

# “Synteza najcięższych jąder atomowych”

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*Nathan Brewer (UTK→ORNL)*

**wyniki z lat 2011-2013 otrzymane we współpracach**

## **Rosja i USA**

*JINR Dubna - ORNL Oak Ridge - LLNL Livermore*

*UT Knoxville - RIAR Dmitrovgrad - Vanderbilt Nashville  
i Uniwersytet Warszawski (!)*

## **Niemcy i reszta świata**

*Darmstadt-Mainz-Lund-Oak Ridge-Berkeley  
i inne laboratoria (Warszawa)*



**Warszawa, 16 stycznia 2014**



*Rose Boll and Shelley Van Cleve  
przygotowujące próbke  $^{249}\text{Bk}$  (~40 Ci)*



*Vladimir Utyonkov  
analizujący dane  $^{48}\text{Ca}+^{239}\text{Pu}$*

**OAK RIDGE NATIONAL LABORATORY**  
MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

# Long term research goals

- **New Heaviest Elements and Nuclei**
  - *how many protons and neutrons a nucleus can hold ?*
  - *unified description of nuclear properties across varying proton and neutron numbers*
  - *new energy gaps, magic numbers and Island of Stability*
  - *or rather enhanced stability without shell gaps and magic numbers*
  - *understanding fission process competing with other decay modes ( $\alpha$ , EC)*
  - *structure beyond ground-state properties of super heavy nuclei*
- **Understanding production mechanism of the heaviest nuclei**
  - *hot and cold fusion reactions with stable and radioactive nuclei*
  - *multi-nucleon transfer between very heavy nuclei*
- **Expansion of Periodic Table of Elements**
  - *relativistic effects in chemical properties of atoms*
  - *super heavy atoms in the Universe*

Oak Ridge High School, April 2010



International team discovers element 117

***“Element 117”*** ice cream  
Razzleberry Ice Cream Lab  
Oak Ridge



*Periodic Table of Elements 2013*

1																	18
1 H	2											13	14	15	16	17	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac <sup>+</sup>	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113	114 Fl	115	116 Lv	117	118

\* Lanthanides

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
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+ Actinides

90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
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- Metals
- Non-metals
- Not confirmed

2012:

~ 150 days search for  $Z=119$  in  $^{249}\text{Bk} + ^{50}\text{Ti}$  reaction, TASCA, GSI,  $\sigma < 70 \text{ fb}$

~ 34 days search for  $Z=120$  in  $^{248}\text{Cm} + ^{54}\text{Cr}$  reaction, SHIP, GSI,  $\sigma < 600 \text{ fb}$

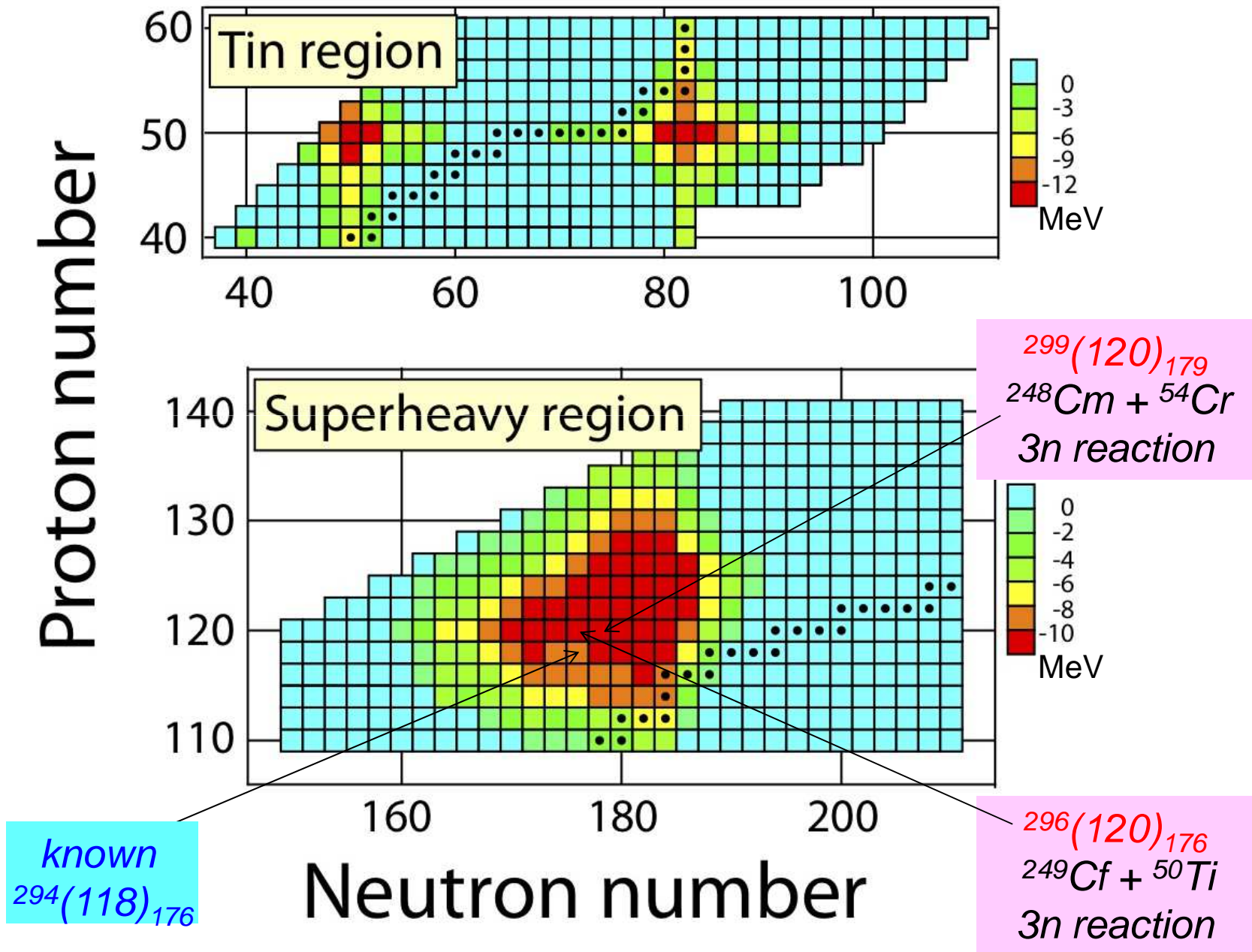
( ???!! )

*discovery of  $Z=121$  can start a group of next 18 elements...*



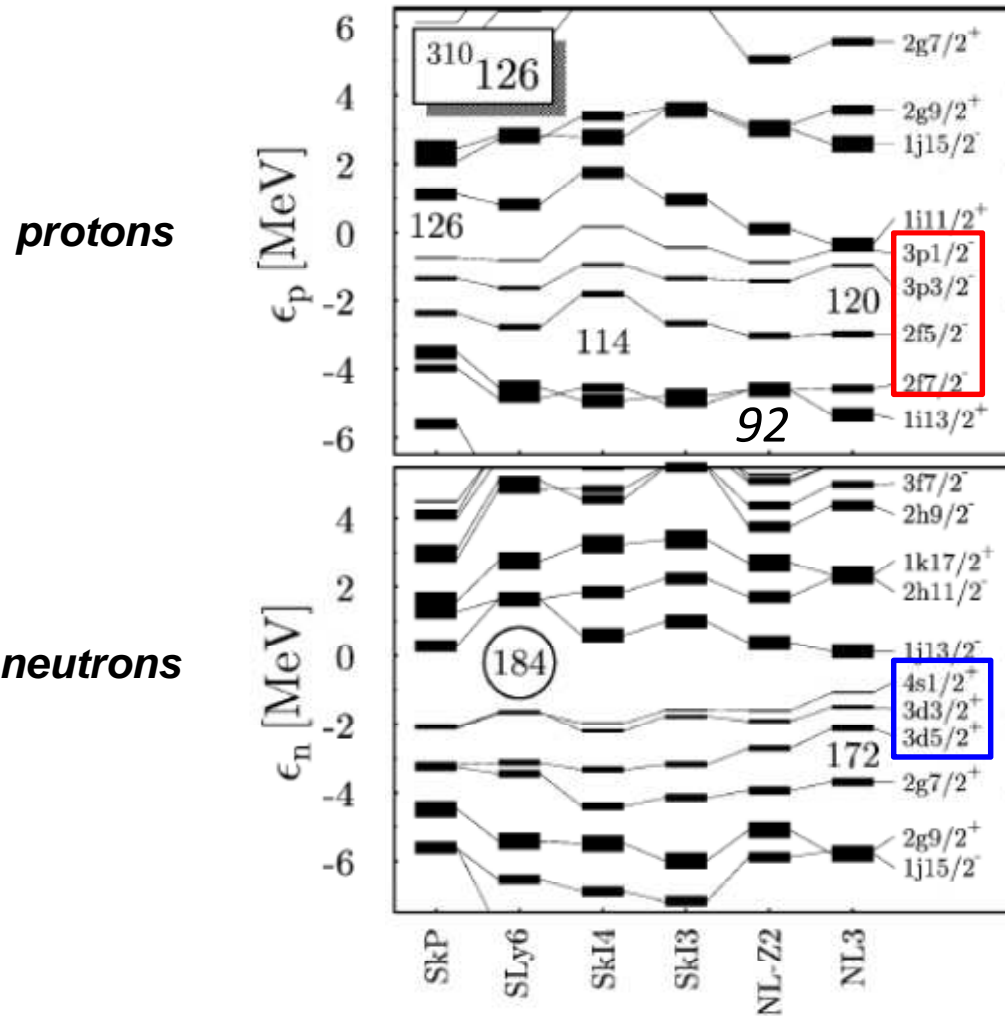


**M. Bender, W. Nazarewicz, P.-G. Reinhard, Phys. Lett. B 515, 42, 2001**  
**“Shell stabilization of super- and hyper-heavy nuclei without magic gaps”**



# Single-particle levels in the region of super heavy nuclei

M. Bender, W. Nazarewicz, P.- G. Reinhard, PL B 515, 42, 2001



# 2009-2010: Synthesis of a New Element with Atomic Number Z=117

Dubna-Oak Ridge-Las Vegas-Nashville-Livermore-Dmitrovgrad

The identification of a new element  $Z=117$  among the products of the  $^{249}\text{Bk}+^{48}\text{Ca}$  reaction was enabled by the close collaboration and unique capabilities of the US and Russia laboratories, neither country could achieve it alone. The 330 days half-life of radioactive  $^{249}\text{Bk}$  required a coordination of two years neutron irradiation and chemical separation at Oak Ridge followed by a target production at Dimitrovgrad and six months experiment with an intense  $^{48}\text{Ca}$  beam at Dubna.

