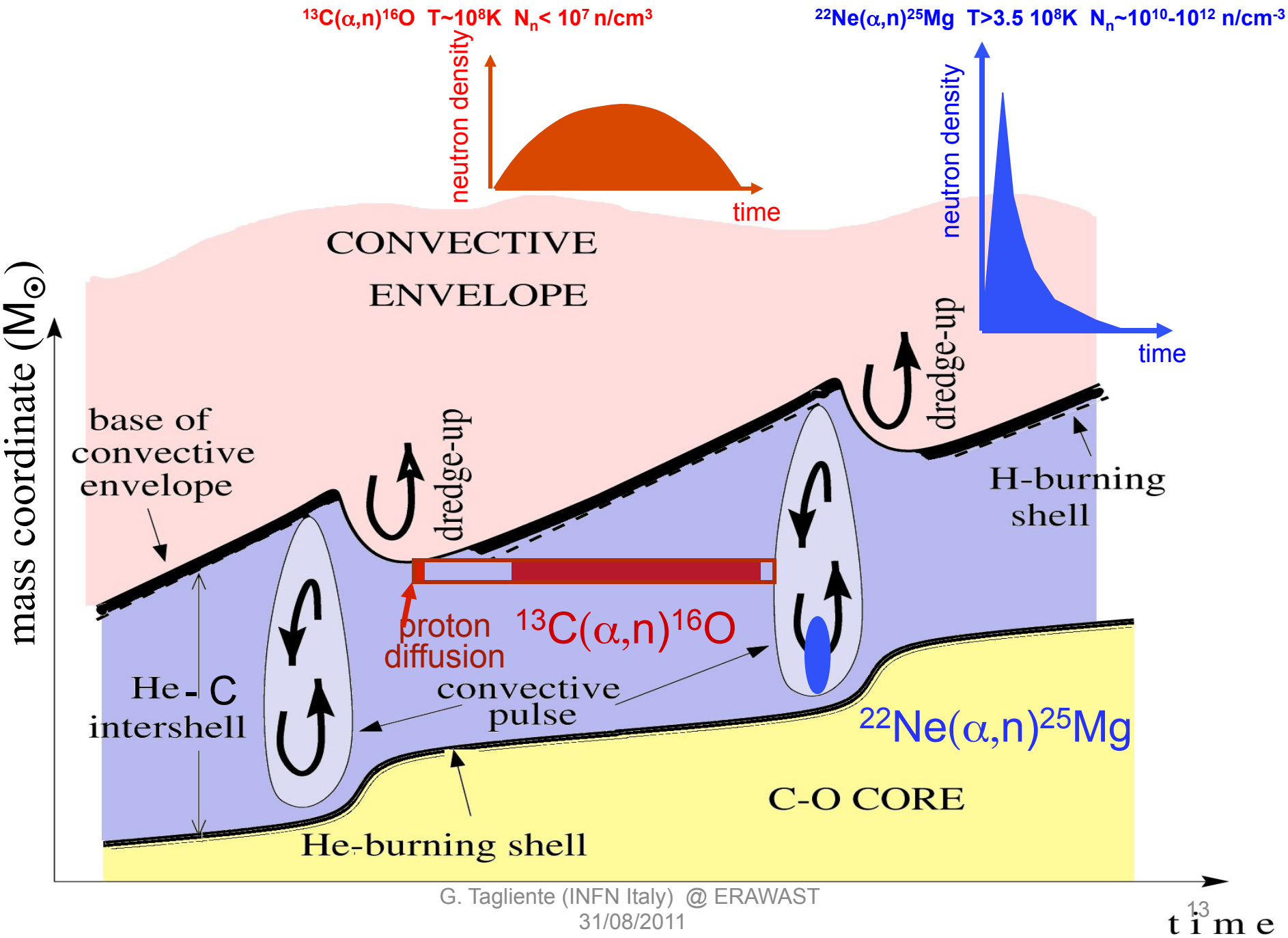
False-color picture of CO molecules tracing material around the AGB star TT-Cygni

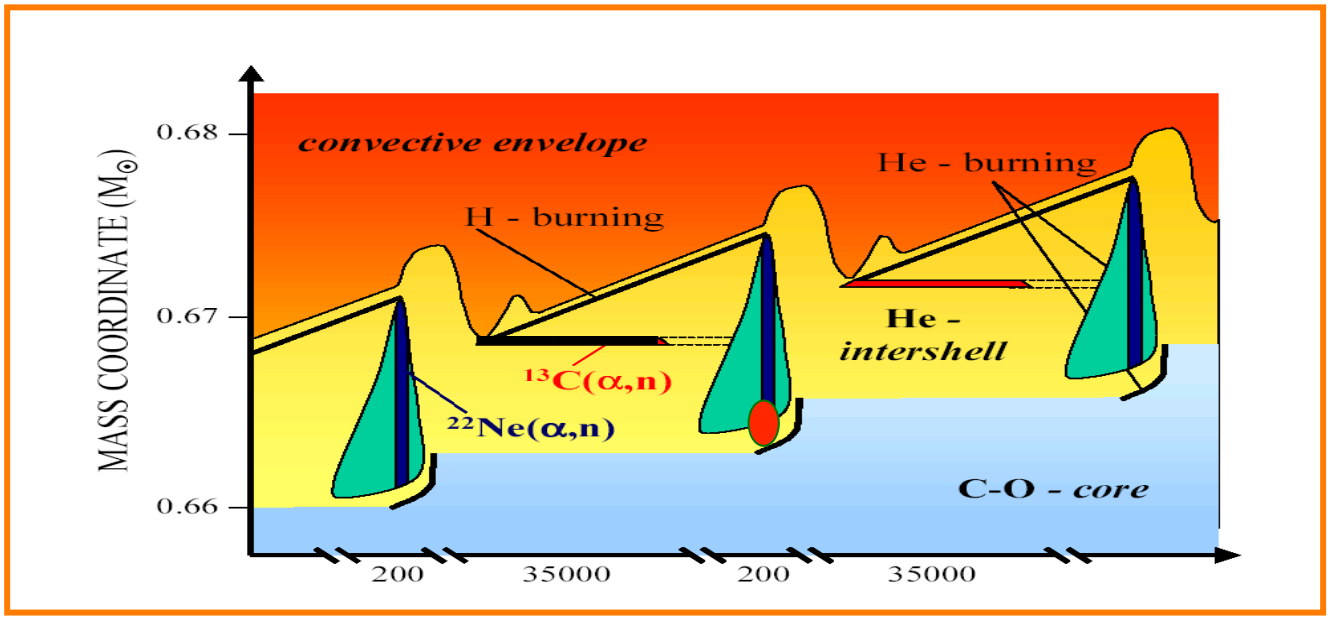




Strong mass loss driven by stellar pulsations and radiation pressure on dust.

Newly synthetised elements and dust grains are shed into the surrounding interstellar medium


$^{13}C(\alpha,n)^{16}O$ T~10^8K N_n< 10^7 n/cm^3
$^{22}Ne(\alpha,n)^{25}Mg$ T>3.5 10^8K N_n~10^{10}-10^{12} n/cm^{-3}
neutron density
time
neutron density
time
CONVECTIVE
ENVELOPE
mass coordinate ($M_\odot$)
base of
convective
envelope
dredge-up
dredge-up
H-burning
shell
proton
diffusion
$^{13}C(\alpha,n)^{16}O$
He - C
intershell
convective
pulse
$^{22}Ne(\alpha,n)^{25}Mg$
C-O CORE
He-burning shell
time



^{92}Mo 14.84	^{93}Mo 4.00 ka	^{94}Mo 9.25	^{95}Mo 15.92	^{96}Mo 16.68	^{97}Mo 9.55	^{98}Mo 24.13	^{99}Mo 2.75 d	^{100}Mo 9.63
^{91}Nb 680.04 a	^{92}Nb 34.70 Ma	^{93}Nb 100	^{94}Nb 20.30 ka	^{95}Nb 34.99 d	^{96}Nb 23.35 h	^{97}Nb 1.20 h	^{98}Nb 2.86 s	^{99}Nb 15.00 s
^{90}Zr 51.45	^{91}Zr 11.22	^{92}Zr 17.15	^{93}Zr 1.53 Ma	^{94}Zr 17.38	^{95}Zr 64.03 d	^{96}Zr 2.8	^{97}Zr 16.74 h	^{98}Zr 30.70 s
^{89}Y 100	^{90}Y 2.67 d	^{91}Y 58.51 d	^{92}Y 3.54 h	^{93}Y 10.18 h	^{94}Y 18.70 m	^{95}Y 10.30 m	^{96}Y 5.34 s	^{97}Y 3.75 s

The Thermal Pulse Stellar model: the Zr case

There is some inconsistency using the TP stellar model to calculate the N_s abundances with actual values of the Zr cross sections.

