



Nikola Cindro (1931 - 2001)

# EXTENSIONS OF FISSION THEORY OF NUCLEAR CLUSTER DECAY

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# HFR and resonances

N. Cindro and M. Božin

Heavy-fragment radioactivities and resonances in heavy-ion collisions: A correlation, *Phys. Rev.* **39** (1989) 1665.

**Abstract** A strong correlation is found to exist between the phenomena of heavy-fragment radioactivity and high transparency in the molecular window for the same nuclei taken, in turn, as emitters and composite systems. Such a correlation sheds new light on the nature of heavy-fragment radioactivity and provides a new way of searching for possible resonant phenomena in heavy nuclei.



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# OUTLINE

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- Macroscopic-microscopic method
- Unified approach of cold fission,  $\alpha$ -decay and heavy ion radioactivities within ASAF model
- Experimental confirmations
- Fine structure
- Extensions
  - saddle-point shapes obtained as solution of an Euler-Lagrange equation
  - $\alpha$ -decay of superheavies (ASAF, universal curve, semi-empirical formula)
  - multicluster fission (true ternary, quaternary, etc)
  - atomic cluster on a surface and fission of charged clusters



# Macroscopic-microscopic method

Accounting for quantum single-particle structure and classical collective properties.

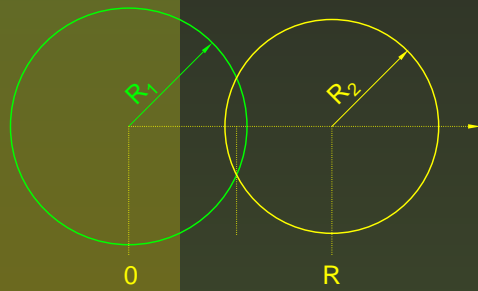
- Liquid Drop Model:  $E_{LD}$
- Single-particle shell model (SPSM): energy levels vs. deformation. *Two-center shell model for fission and fusion.*
- Shell correction method:  $\delta E = \delta U + \delta P$
- Total deformation energy:  $E_{def} = E_{LD} + \delta E$

The potential of SPSM Hamiltonian should admit the drop eq.  $\rho = \rho(z)$  as an equipotential surface.

**Semi-spheroidal shape**, allows to obtain analytical results for atomic clusters on a surface.



# Intersected spheres



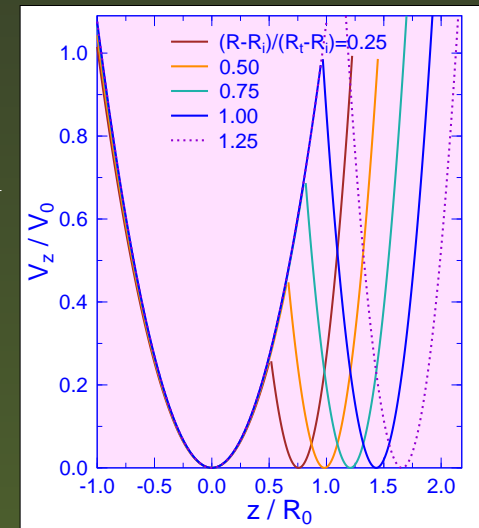
Two intersected spheres. Volume conservation and  $R_2 = \text{const.}$  One deformation parameter: separation distance  $R$ . Surface equation  $\rho = \rho(z)$ . Initial  $R_i = R_0 - R_2$ . Touching point  $R_t = R_1 + R_2$ .

Example:  $^{232}\text{U} \rightarrow ^{24}\text{Ne} + ^{208}\text{Pb}$

Two center shell model (Frankfurt) potential



Sequence of shapes



# Liquid drop model

Nucleus considered a uniformly charged drop. Two variants: LDM and Yukawa-plus-exponential (Y+EM).