High Flux Isotope Reactor  
ORNL, Oak Ridge



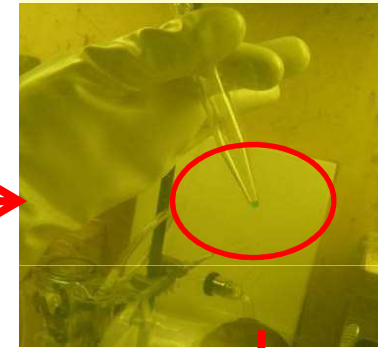
25-ton Q-ball transporting  
irradiated Am/Cm seed material



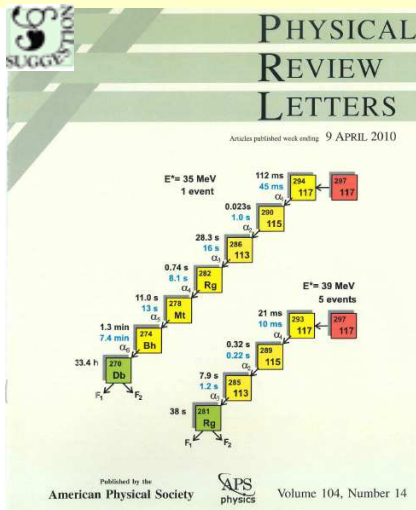
Chemical separation in hot cell  
REDC, ORNL, Oak Ridge



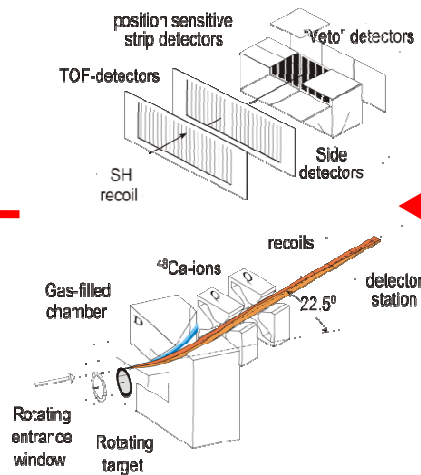
Pure 22 mg of  $^{249}\text{Bk}$



Decay chains of  $^{294}(117)$  and  $^{293}(117)$



Dubna Gas Filled Recoil Separator



U-400 cyclotron  
at JINR Dubna



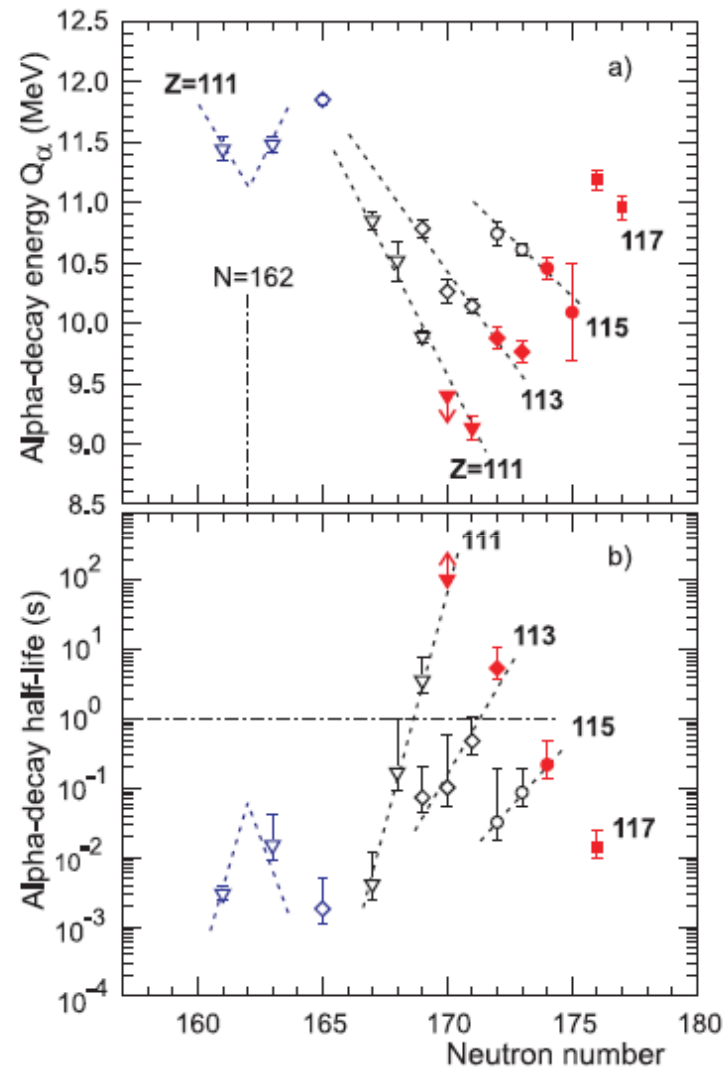
$^{249}\text{Bk}$  target wheel made  
at RIAR Dimitrovgrad





# Towards the Island of (enhanced) Stability

Oganessian et al., PRL 104, 142502, 2010 and PR C83, 054315, 2011

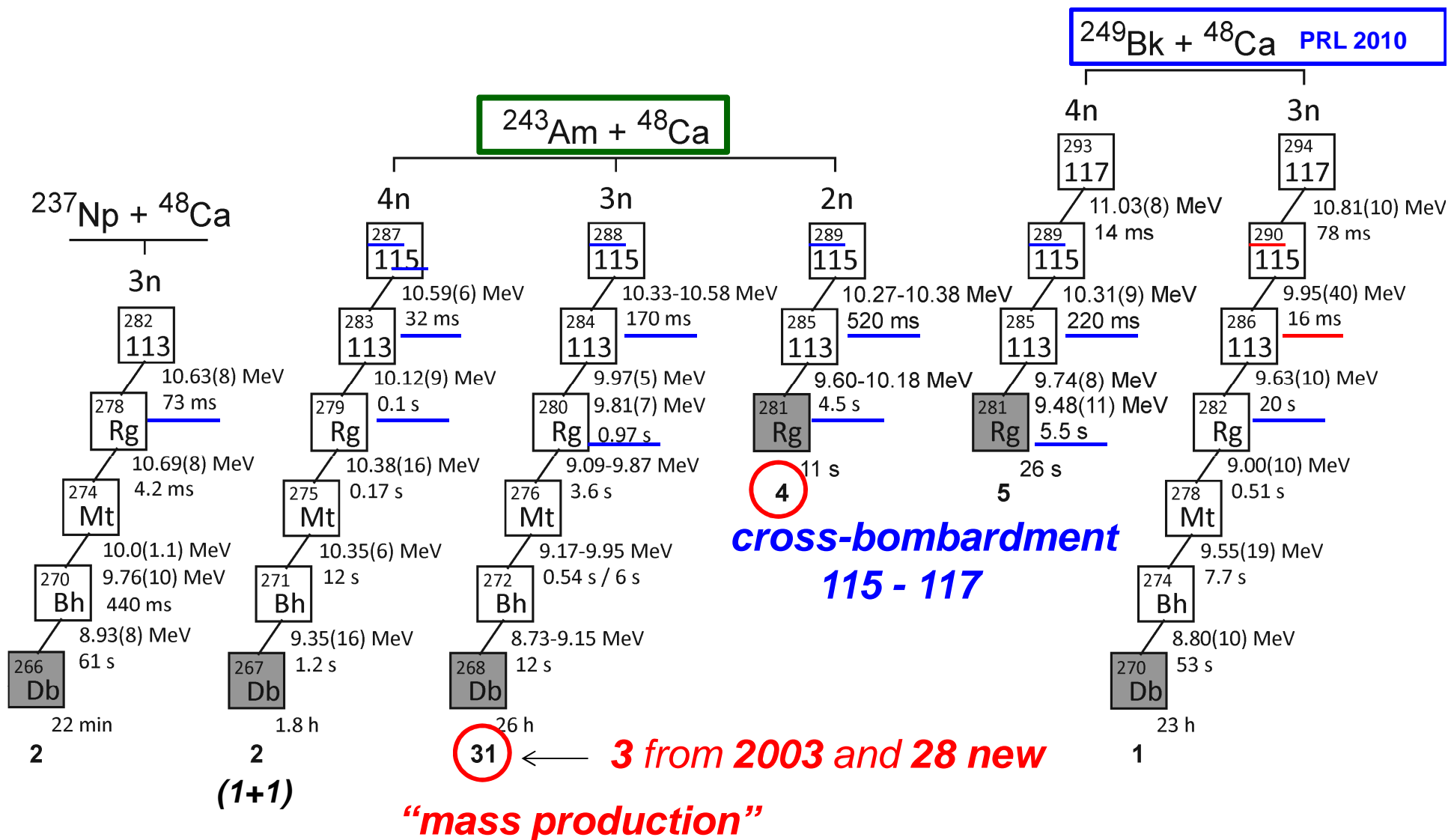


Indeed we observe an increased stability with larger neutron number  $N$ , but alpha half-lives are getting shorter with increased atomic number  $Z$  (indicating a need for fast detection in a next generation of SHE studies!)