Nucleus	$N_\odot$ Normalized to N(Si) $=10^6$ atoms	$N_s / N_\odot$
^{90}Zr	5.546	0.789
^{91}Zr	1.21	1.066
^{92}Zr	1.848	1.052
^{94}Zr	1.873	1.217
^{96}Zr	0.302	0.842

The uncertainty on the $N_\odot$ is 10%

The uncertainty on Zr cross sections ranges from 5% to 20% (depending on the isotopes).

There are discrepancies of 50% on the results of some measurements

New measurements with high accuracy are needed !

The n_TOF facility at CERN

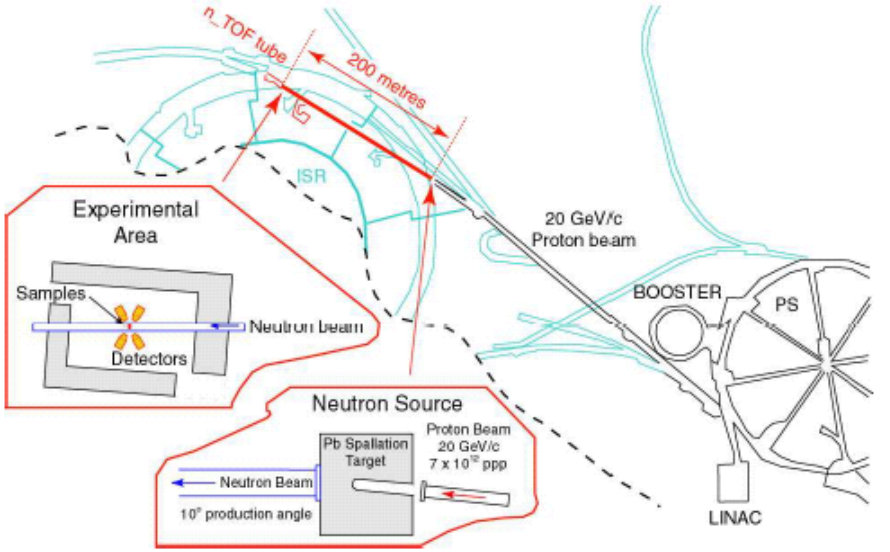
somewhere around here

The n_TOF facility at CERN

- Spallation of high-energy proton beam on a lead target (~360 neutrons/proton)

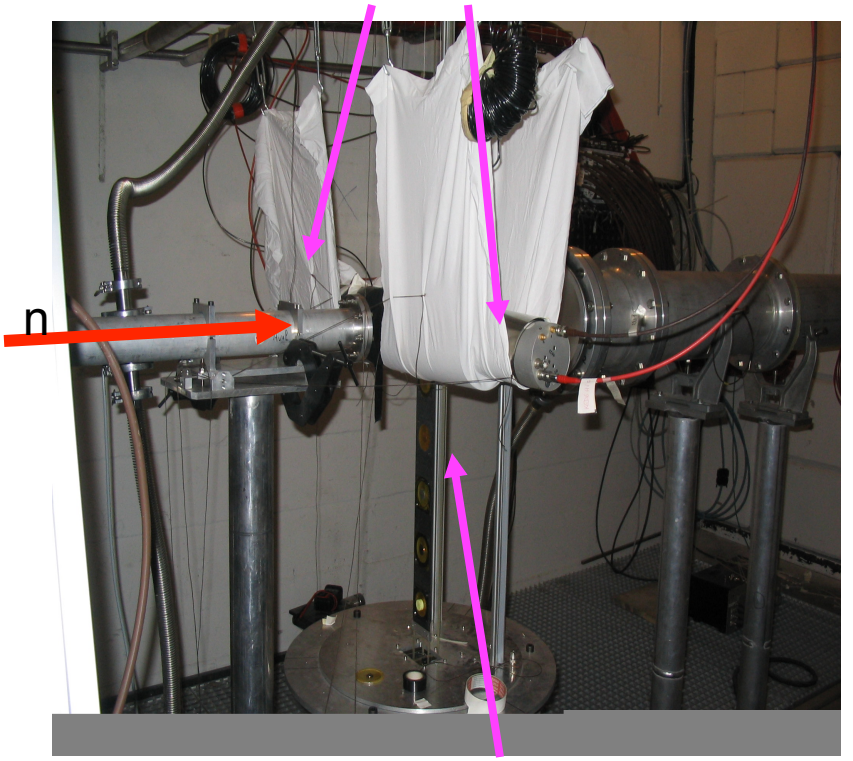
- 7×10^{12} protons/bunch @ 20 GeV/c from the PS accelerator (6 ns time resolution)

- 0.8 Hz maximum repetition rate



Very high instantaneous neutron flux
fundamental for studying small samples and radioactive isotopes

n_TOF phase I: Zr measurements



Sample changer

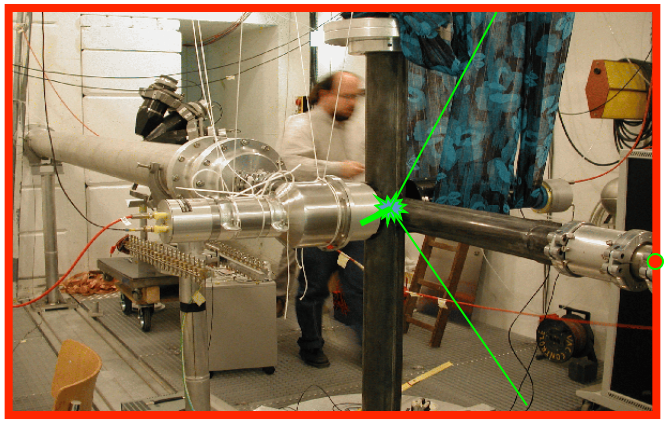
Specifically designed **low neutron sensitivity** C_6D_6 liquid scintillators

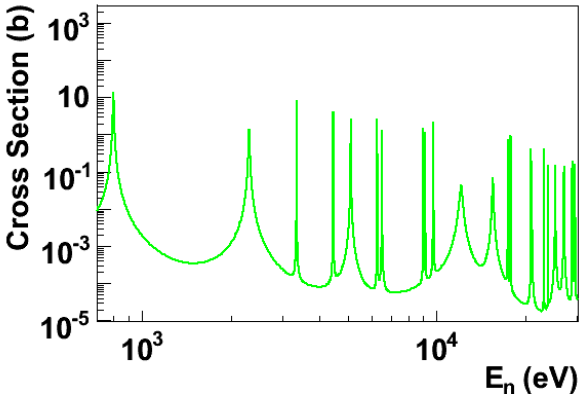
Sample	Measurement
$^{90,91,92,94,96}Zr$	June-Aug. 2003
^{93}Zr	Oct. 2004

(n,γ) Total energy detection

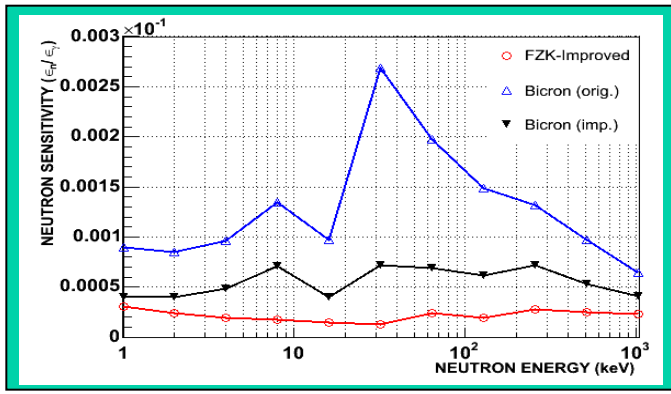
Improvements in the Experimental Setup & Data Analysis

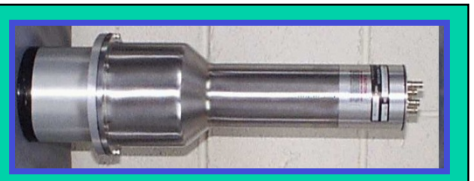
- Lowest neutron sensitivity ➡ No neutron background corrections !