LDM (surface + Coulomb) deformation energy

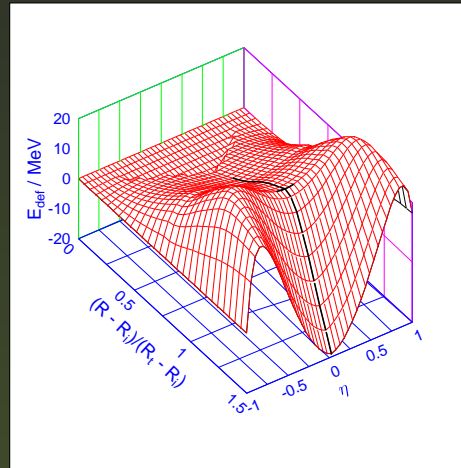
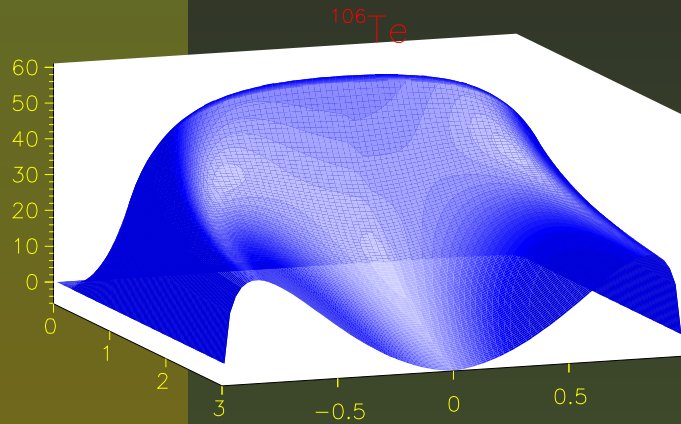
$$\begin{aligned} E_{LDM} &= E - E^0 = (E_s - E_s^0) + (E_C - E_C^0) \\ &= E_s^0(B_s - 1) + E_C^0(B_C - 1) \end{aligned}$$

For spherical shapes  $E_s^0 = a_s(1 - \kappa I^2)A^{2/3}$ ;  $I = (N - Z)/A$ ;  
 $E_C^0 = a_c Z^2 A^{-1/3}$ . Nuclear fissility  $X = E_C^0 / (2E_s^0)$ .

Parameters obtained by fit to experimental data on nuclear masses, quadrupole moments and fission barriers:  $a_s = 17.9439$  MeV,  $\kappa = 1.7826$ ,  $a_c = 3e^2 / (5r_0)$ ,  $e^2 = 1.44$  MeV·fm,  $r_0 = 1.2249$  fm.  
W.D. Myers and W.J. Swiatecki, Nucl. Phys. A **81** (1966) 1



# LDM PES and saddle-point shapes



Potential energy surfaces (PES) for  $^{106}\text{Te}$  (left) and  $^{232}\text{Th}$  (right)

$X = 0.60$

$X = 0.70$

$X = 0.82$

Saddle point shapes for fissility parameter  $X = 0.60, 0.70, 0.82$  ( $^{170}\text{Yb}$ ,  $^{204}\text{Pb}$ ,  $^{252}\text{Cf}$  nuclei) obtained by solving an integro-differential equation.

D.N. Poenaru, R.A. Gherghescu, W. Greiner, *Nucl. Phys. A* **747** (2005) 182–205.



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# Shell corrections

The total energy of the uniform level distribution

$$\tilde{u} = \tilde{U} / \hbar\omega_0^0 = 2 \int_{-\infty}^{\tilde{\lambda}} \tilde{g}(\epsilon) \epsilon d\epsilon$$

In units of  $\hbar\omega_0^0$  the shell corrections are calculated for each deformation  $\varepsilon$

$$\delta u(n, \varepsilon) = \sum_{i=1}^n 2\epsilon_i(\varepsilon) - \tilde{u}(n, \varepsilon)$$

$n = N_p/2$  particles. Then  $\delta u = \delta u_p + \delta u_n$ .