# *new experiments at the DGFRS (JINR Dubna)*

- $^{243}\text{Am}+^{48}\text{Ca}$ , November 1<sup>st</sup>, 2010 - February 27<sup>th</sup>, 2012, beam dose  $3.3 \cdot 10^{19}$
- *verification of 2003-2004 results on Z=115, 113, and “mass production”*
- *$\alpha$ -decay properties (fine structure ?)*
- *excitation function*
- $^{289}115$  from **2n** reaction channel - *“cross bombardment”* for  $^{293}117$

# New study of Z=115 decay chains



total number of observed decay chains (2003, 2010 - 2012) is listed at the end of the chain

## *new experiments at the DGFRS (JINR Dubna)*

- $^{249}\text{Bk} + ^{48}\text{Ca}$ , from April 23<sup>rd</sup>, 2012 till October 23<sup>rd</sup>, 2012, beam dose  $\sim 4.6 \cdot 10^{19}$ 
  - $\alpha$ -decay properties, in particular of  $^{294}117$  chain (only 1 event detected earlier)
  - new isotope  $Z=109$   $^{277}\text{Mt}$  and its very fast fission !
  - excitation function (search for  $^{295}117$  produced in  $2n$  reaction channel)
  - evidence for  $^{294}(118)$  decay chain

# New studies of super heavy nuclei with $Z=97$ $^{249}\text{Bk}$ and $Z=98$ $^{249}\text{Cf}$ target materials at Dubna (2012)

## 11 new events of $^{293}(117)$

ER	# 9	# 23	# 12	# 25	# 7
	9.91 MeV	13.23 MeV	11.76 MeV	11.00 MeV	9.36 MeV
$^{293}_{117}$	10.90 <sup>10</sup> MeV <sup>a</sup> 7.525 ms	11.142 <sup>65</sup> MeV 3.305 ms	11.114 <sup>89</sup> MeV <sup>a</sup> 153.948 ms	10.914 <sup>68</sup> MeV 10.547 ms	10.598 <sup>49</sup> MeV 109.878 ms
$^{289}_{115}$	10.37 <sup>28</sup> MeV <sup>b</sup> 0.2665 s	10.310 <sup>65</sup> MeV 0.1719 s	missing $\alpha$	10.198 <sup>68</sup> MeV 1.4348 s	10.217 <sup>49</sup> MeV 0.1510 s
$^{285}_{113}$	9.857 <sup>40</sup> MeV 1.5155 s	missing $\alpha$	9.631 <sup>67</sup> MeV 19.0456 s	9.36 <sup>30</sup> MeV <sup>b</sup> 1.3153 s	9.683 <sup>49</sup> MeV 18.3997 s
$^{281}_{\text{Rg}}$	204.0 MeV <sup>c</sup> 9.4192 s	222.0 MeV <sup>c</sup> 7.4538 s	212.8 MeV <sup>c</sup> 1.4809 s	162.9 MeV <sup>c</sup> 103.406 s	150.8 MeV 42.1349 s

## 3 new events of $^{294}(117)$

ER	# 16	# 8
	12.64 MeV	13.11 MeV
$^{294}_{117}$	10.960 <sup>70</sup> MeV 100.72 ms	10.967 <sup>73</sup> MeV 3.986 ms
$^{290}_{115}$	10.28 <sup>12</sup> MeV <sup>a</sup> 0.3010 s	9.775 <sup>73</sup> MeV 0.6976 s
$^{286}_{113}$	9.61 <sup>11</sup> MeV <sup>a</sup> 5.7895 s	9.750 <sup>11</sup> MeV <sup>a</sup> 3.6525 s
$^{282}_{\text{Rg}}$	9.18 <sup>30</sup> MeV <sup>b</sup> 145.34 s	9.04 <sup>11</sup> MeV <sup>a</sup> 29.202 s
$^{278}_{\text{Mt}}$	9.396 <sup>70</sup> MeV 4.1713 s	9.382 <sup>73</sup> MeV 7.2154 s
$^{274}_{\text{Bh}}$	8.791 <sup>70</sup> MeV 102.58 s	8.69 <sup>30</sup> MeV <sup>b</sup> 55.722 s
$^{270}_{\text{Db}}$	142.4 MeV 37.49 h	195.9 MeV <sup>c</sup> 23.49 h

$Z=97$   $^{249}\text{Bk} \rightarrow Z=98$   $^{249}\text{Cf}$

$T_{1/2} = 330$  days

$E^*(^{297}118) \sim 31$  MeV

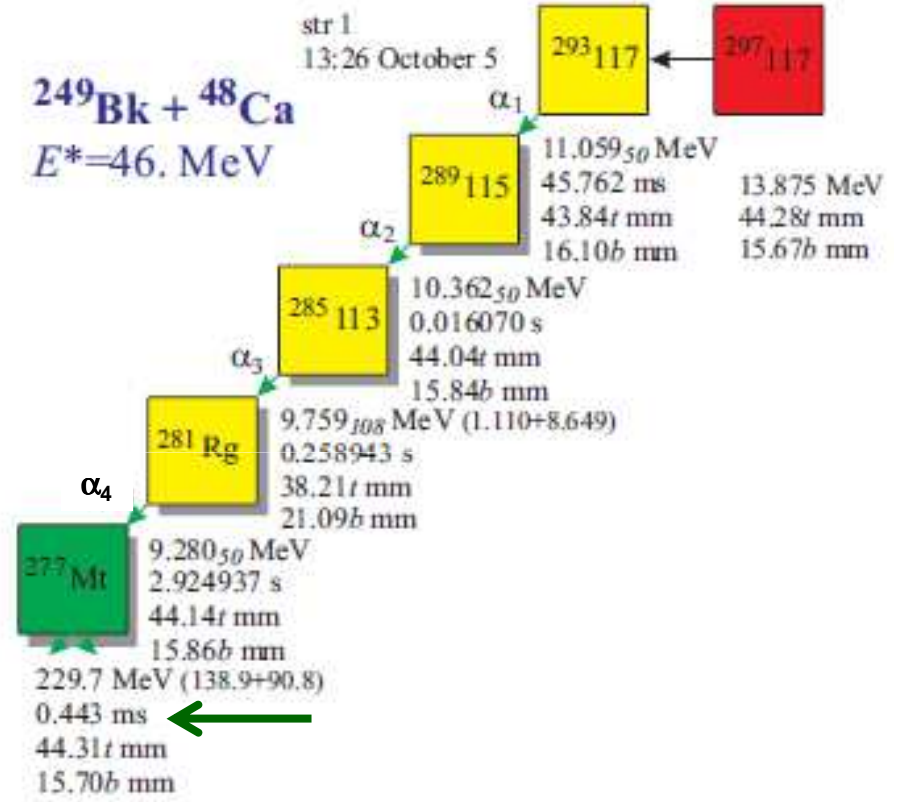
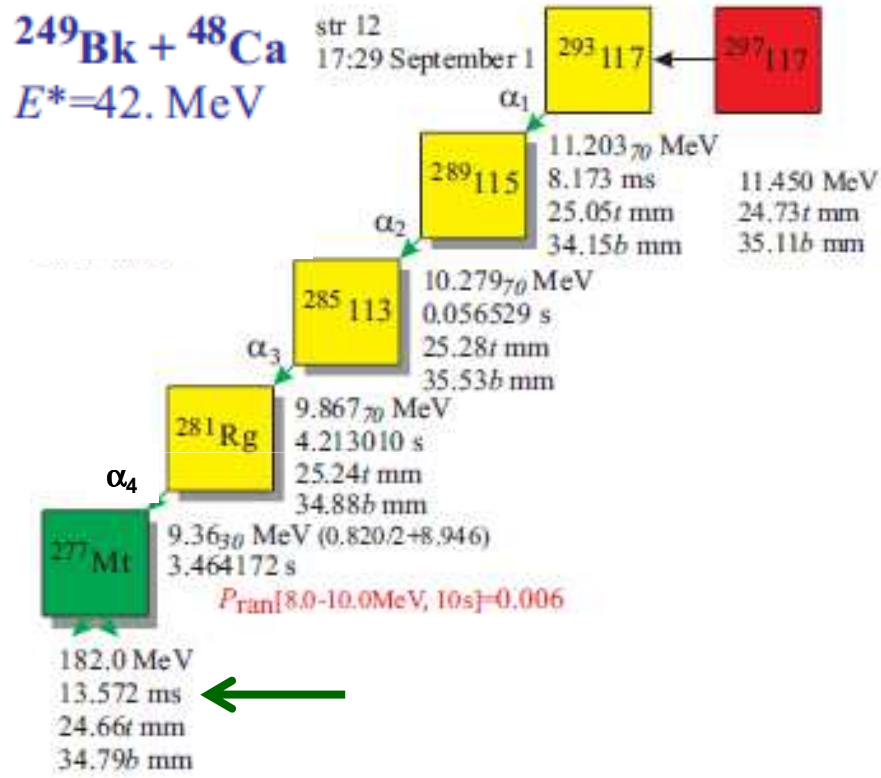
$^{249}\text{Cf} + ^{48}\text{Ca}$