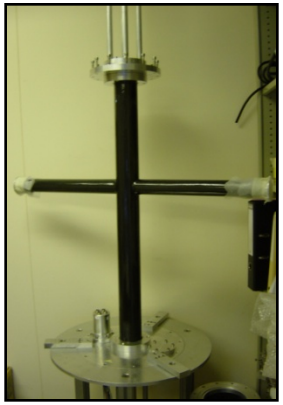




(n,γ)



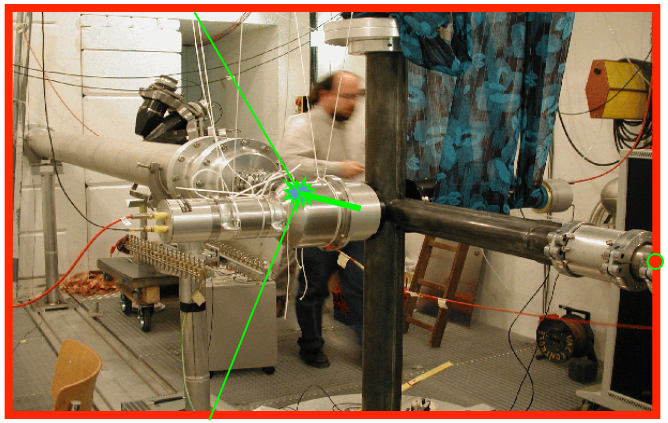


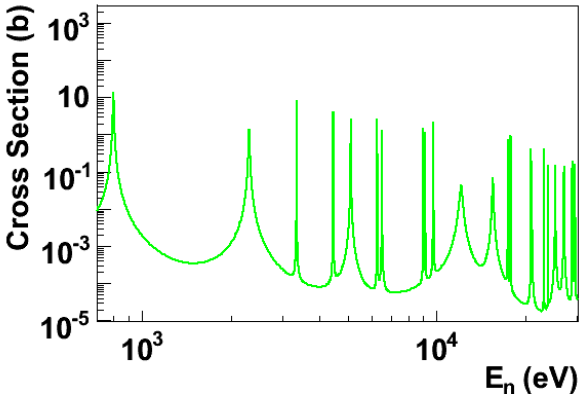


(n,γ) Total energy detection

Improvements in the ExperimentalSetup & Data Analysis

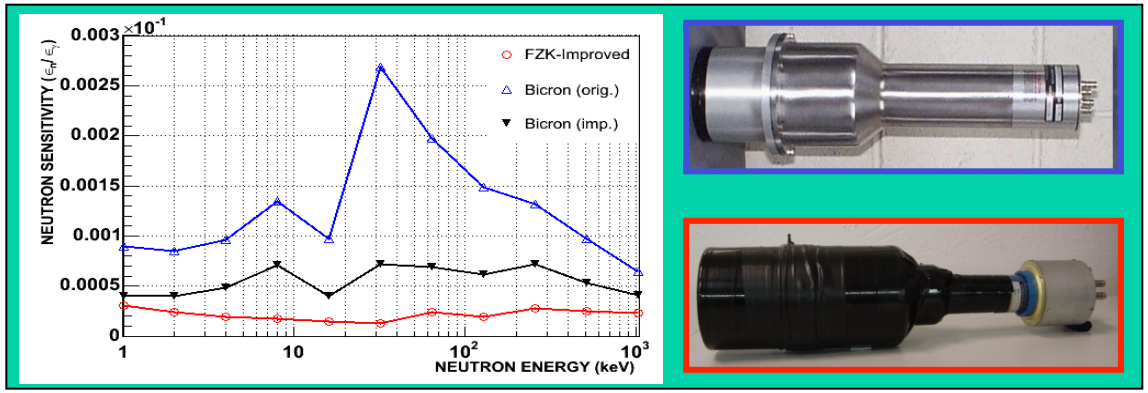
- Lowest neutron sensitivity ➡ No neutron background corrections !

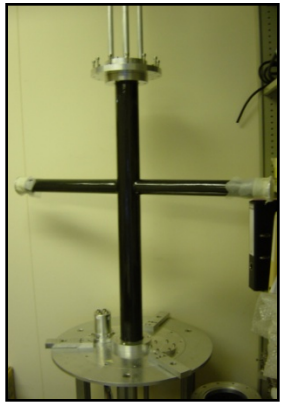




(n,n)

(n,γ)





Zr isotope samples

Sample	Isotopic content (%)					
	^{90}Zr	^{91}Zr	^{92}Zr	^{93}Zr	^{94}Zr	^{96}Zr
^{90}Zr	97.7	0.87	0.6	-	0.67	0.16
^{91}Zr	5.43	89.9	2.68	-	1.75	0.24
^{92}Zr	4.65	1.62	91.4	-	2.03	0.3
*^{93}Zr**	*1.5*	*19.0*	*20.0*	*20.0*	*20.0*	*19.0*
^{94}Zr	4.05	1.18	1.93	-	91.8	1.04
^{96}Zr	19.41	5.21	8.2	-	8.68	58.5

Admixture: Hf, Na, Mg, Al ...

* Radio isotope ($T_{1/2}$ =1.5x10^6 year)

Zr isotope samples

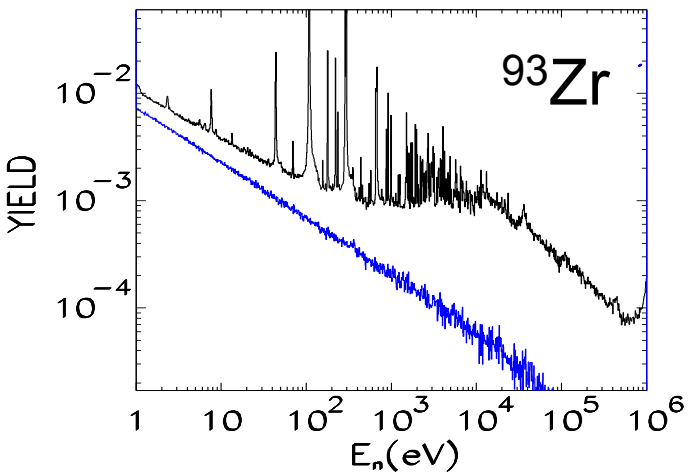
	^{90}Zr	^{91}Zr	^{92}Zr	^{93}Zr	^{94}Zr	^{96}Zr	^{197}Au	Lead
Mass (g)	2.717	1.404	1.349	4.88	2.015	3.398	1.871	3.895
Thickness (cm)	0,127	0,065	0,062	0,37	0,091	0,151	0.025	0.09
Chemical form	ZrO_2	ZrO_2	ZrO_2	ZrO_2	ZrO_2	ZrO_2	Metal	Metal
Enrichment (%)	97.7	89.9	91.4	*20.0*	91.8	58.5	100	Nat.

Samples 2.2 cm in diameter, 1 mm thick
Stable Zr isotopes encapsulated in 0.2 mm Al can
^{93}Zr isotope encapsulated in 0.2 mm Al + 0.2 mm Ti