# Pairing corrections

The gap  $\Delta$  and Fermi energy  $\lambda$  are solutions of the BCS eqs:

$$0 = \sum_{k_i}^{k_f} \frac{\epsilon_k - \lambda}{\sqrt{(\epsilon_k - \lambda)^2 + \Delta^2}} ; \quad \frac{2}{G} = \sum_{k_i}^{k_f} \frac{1}{\sqrt{(\epsilon_k - \lambda)^2 + \Delta^2}}$$

$$k_i = Z/2 - n + 1, \quad k_f = Z/2 + n', \quad \frac{2}{G} \simeq 2\tilde{g}(\tilde{\lambda}) \ln \left( \frac{2\Omega}{\tilde{\Delta}} \right).$$

The pairing correction  $\delta p = p - \tilde{p}$ , represents the difference between the pairing correlation energies for the discrete level distribution

$$p = \sum_{k=k_i}^{k_f} 2v_k^2 \epsilon_k - 2 \sum_{k=k_i}^{Z/2} \epsilon_k - \frac{\Delta^2}{G} \text{ and for the continuous level}$$

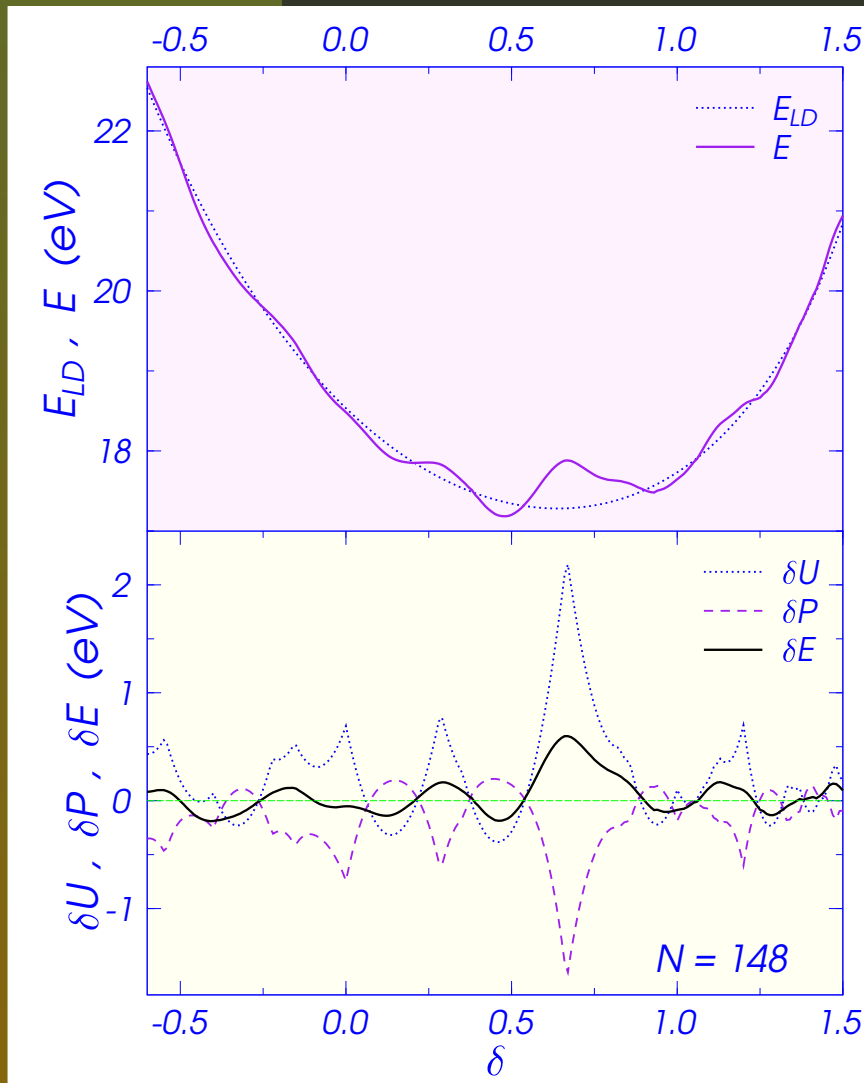
distribution  $\tilde{p} = -(\tilde{g}\tilde{\Delta}^2)/2 = -(\tilde{g}_s\tilde{\Delta}^2)/4$ . Compared to shell correction,

the pairing correction is out of phase and smaller. One has again

$$\delta p = \delta p_p + \delta p_n, \text{ and } \delta e = \delta u + \delta p.$$