ER	# 23
	11.63 MeV
$^{294}_{118}$	11.65 <sub>6</sub> MeV 0.89 <sup>+1.07</sup> <sub>-0.31</sub> ms
$^{290}_{\text{Lv}}$	10.84 <sub>8</sub> MeV 7.1 <sup>+3.2</sup> <sub>-1.7</sub> ms
$^{286}_{\text{Fl}}$	10.19 <sub>6</sub> MeV 129 <sup>+39</sup> <sub>-24</sub> ms
$^{282}_{\text{Cn}}$	0.82 <sup>+0.30</sup> <sub>-0.18</sub> ms
ER	# 23
	11.63 MeV
$^{294}_{118}$	11.679 <sup>65</sup> MeV 0.135 ms
$^{290}_{\text{Lv}}$	10.848 <sup>65</sup> MeV 29.437 ms
$^{286}_{\text{Fl}}$	219.3 MeV <sup>c</sup> 3.486 ms

new event of  $^{294}(118)$

consistent data on  $Z=117$  and  $Z=118$  production and decay

# First observation of $^{281}\text{Rg}$ $\alpha$ -decay and fast fission of new isotope $^{277}\text{Mt}$



$Z=111$   $^{281}\text{Rg}$  SF  $\sim 20$  s ( $\alpha \sim 10\%$ )

$Z=109$   $^{277}\text{Mt}$  SF  $\sim 5 (+9, -2)$  ms “fission corridor”

nuclear structure dramatically influences fission probability

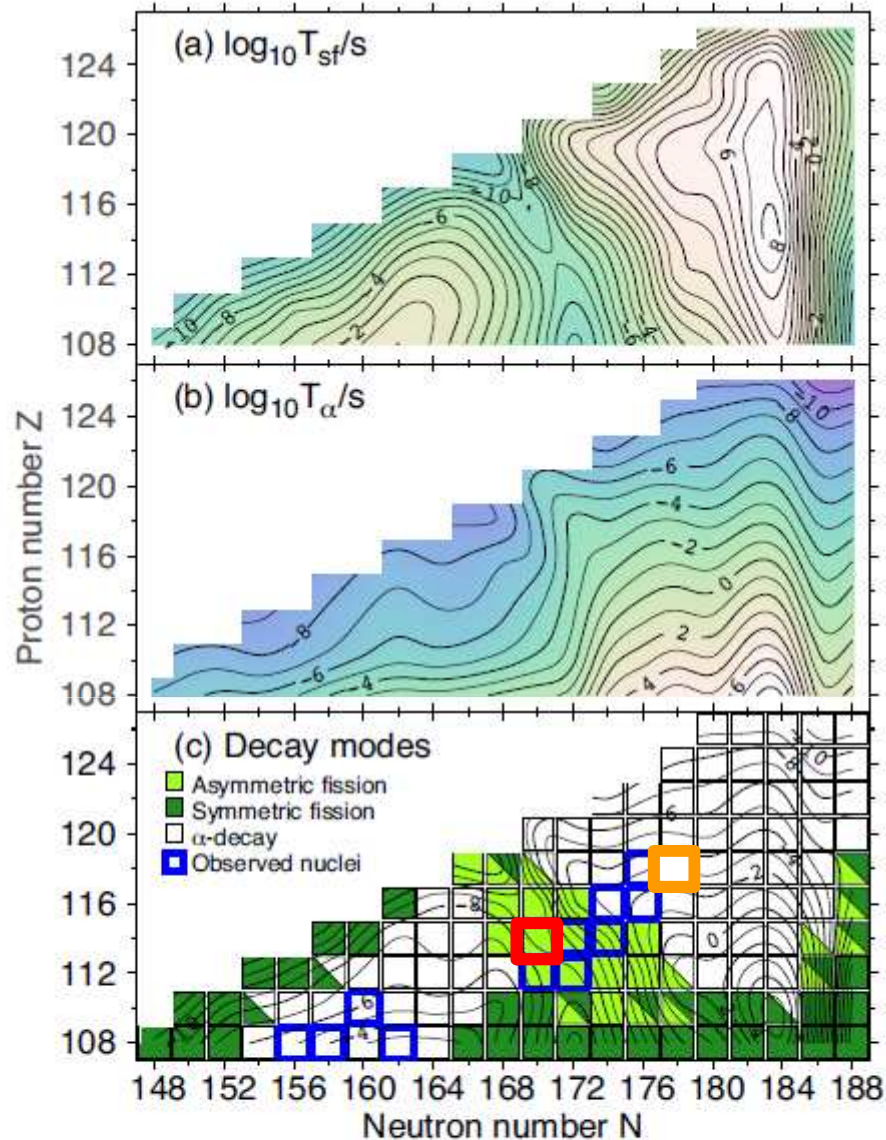


FIG. 4. (Color online) Summary of our SkM\* results for decay modes of SHE nuclei. (a) SF half-lives  $\log_{10} T_{sf}$  (in seconds). (b)  $\alpha$ -decay half-lives  $\log_{10} T_{\alpha}$  (in seconds). (c) Dominant decay modes. If two modes compete, this is marked by coexisting triangles.

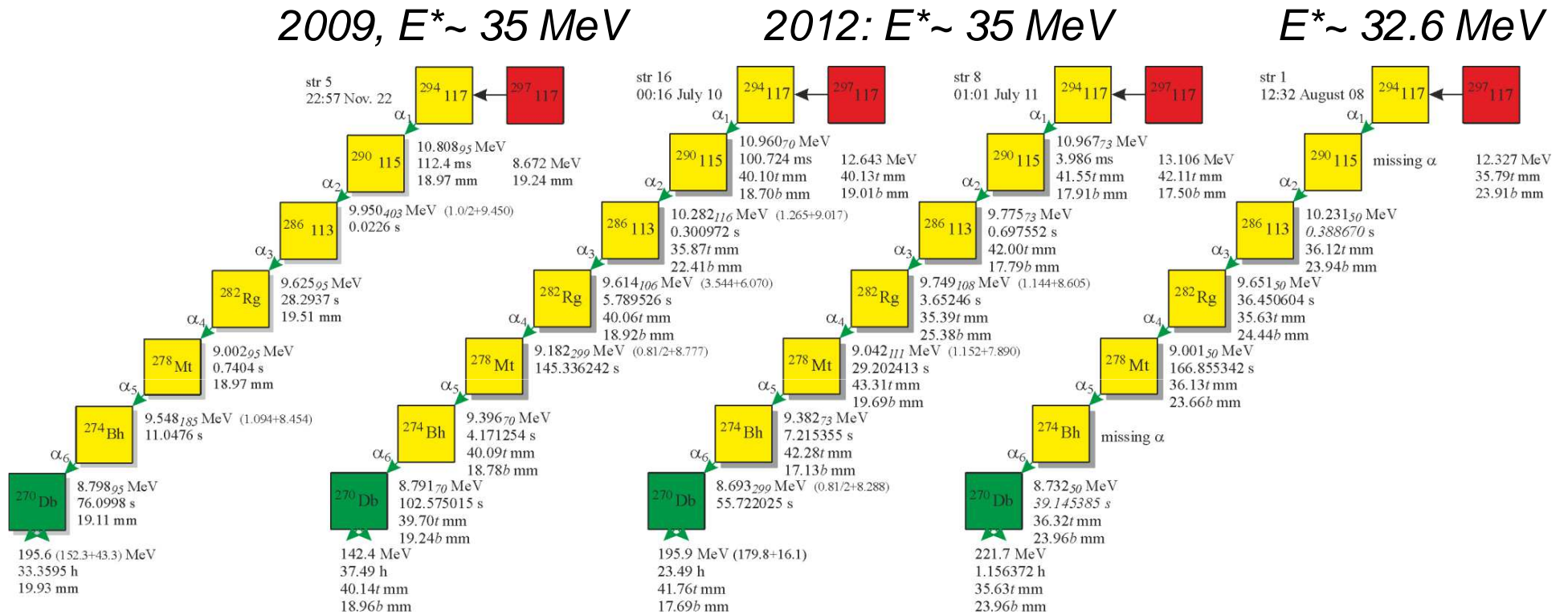
A. Staszczak, A. Baran and W. Nazarewicz  
 Phys. Rev. C 87, 024320, 2013  
**EVEN-EVEN SHE nuclei**  
 advanced analysis of shapes and  
 symmetric vs asymmetric fission modes  
 affecting SHE lifetimes

**Pretty good agreement**  
 for  $T_{1/2}$ 's of even-even alpha emitters,  
 but the SF mode  
 at the end of "Dubna Island" decay chains  
 appears to be much too fast in the calculations.  
 However, the large gradient of SF life-time  
 is indeed observed in experiment.