Chemical form: ZrO_2

^{93}Zr isotope activity 92.5 MBq

Data Analysis - ^{93}Zr yield

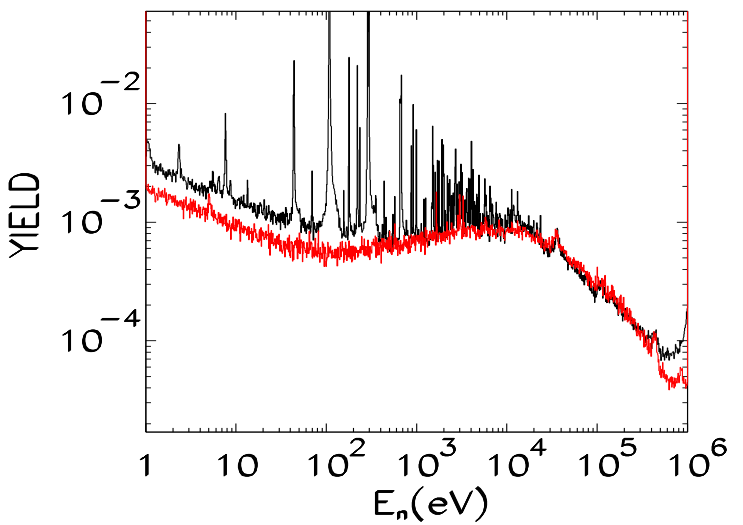


raw Yield

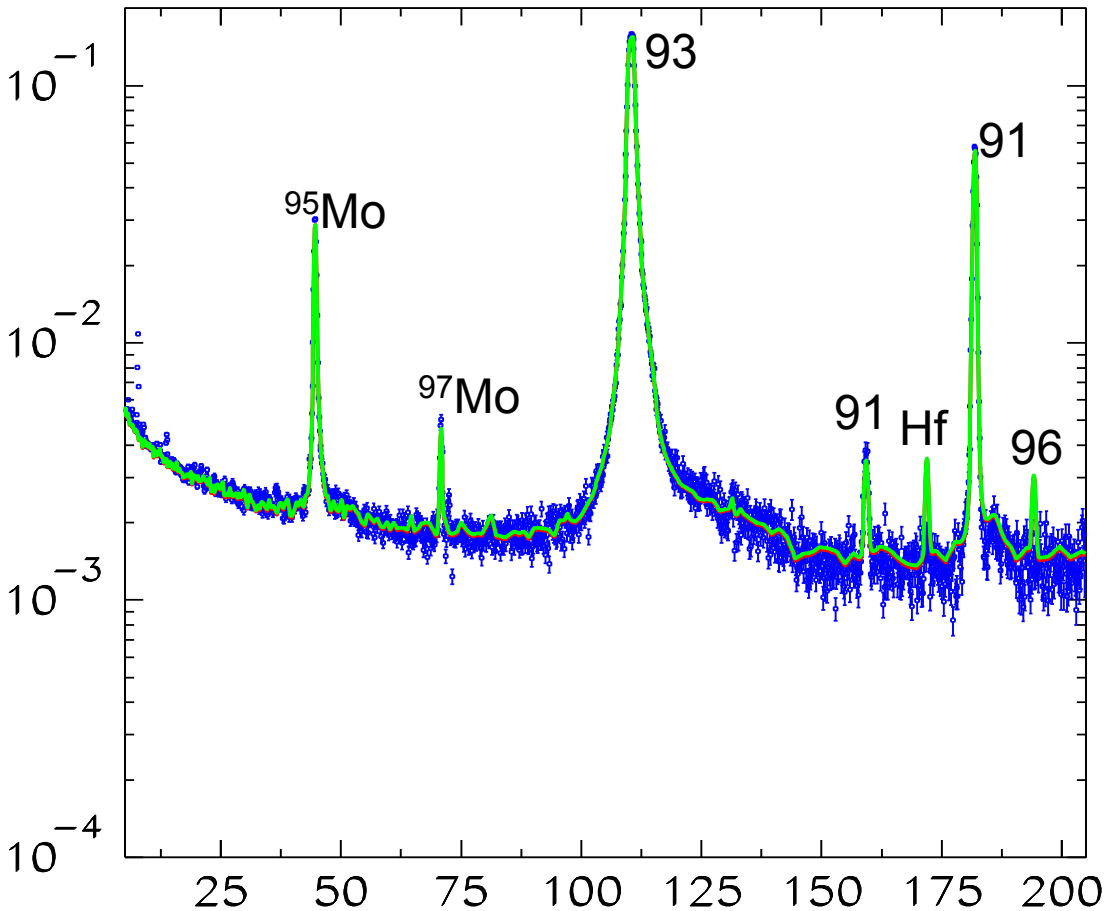
Natural radioactivity of the sample

Yield – nat. radioactivity of the sample

overall background



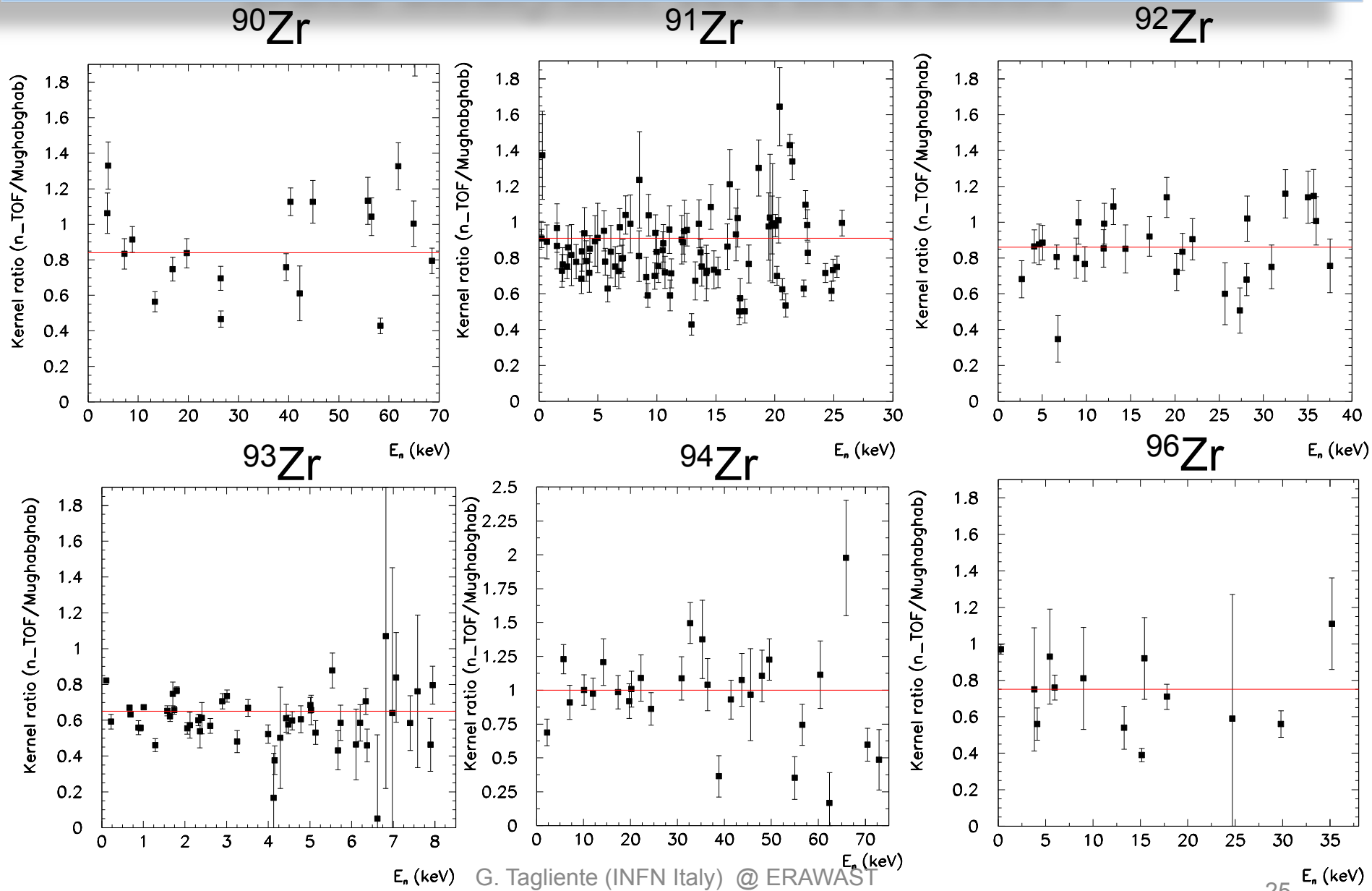
Resonance Analysis ^{93}Zr



$$A_\gamma = 2n\pi^2 K\lambda\!\!\!^{-}$$

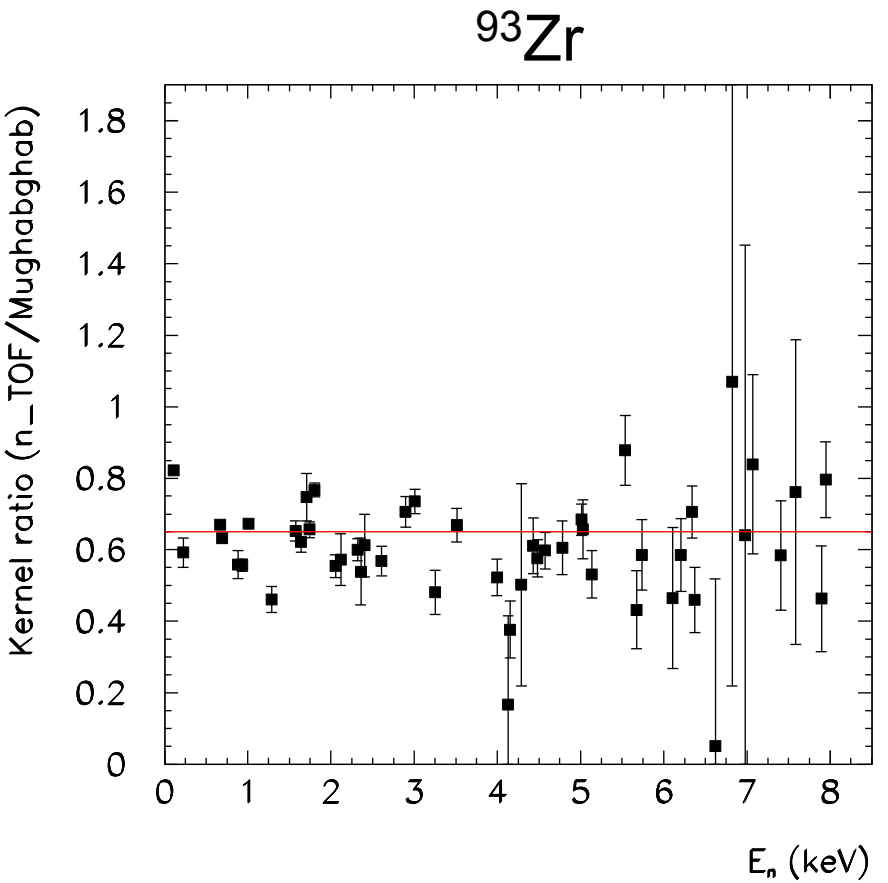
$$K = g\frac{\Gamma_n\Gamma_\gamma}{\Gamma}$$