# Example: Na<sub>148</sub> atomic cluster



$E_v = -333$  eV was not included in  $E_{LD}$  and  $E$ . Liquid drop and total deformation energy (top). Shell plus pairing corrections for hemispheroidal harmonic oscillator energy levels (bottom).

**Smoothing effect of pairing.**

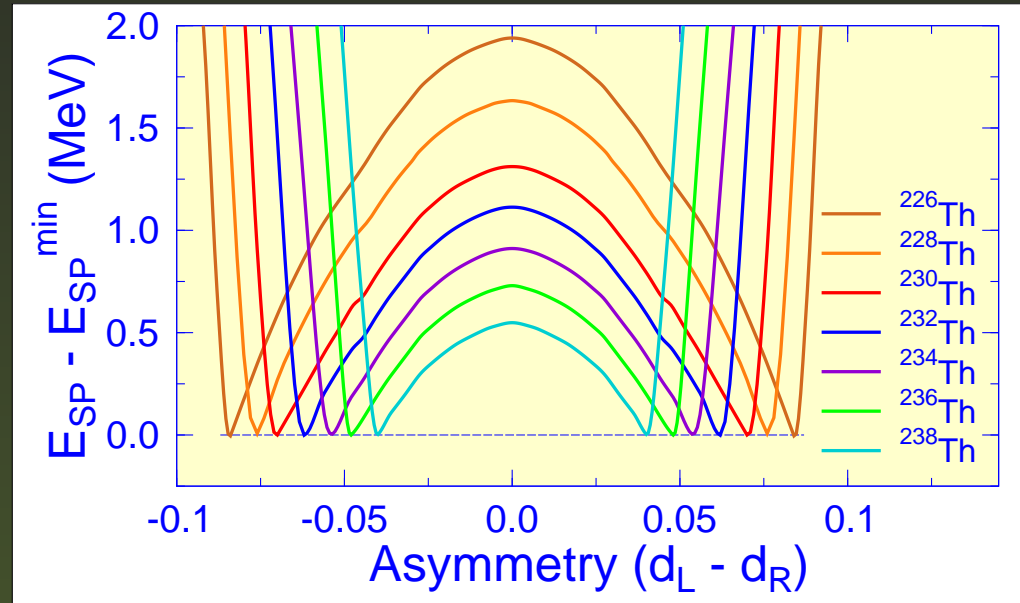
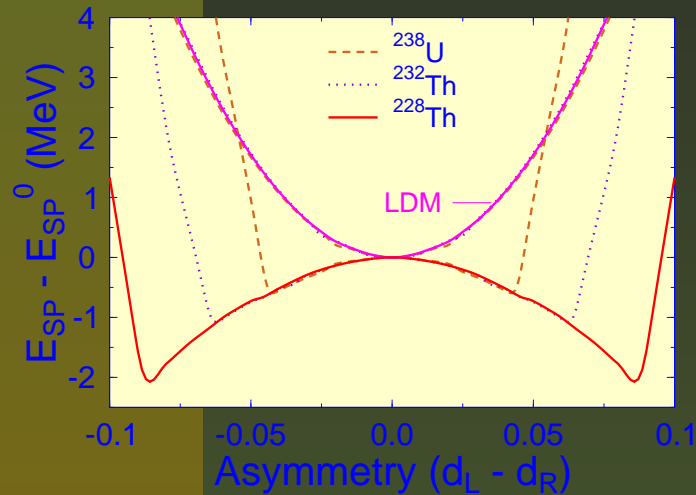
Ground state shape prolate