□  $^{284}\text{Fl}$  (now searched for)  
 $^{239,240}\text{Pu} + ^{48}\text{Ca}$ , 3,4n channel

□  $^{296}\text{118}$  (to be searched for)  
 $^{251}\text{Cf} + ^{48}\text{Ca}$ , 3n channel

# Total of four $^{294}_{117}$ decay chains found at Dubna (3n channel of $^{249}\text{Bk}+^{48}\text{Ca}$ reaction)



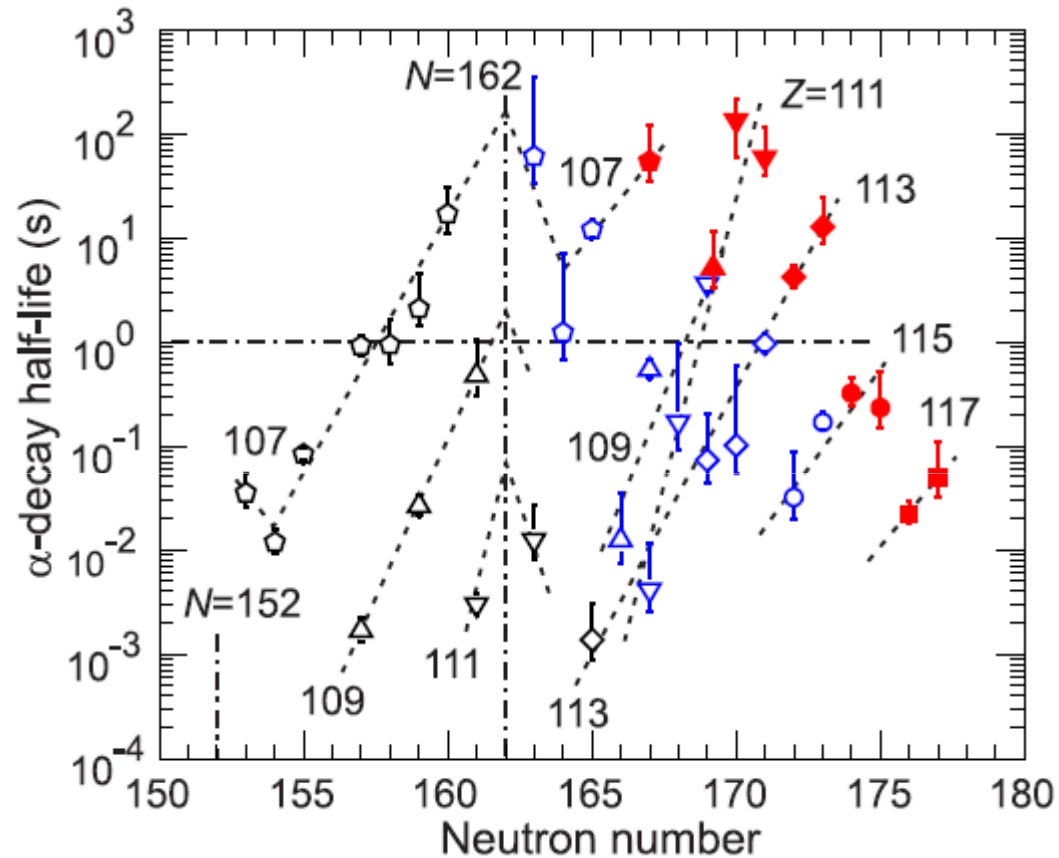
all decay chains ended with SF of  $Z=105$   $^{270}\text{Db}$ ,  $T_{1/2}(\text{SF}) \sim 17$  h

$E^* \sim 35$  MeV denotes  $E^*(^{297}117)$  ranging from 32.8 MeV to 37.5 MeV

Oganessian et al., PRL 109, 162501, 2012, PRC 87, 054621, 2013



# ***Towards the Island of (enhanced) Stability 2013***



***Better statistics on Z=117 and Z=115 decay chains confirms enhanced stability with increasing neutron number***

## *experiments at TASCA (GSI Darmstadt)*

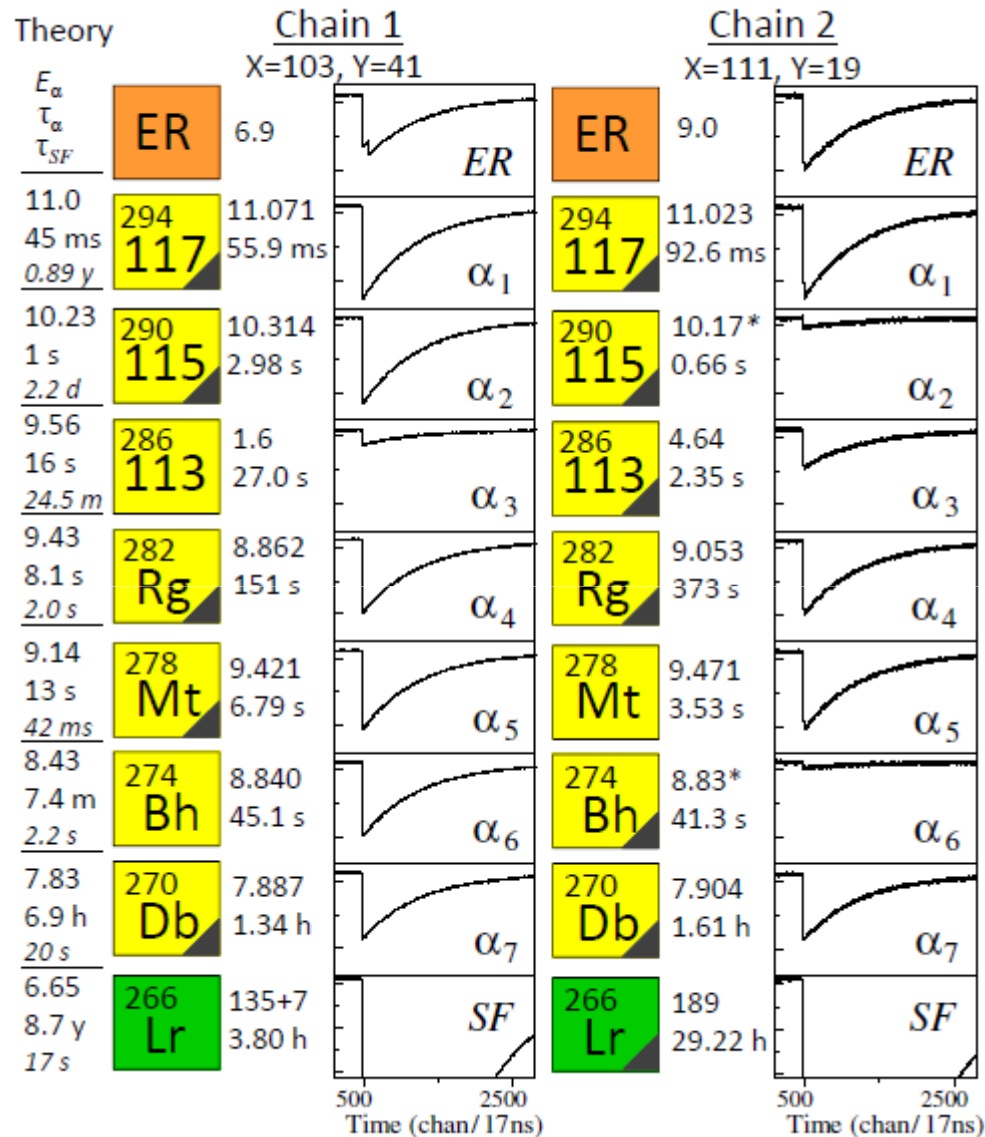
- $^{249}\text{Bk}+^{50}\text{Ti}$ , TASCA, April – September 2012, beam dose  $\sim 3.6 \cdot 10^{19}$ 
  - search for isotopes of new element  $Z=119$ ,  $^{295,296}(119)_{174,175}$
  - cross section limit of about **70 femtobarn** reached
- $^{249}\text{Bk}+^{48}\text{Ca}$ , TASCA, September - October 2012, beam dose  $\sim 1.1 \cdot 10^{19}$ 
  - search for isotopes of element  $Z=117$ ,  $^{293,294}(117)$  */GSI-ORNL scientific agreement/*



**12.7 mg  $^{249}\text{Bk}$  from ORNL**  
**impressive production of  $^{249}\text{Bk}$  target at Mainz**  
(Klaus Eberhardt, Norbert Trautmann,  
Jörg Runke and their collaborators)

*Rotating target survived  $\sim 4$  part \* microAmps  
i.e.,  $2.4 \cdot 10^{13}$   $^{50}\text{Ti}$  projectiles per 0.25 second  
over several months of irradiations*