Data analysis: Kernel ratios



- Weighted average ratio

Data analysis: Kernel ratios



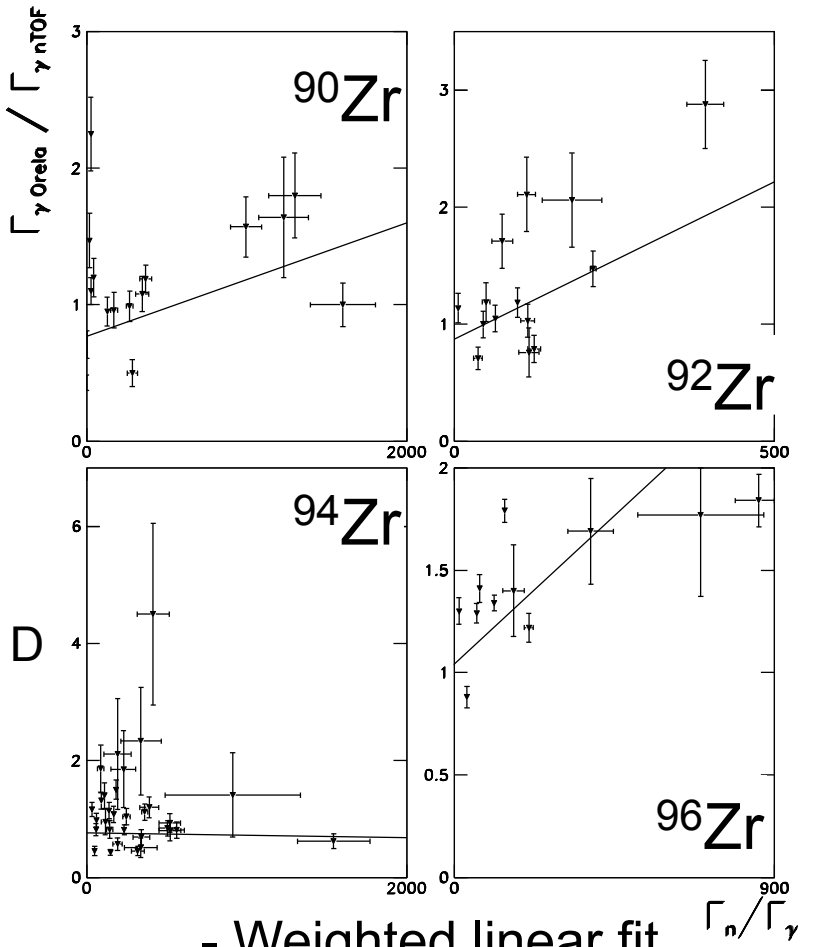
Prelimirary

The ^{93}Zr new kernels are 37% lower than the previous measurements

Data analysis

Sample	Weighted $\langle K_{n\_TOF}/K_{ORELA} \rangle$
^{90}Zr	0.84±0.01
^{91}Zr	0.91±0.01
^{92}Zr	0.86±0.02
^{93}Zr	0.66±0.01
^{94}Zr	1.03±0.01
^{96}Zr	0.73±0.02

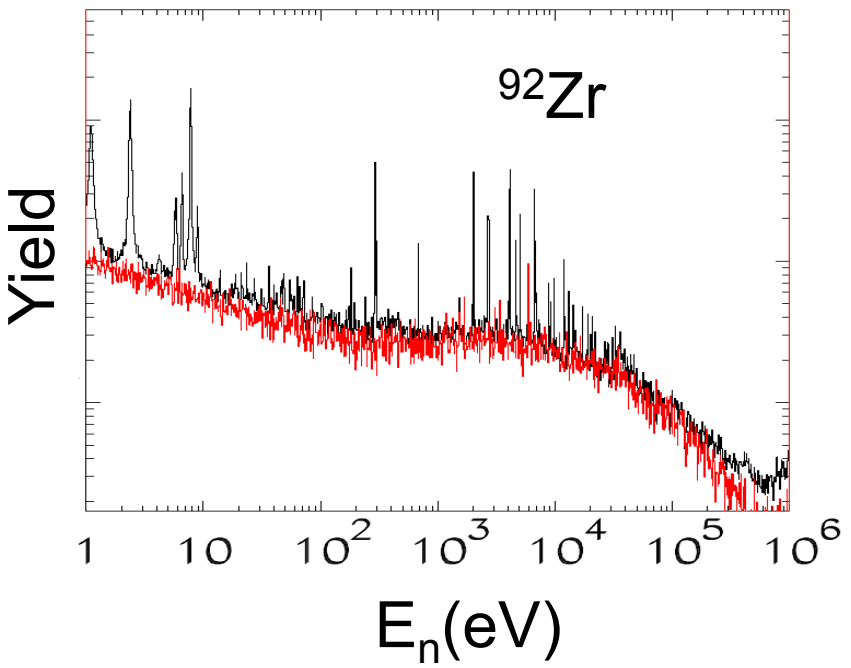
ORELA detectors C_6F_6: F has an higher neutron capture cross section than D

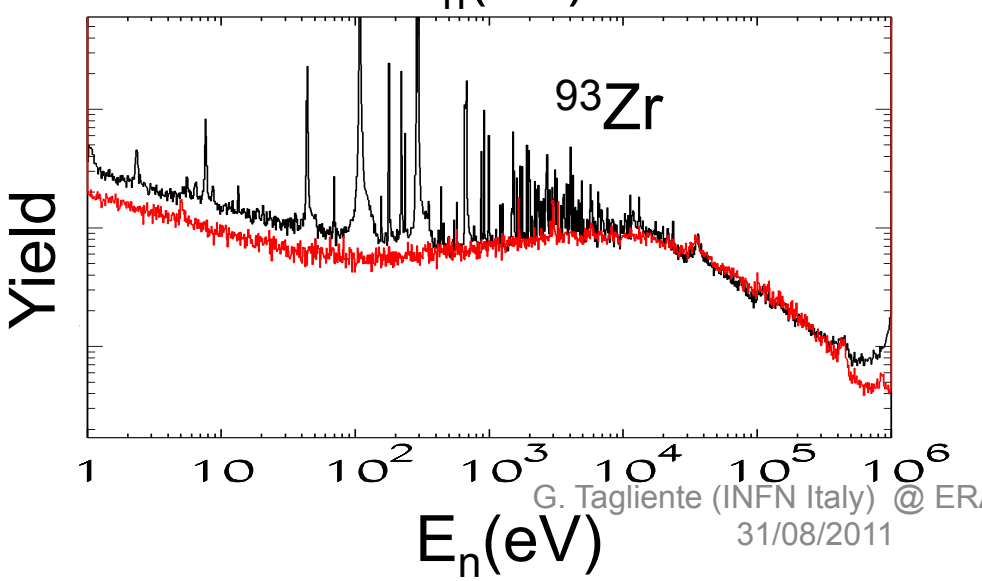


- Weighted linear fit

MACS: Experimental energy ranges

MaxwellianAveraged Cross Sections(MACS)