$$\delta = 0.47$$

$$\text{Semiaxes ratio } \frac{a}{c} = \frac{2-\delta}{2+\delta}$$



# Mass asymmetry



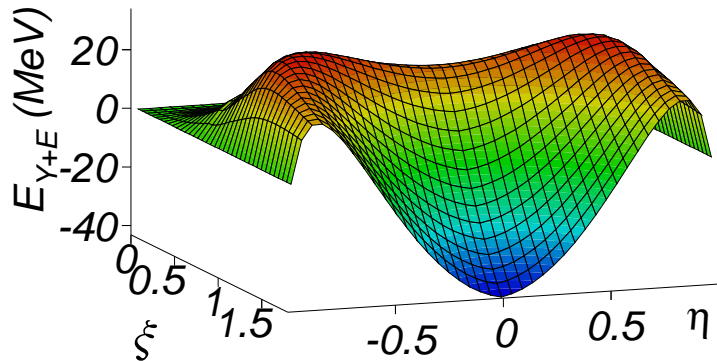
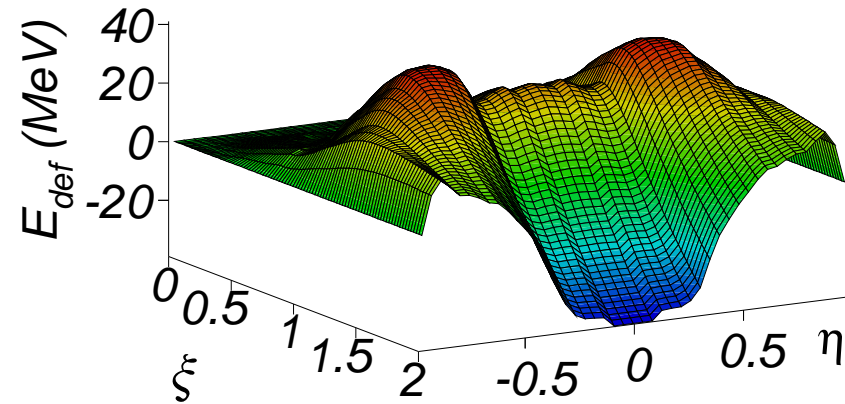
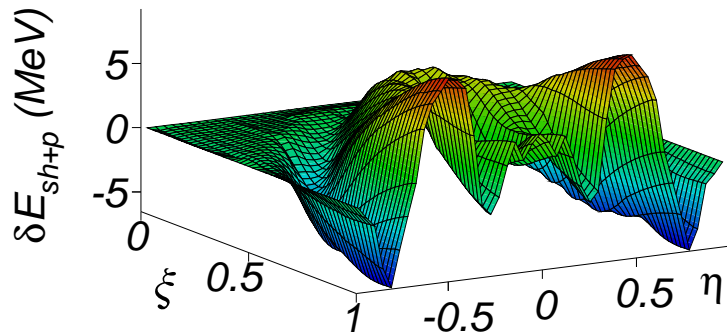
Shell effects explain the mass asymmetry. Saddle point nuclear shape obtained as a solution of integro-differential equation.

D.N. Poenaru, R.A. Gherghescu, W. Greiner, *Nucl. Phys. A* **747** (2005) 182–205.



$^{222}\text{Ra}$

$E_{Y+EM}, \delta E_{shell+pair}, E_{def}$  PES



separation distance

$$\xi = (R - R_i)/(R_t - R_i)$$

mass asymmetry

$$\eta = (A_1 - A_2)/(A_1 + A_2)$$

Poenaru, Gherghescu, W.Greiner, *Phys. Rev. C* 73 (2006) 014608



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# Basic relationships

Parent  $\rightarrow$  emitted ion + daughter nucleus,  ${}^A Z \rightarrow {}^{A_e} Z_e + {}^{A_d} Z_d$

Measurable quantities

- Kinetic energy of the emitted cluster  $E_k = Q A_1 / A$  or the released energy  $Q = M - (M_e + M_d) > 0$ .
- Decay constant  $\lambda = \ln 2 / T$  or Half-life ( $T < 10^{32}$  s) or branching ratio  $b_\alpha = T_\alpha / T$  ( $b_\alpha > 10^{-17}$ )