# Element 117 independently confirmed at TASCA !



*Jadambaa Khuyagbaatar et al.,  
→ PRL 2014*

*among coauthors:*

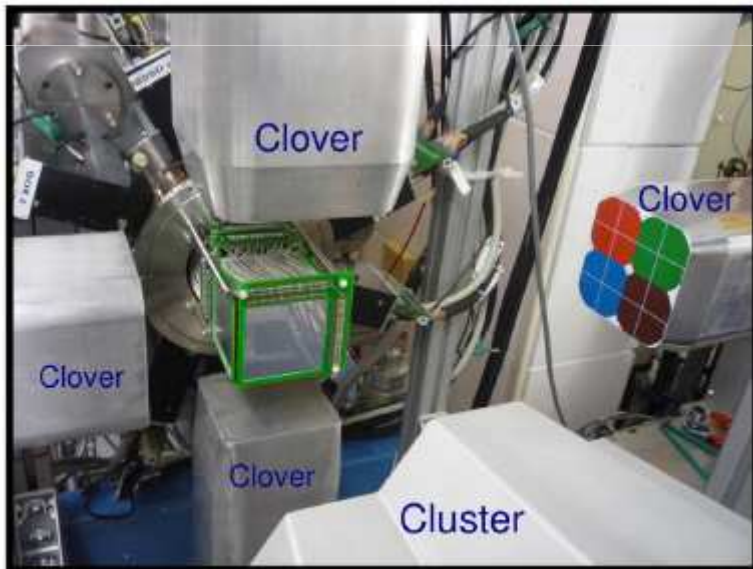
*K. Miernik*

*M. Węgrzecki (ITME)*

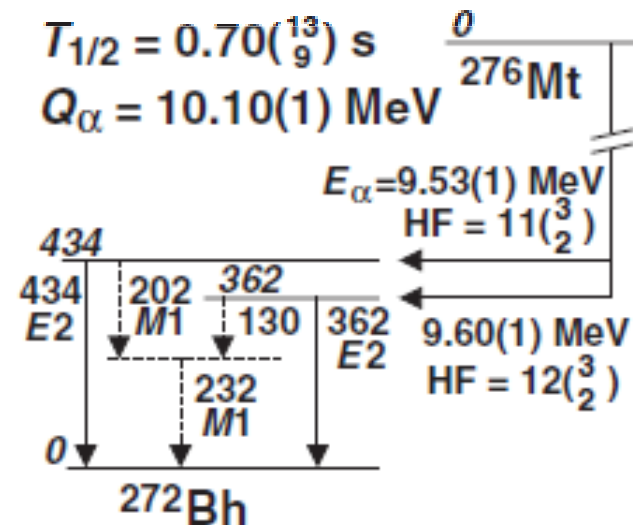
**NEW:**  
 **$\alpha$ -decay of  $^{270}\text{Db}$**   
**and new SF isotope  $^{266}\text{Lr}$**

**GSI Darmstadt**     *D. Rudolph et al., PRL 111, 112502, 2013*

- $^{243}\text{Am} + ^{48}\text{Ca}$ , TASCA-TASISpec,  $^{243}\text{Am}$  (ORNL),  $0.8 \text{ mg/cm}^2$  target (Mainz)
  - 3 weeks in Nov. 2012, beam dose  $\sim 7 \cdot 10^{18}$
  - study of known isotopes of element  $Z=115$ , e.g.,  $^{288}(115)$
  - independent confirmation of 2003-2004 and 2010-2012 Dubna data on  $Z=115, 113, \dots$
  - attempt to directly measure atomic number  $Z$  through  $\alpha$ -X ray correlation
  - **30 decay chains** of  $Z=115$  isotopes observed in three weeks !



$\sim 40\%$  efficiency at KX-ray region



*first  $\gamma$ -spectroscopy at Hot Fusion Island*

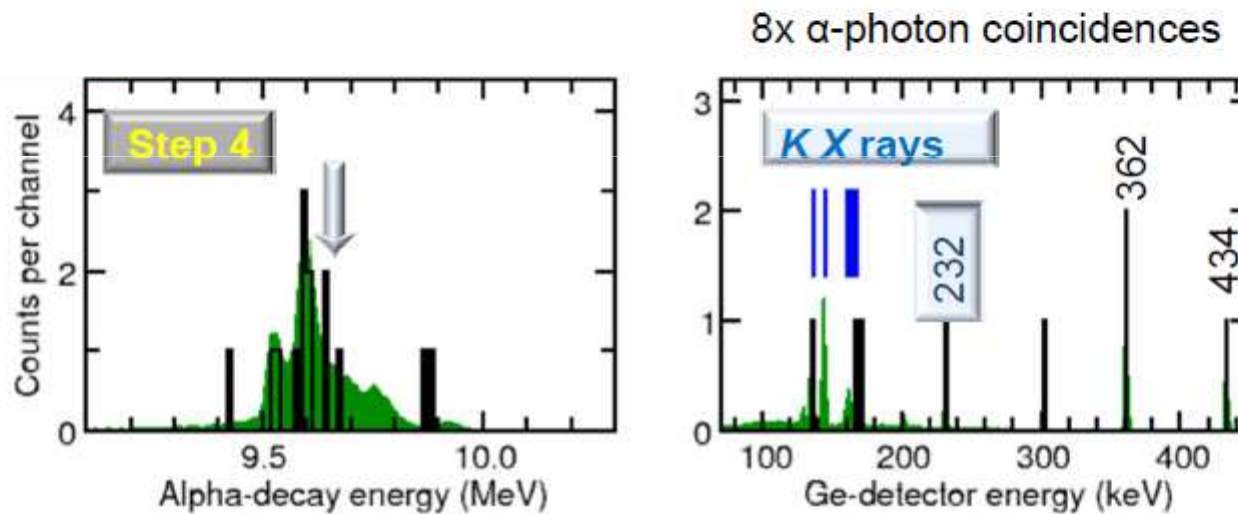
# Results – $^{288}\text{115}$ (3n-chain)

**X-ray Case?**



Chain 5 :  $E_{\alpha} = 9.65(1)$  MeV  
 $E_{\text{ph}} = 232(1)$  keV

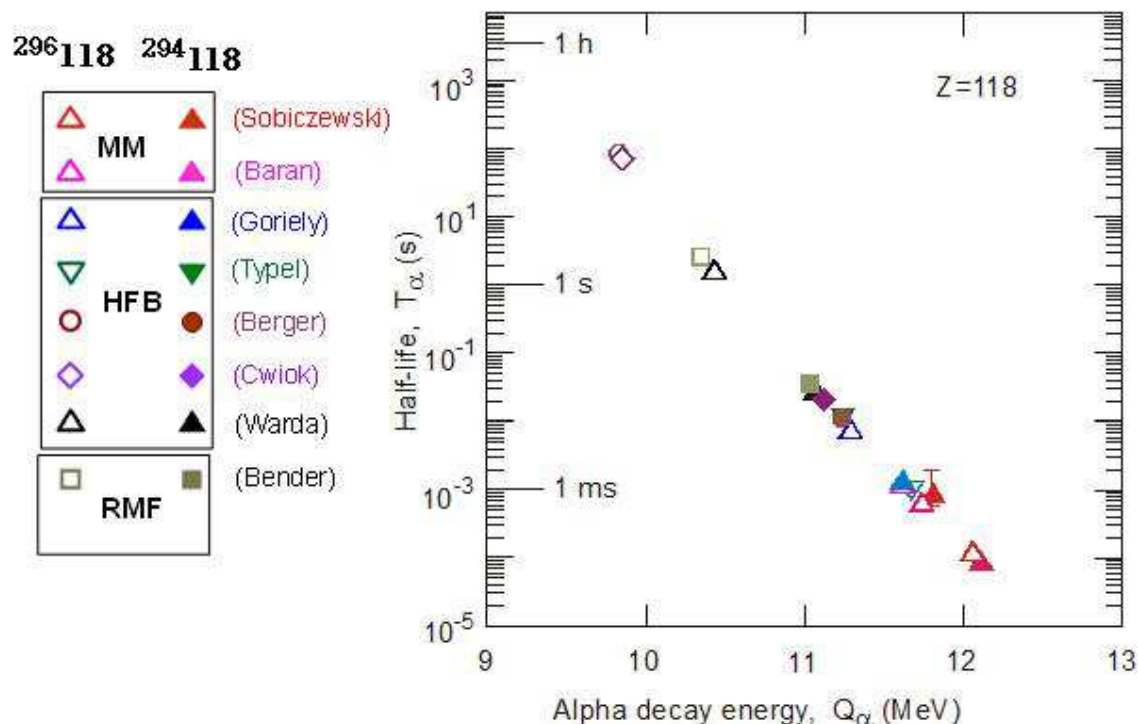
cascade (of 2 K X rays?)



GEANT4 simulations: 100000 decays, normalized to number of  $\alpha$ 's

# Plans for 2014-2015: search for Z=118 isotopes

Models do not agree about the decay properties of  $^{294}118$  and  $^{296}118$



exp data on  $^{294}118$   $\alpha$ -decay:  
 $E_{\alpha}=11.7$  MeV,  $T_{1/2} \sim 0.7$  ms

Staszczak, Baran, Nazarewicz  
*Phys. Rev. C* 87, 024320, 2013

$^{294}118$ :  $E_{\alpha}=11.8$  MeV,  $T_{1/2}=0.66$  ms  
 $^{296}118$ :  $E_{\alpha}=11.7$  MeV,  $T_{1/2}=1$  ms

long experiment with a mixed-Cf target from ORNL and  $^{48}\text{Ca}$  beam  
has a potential to identify new isotopes  $^{293}118$ ,  $^{295}118$  and  $^{296}118$

# Mixed-Cf target ORNL 2013-2014

## Radiochemical Engineering Development Center REDC

~ 40 years old Cf-sources processed at REDC to obtain mixed-Cf material

$^{249}\text{Cf}$ (351 y)	$^{250}\text{Cf}$ (13 y)	$^{251}\text{Cf}$ (898 y)	$^{252}\text{Cf}$ (2.65 y)
7.6 mg	2.5 mg	5.7 mg	0.007 mg
48.1%	15.6%	30.3%	0.04%