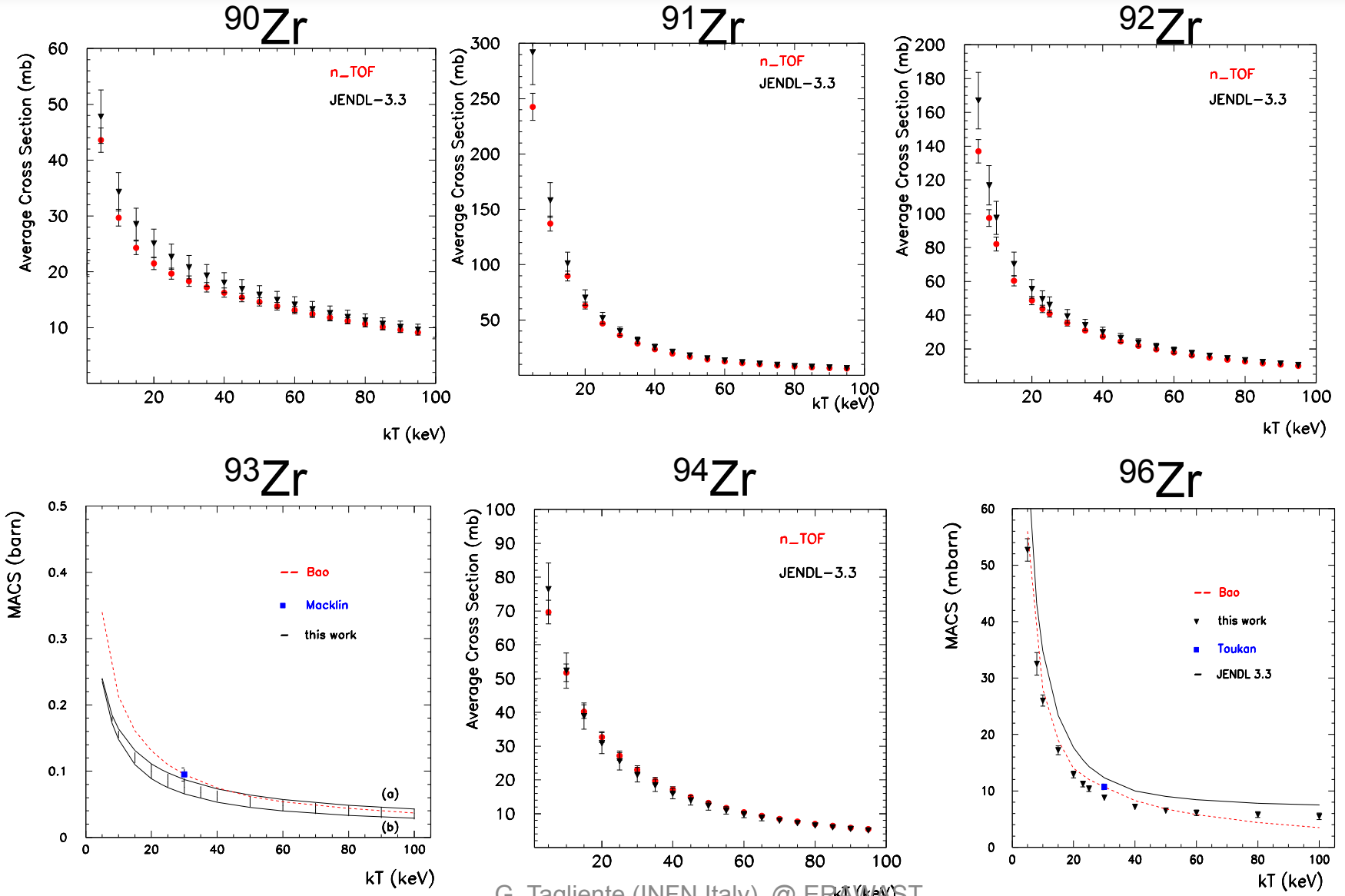




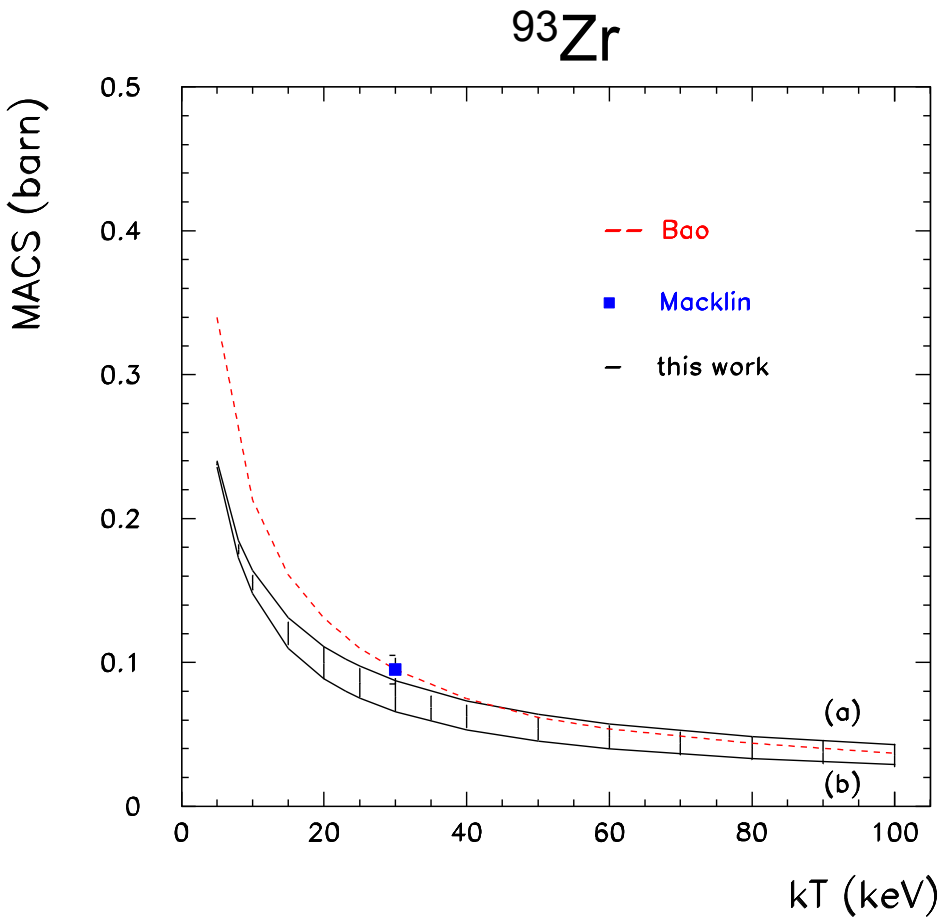
	Energy range (keV) n_TOF
^{90}Zr	0.01 - 66
^{91}Zr	0.01 - 26
^{92}Zr	0.01 – 40
^{93}Zr	0.01 – 7
^{94}Zr	0.01 – 74
^{96}Zr	0.01 – 42

The n_TOF data have to be complemented with a data library Jendl 3.3 and ENDF IV

MACS: results



MACS: results

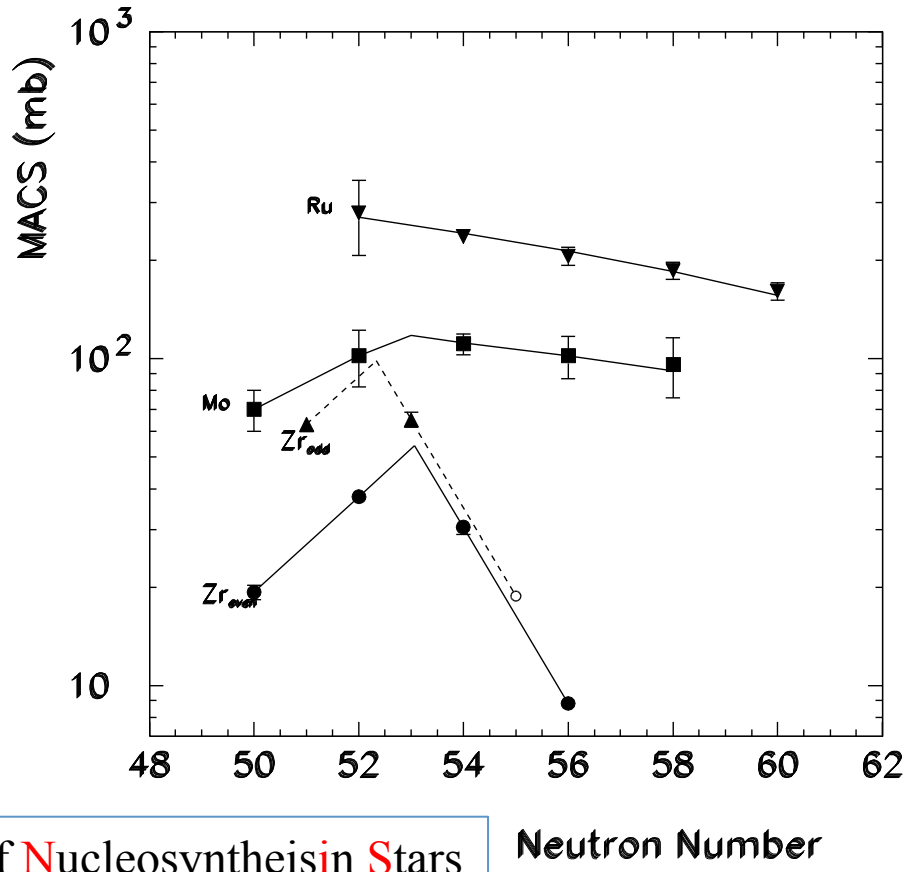


Prelimirary

MACS:@ 30 keV

MACS in mbarn

Isotope	KADoNiS	n_TOF	MOST
^{90}Zr	21 ± 2	19.3 ± 0.9	
^{91}Zr	60 ± 8	63 ± 4	
^{92}Zr	34 ± 6	38 ± 3	
^{93}Zr	95 ± 10	**65.1 ± 3**	67.8
^{94}Zr	26 ± 1	30.5 ± 2	
^{95}Zr	79	**18.9 ± 0.9**	24.2
^{96}Zr	10.7 ± 0.5	8.9 ± 0.5	