Model dependent quantities ( $\lambda = \nu S P_s$ )

- $\nu$  frequency of assaults or  $E_\nu = h\nu / 2$
- $S$  preformation probability
- $P_s$  penetrability of external barrier



# Fission theory

Shape parameters: fragment separation,  $R$ , and mass asymmetry

$$\eta = (A_d - A_e)/A.$$

Our method to estimate preformation as penetrability of internal barrier:  $S = \exp(-K_{ov})$ . **DNP, WG, *Physica Scripta* 44 (1991) 427.**

Similarly  $P = \exp(-K_s)$  for external barrier.

Action integral calculated within Wentzel-Kramers-Brillouin (WKB) quasiclasical approximation

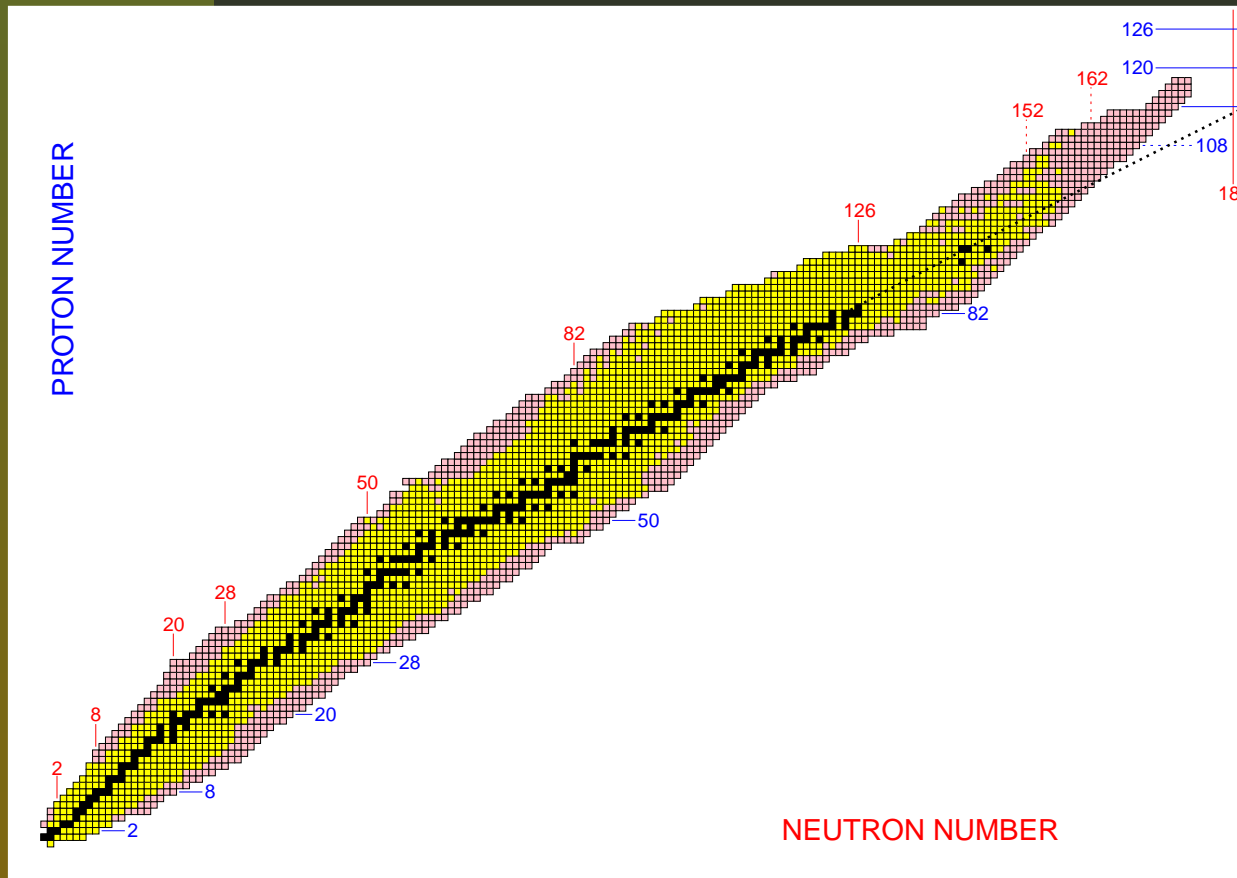
$$K_{ov} = \frac{2}{\hbar} \int_{R_i}^{R_t} \sqrt{2B(R)E(R)} dR$$