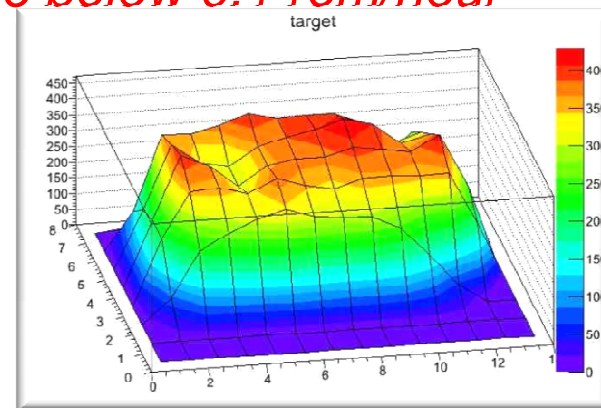
~  $3 \times 10^7$  n/s, unshielded dose in 6" ~ 1 rem/hour  
 polyethylene shield reduces the radiation dose below 0.1 rem/hour



work on mixed-Cf  
at REDC



first target sector  
ready in Dec. 2013



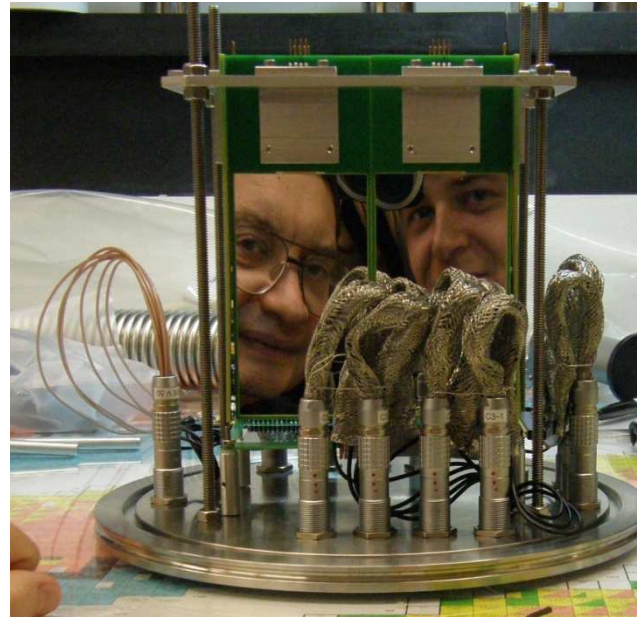
radiographic image  
of Sm test target

# ***New ORNL-UTK detectors and digital data acquisition system*** (similar DAQ at SHIP Z=120 exp was serving PSSD+Si-box+MCPs)

## *MICRON*

*128 x 48 mm  
1 mm wide strips DSSD  
and  
matching Si-veto detector*

*six 120 x 65 mm single Si  
forming Si-box*

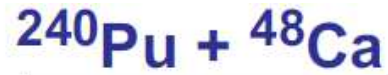


*MESYTEC  
lin-log preamps  
XIA Pixie16 rev D  
(208 channels)  
Dell Power Edge*





# Connecting Hot Fusion Island with Nuclear Mainland search for new Z=114 Flerovium isotopes and their decay products



4n    3n    2n

Fl/283 4 ms 10.8	Fl/284 2 ms 10.6	Fl/285 0.1 s 10.3	Fl/286 0.12 s 10.19	Fl/287 0.48 s 10.02	Fl/288 0.80 s 9.94	Fl/289 2.6 s 9.82
Cn/279 1 ms 10.9	Cn/280 0.4 ms 10.6	Cn/281 0.1 s 10.31	Cn/282 0.82 ms 9.54	Cn/283 3.8 s 9.15	Cn/284 97 ms 9.15	Cn/285 29 s 9.15
Ds 275 50 μs 11.2	Ds 276 0.2 ms 10.8	Ds 277 6 ms 10.57	Ds 278 10 ms 10.1	Ds 279 0.20 s 9.70	Ds 281 11 s 9.15	
Hs 271 1 s 9.3	Hs 272 0.2 s 9.5	Hs 273 0.2 s 9.59	Hs 274 10 ms 9.4	Hs 275 0.19 s 9.30	Hs 277 3 ms 9.15	
Sg 267 80 s 8.2	Sg 268 10 s 8.3	Sg 269 2 min 8.57	Sg 270 1 s 8.6	Sg 271 1.9 m 8.54		
Rf 263 8 s 8.0	Rf 264 5 s 7.8	Rf 265 2 m 7.4	Rf 266 10 s 7.4	Rf 267 1.3 h 7.4		

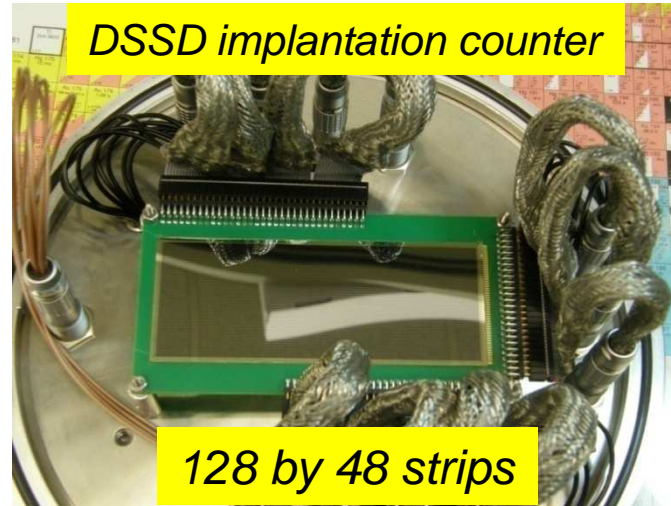
$^{240}\text{Pu}$  material from Oak Ridge  
delivered to Dubna on 27<sup>th</sup> Dec. 2013  
(after 51 weeks of paperwork)



# ORNL-UTK 8-detector and digital data acquisition system operating at the Dubna Gas Filled Recoil Separator (DGFRS)

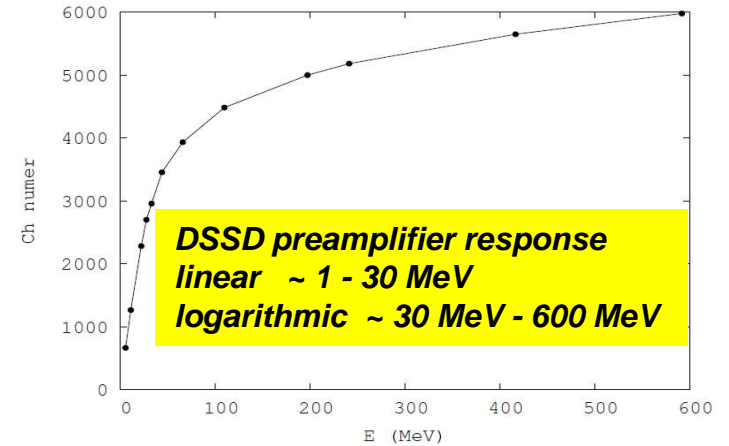


Si-detector stack



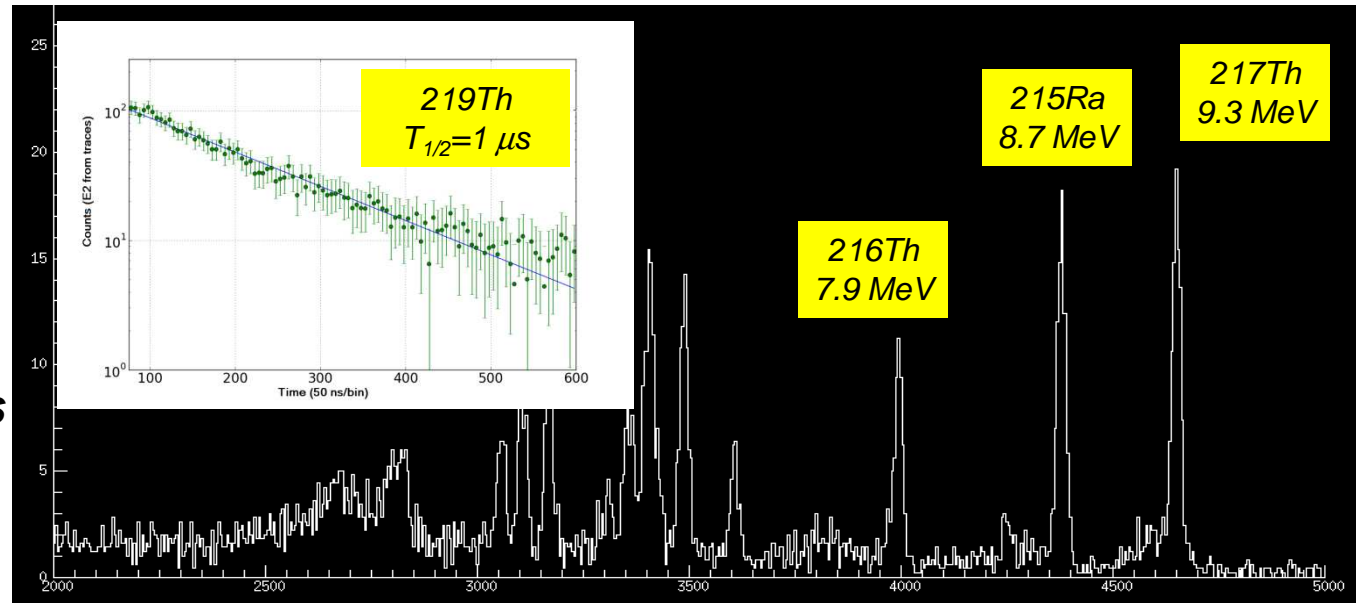
DSSD implantation counter

128 by 48 strips



$\alpha$ -emitters including  
 1  $\mu$ s activity of  $^{219}\text{Th}$   
 studied at the DGFRS  
 during  $^{48}\text{Ca} + ^{\text{nat}}\text{Yb}$  run  
 Nov.-Dec. 2013.