KADoNIS: Karlsruhe Astrophysical Database of Nucleosyntheisin Stars

MOST: Reaction code to calculate nuclear reaction rates for astrophysics applications on the basis of Hauser-Feshbach.(**S. Goriely** Institut d'Astronomie et d'Astrophysique, Université Libre de Bruxelles)

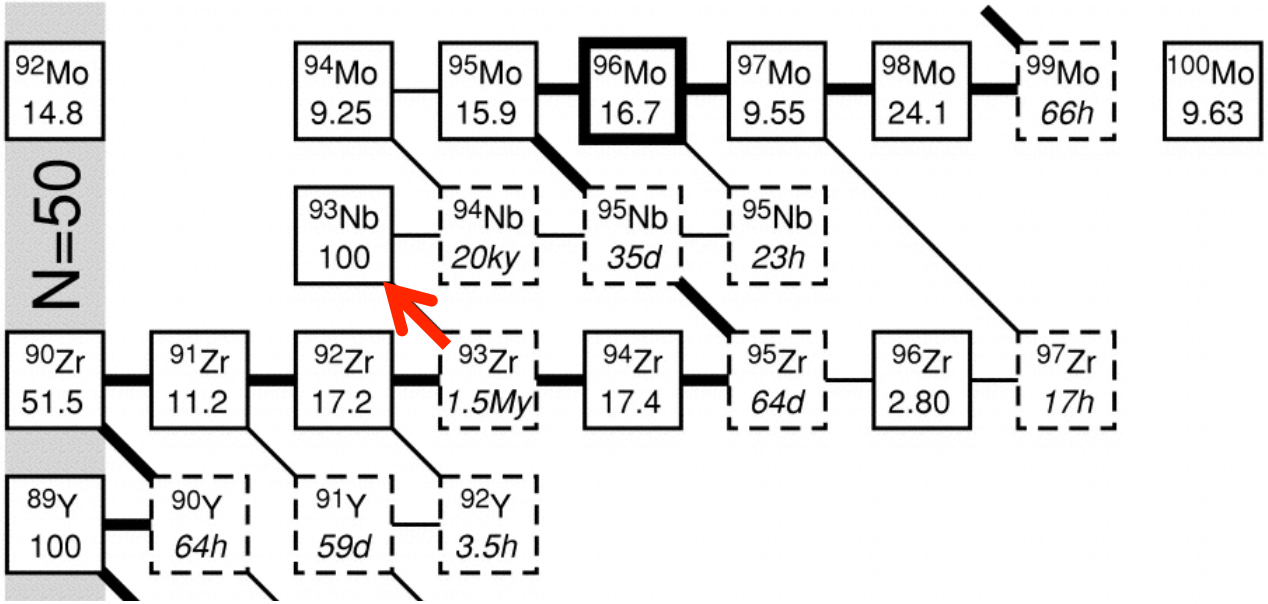
Astrophysical implication: Abundances

Nucleus	N_Θ Normalized to N (Si)=10^6 atoms	N_s/N_Θ % Old MACS	N_s/N_Θ % n_TOF MACS
^{90}Zr	5.546	0.789	0.844
^{91}Zr	1.21	1.066	1.024
^{92}Zr	1.848	1.052	0.981
^{94}Zr	1.873	1.217	1.152
^{96}Zr	0.302	0.842	0.321

Solar abundances, $N_\odot$, from Lodders 2009, accuracy 10%

The s-abundances, N_s, are calculated using the TP stellar model for low mass AGB star (1.5 - 3 $M_\odot$), accuracy 8%.
Old MACS are from the KADoNiS data base 2008. Since 2009 the databases has been update at the new n_TOF data, as the new data are relased.

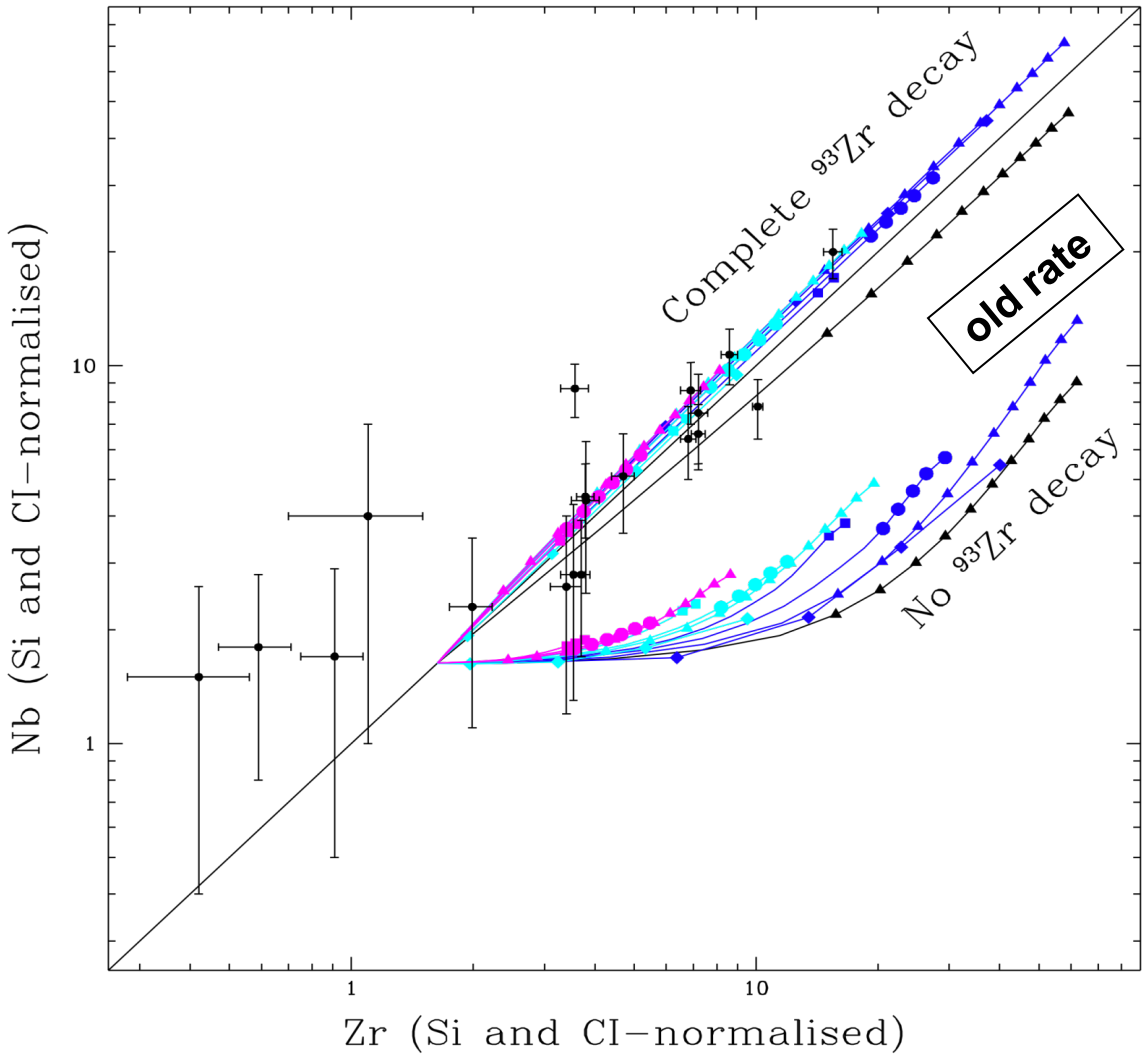
Astrophysical implication: Zr/Nb



A lower ^{93}Zr(n,γ) value means that more ^{93}Zr is produced. After radiogenic decay of ^{93}Zr more Nb will result.

The final result is ~50% more Nb!