Search for new isotopes  
 of element  $Z=114$  (FI)  
 with  $^{239}, ^{240}\text{Pu}$  targets  
 started on 6<sup>th</sup> Dec. 2013

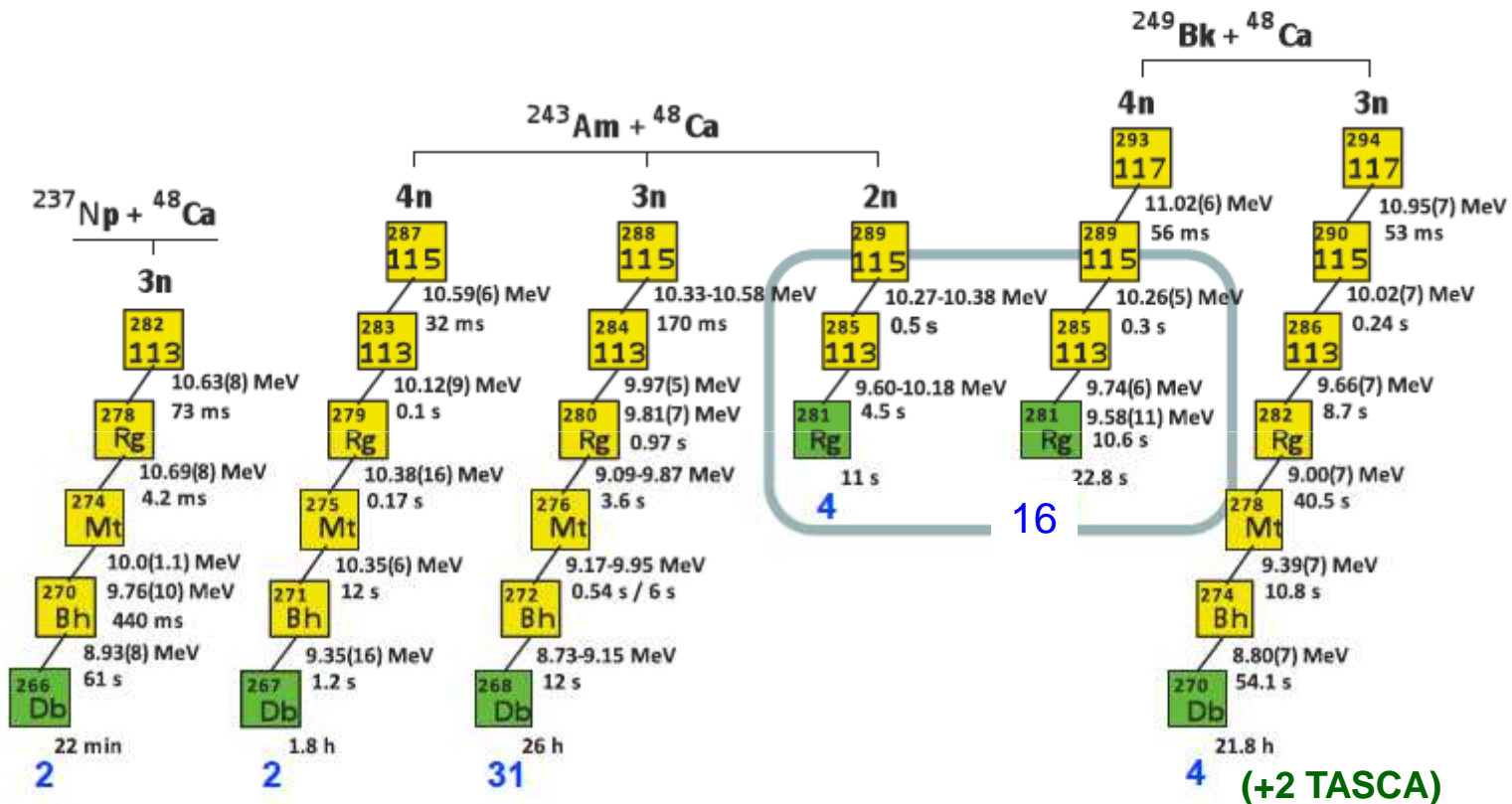


energy of  $\alpha$ -particles

# Summary

2003-2013

odd Z nuclei produced in  $^{48}\text{Ca}$ -induced reactions



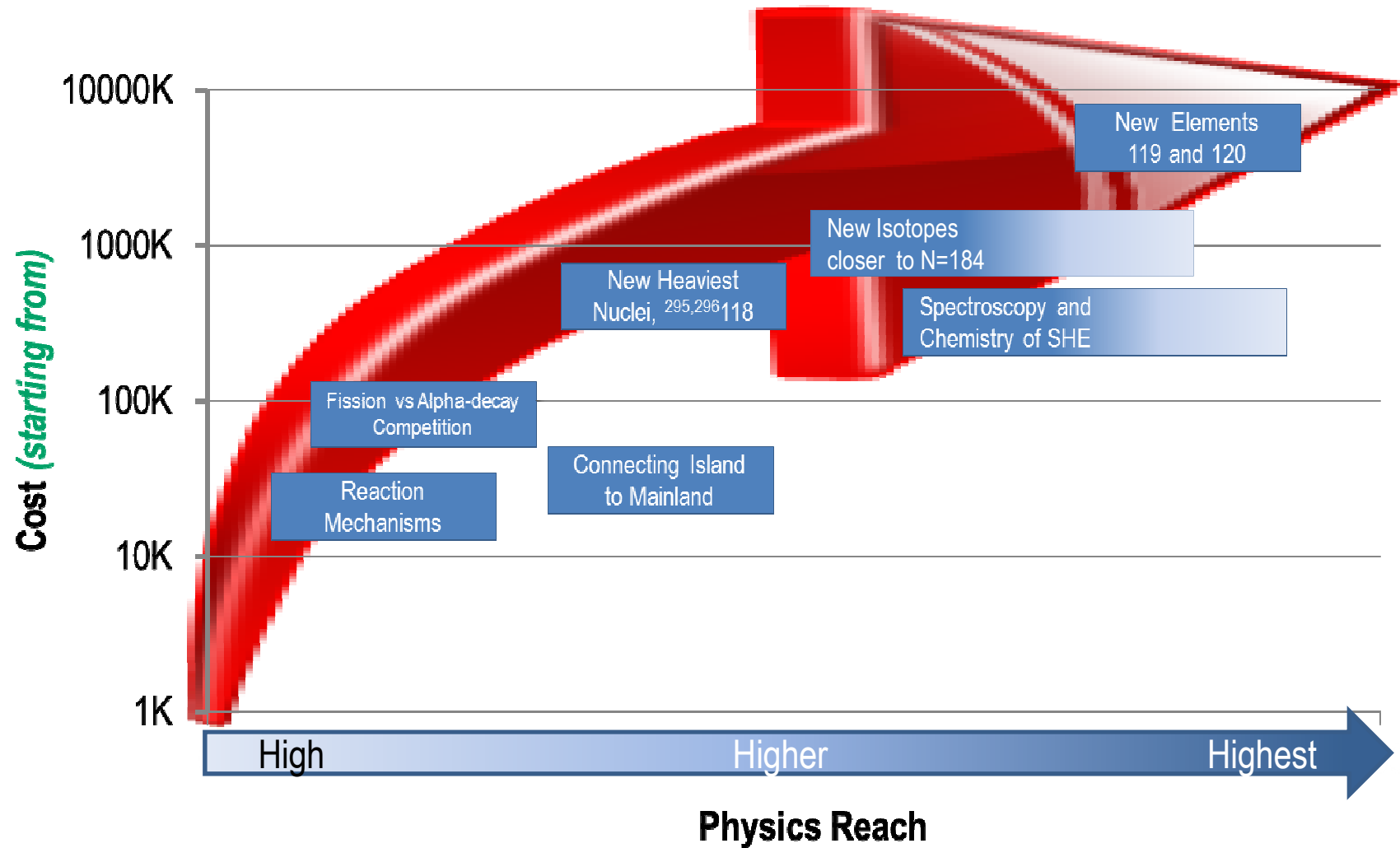
number of the detected chains  
(+30 TASCA + ~ 40 BGS)

Yu.Ts. Oganessian et al.,  
PRL **104**, 142501, 2010, PRL **108**, 022502, 2012, PRL **109**, 162501, 2012.

# Summary

- new data consistent with earlier results on  $Z=117, 115, 113$  and  $118$
- Dubna discoveries of elements  $115$  and  $113$  (2003-2004) and of  $117$  (2010) independently confirmed at TASCA (GSI)
- “cross bombardment” achieved in  $^{48}\text{Ca}$ -induced reactions:  
 $4n$  ( $^{249}\text{Bk}$  target,  $^{293}117$ ) and  $2n$  ( $^{243}\text{Am}$ ,  $^{289}115$ ) reaction channels
- nearly “mass production” of  $Z=115$  isotopes,  $\sigma_{\text{MAX}} \sim 9 \text{ pb}$
- total of  $\sim 100$  decay events of  $^{288}115$  and 16 events of  $^{293}117$  and  $^{294}117$  (4 +2 events) help to determine the decay properties (new isotope  $^{266}\text{Lr}$ )
- an evidence for fine structure in  $\alpha$ -spectra and  $\alpha$ -KX-ray events
- new isotope  $^{277}\text{Mt}$  ( $\sim 5 \text{ ms SF}$ !) ending the  $^{293}117$  decay chain
- $\sigma(117) \sim 2\text{-}3 \text{ pb}$ ,  $^{294}118$  observed with ingrown  $^{249}\text{Cf}$  target component
- **new SHE experiments with important ORNL/UTK contributions at Dubna**

# Approach to Highest Value Experiments (e.g., at Dubna SHE Factory)



# Timeline of SHE Experiments (Dubna-ORNL)