Elemental Nb and Zr abundances in SiC



Mass, Metallicity

◇ 1.8, 0.01 ~ 0.7 $Z_\odot$

△ 3, 0.01 ~ 0.7 $Z_\odot$

○ 3, 0.02 ~ 1.5 $Z_\odot$

□ 3, 0.03 ~ 2 $Z_\odot$

$M_{mix}(M_\odot)$

0.002

0.0005

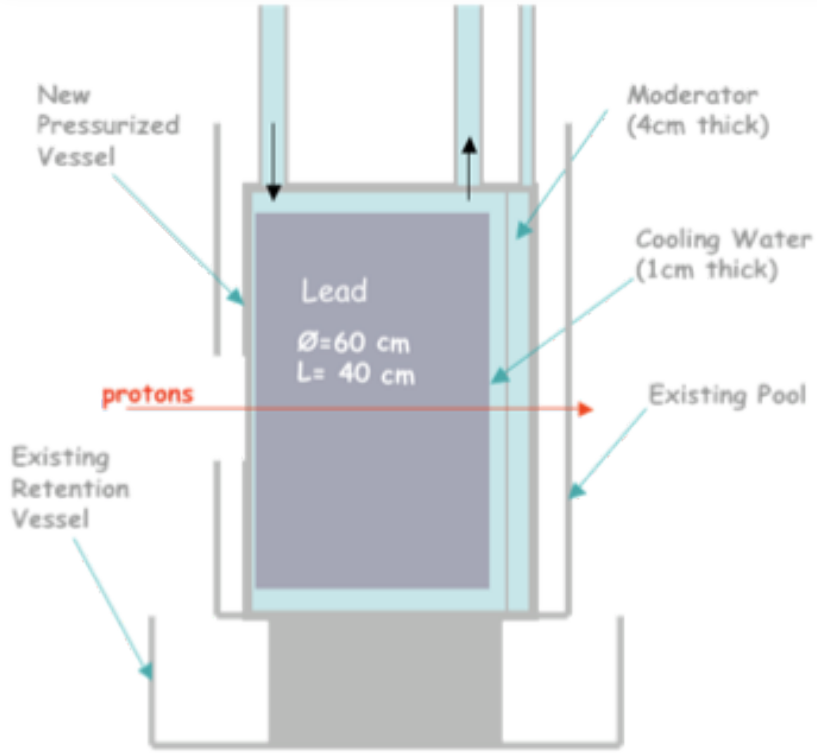
0.0002

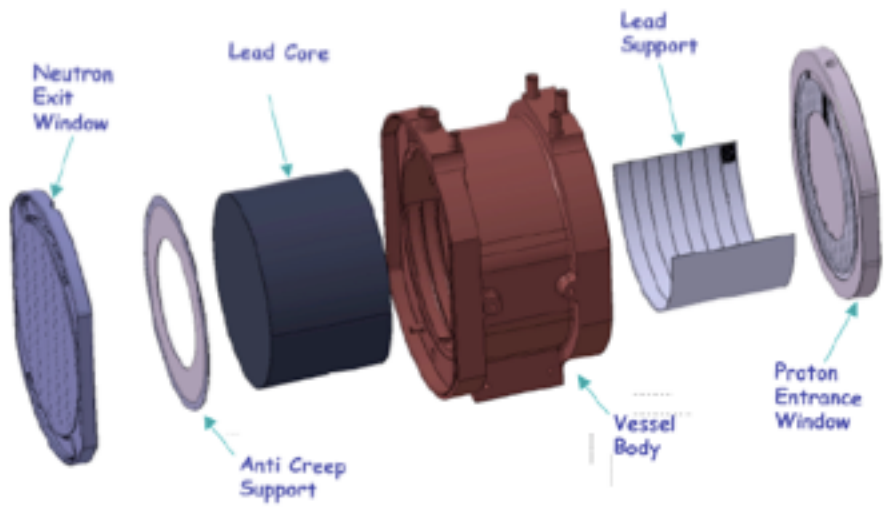
With the new ^{93}Zr(n,γ) cross section the problem is solved.

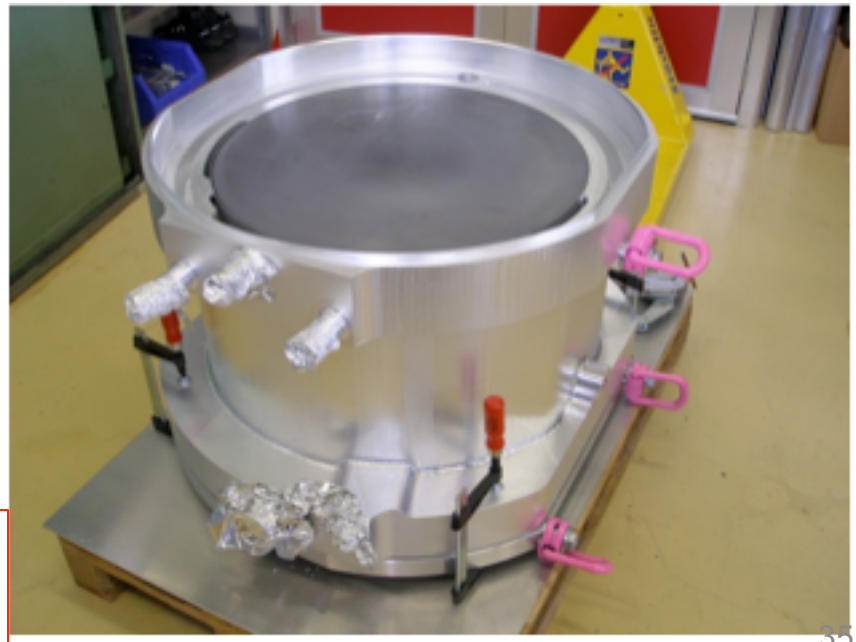
BUT!!!

The data, specially for the ^{93}Zr, have to be confirmed with new data in a wider neutron energy range

n_TOF new spallation target





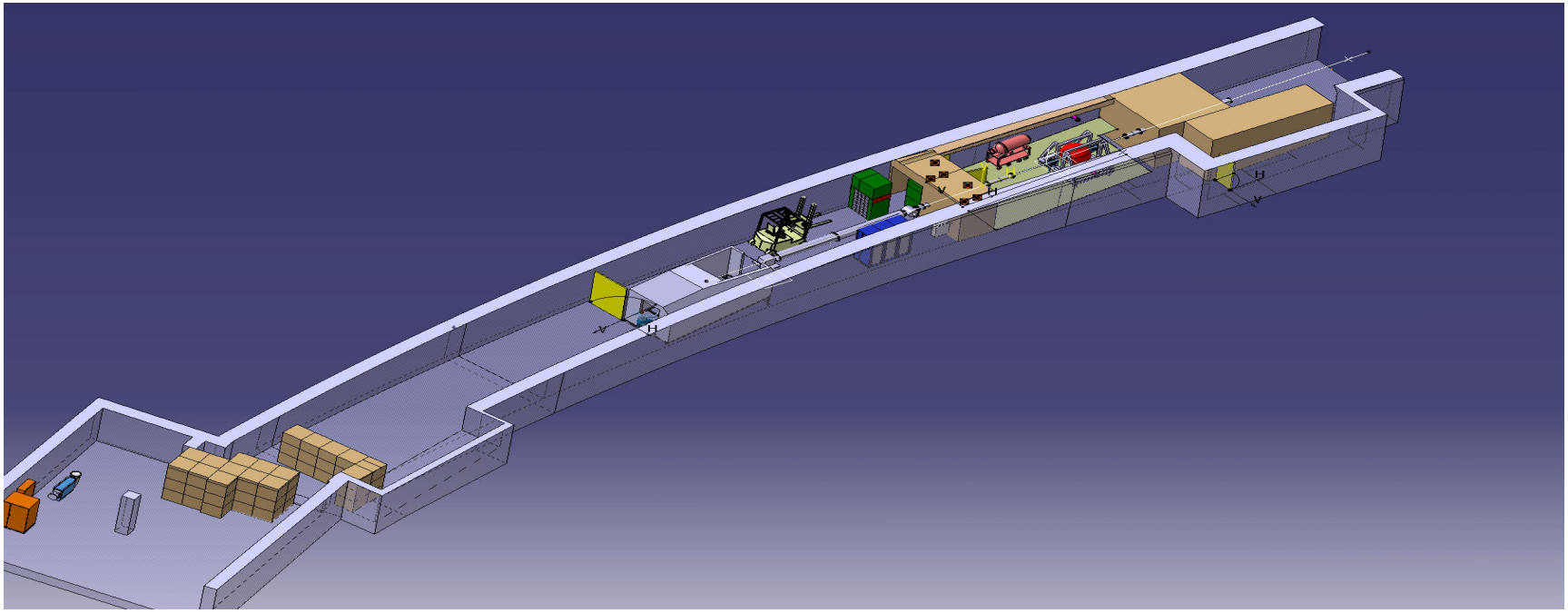


✓Optimized for a better cooling
✓two different circuits for cooling and moderation

The borated water as moderator reduces the background of a factor 10!!

See Claudia Lederer talk 01/09

Work Sector of Type A



The main problem in the ^{93}Zr measurement was the radioprotection issue.

Since 2010 the n_TOF experimental area was transformed in work sector type A.

It will allow us to measure the ^{93}Zr without double canning (Ti + Al).

Conclusion

- New neutron capture measurements on $^{90,91,92,93,94,96}Zr$ were done at n_TOF facility
- MACS calculated from the new data for most of the Zr isotopes are lower than the previous MACS
- MACS uncertainty improved by a factor 2
- The new MACS work much better when used in the TP stellar model to calculate the s-process abundances, proving the validity of the model
- We will propose a new neutron capture measurement for the ^{93}Zr at n_TOF facility. Thanks to the upgrade of the facility now it should be possible to extend the neutron energy range measured at 100 keV.