$E$  – Potential barrier

$B = \mu$  – Nuclear inertia = reduced mass for  $R \geq R_t$



# Experimental masses



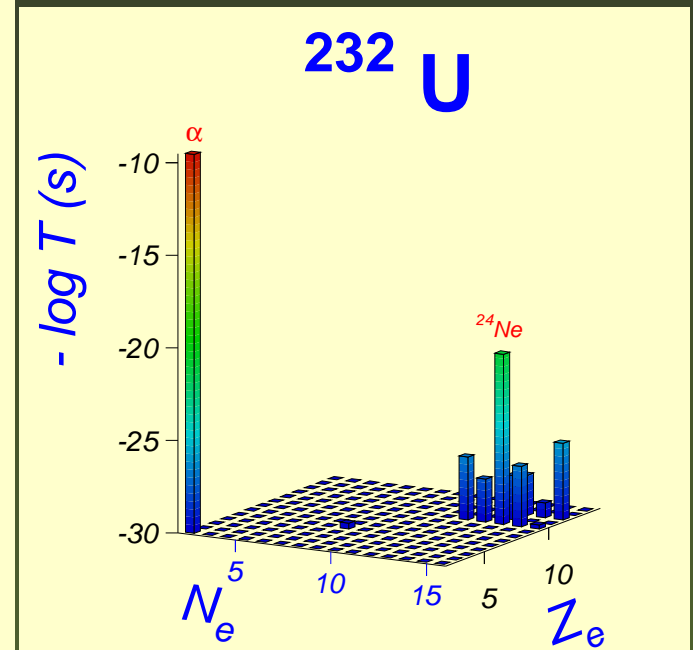
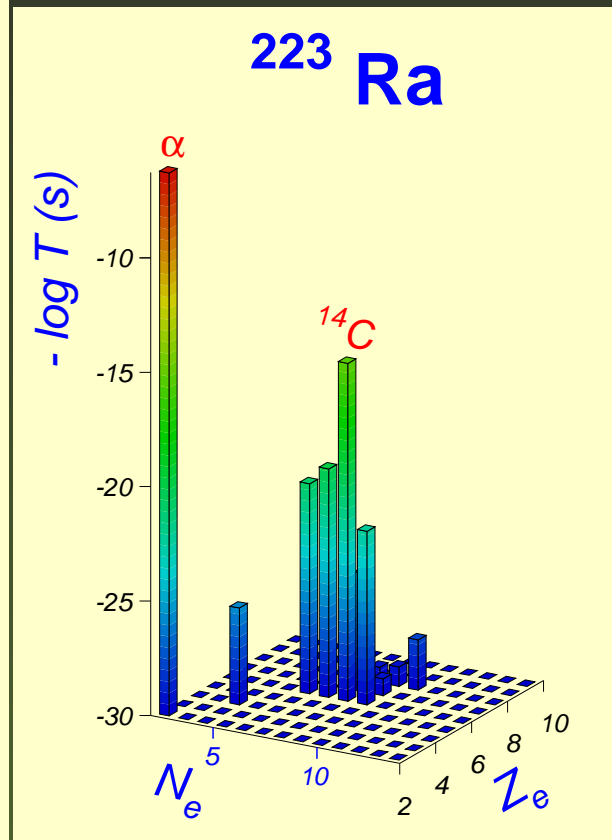
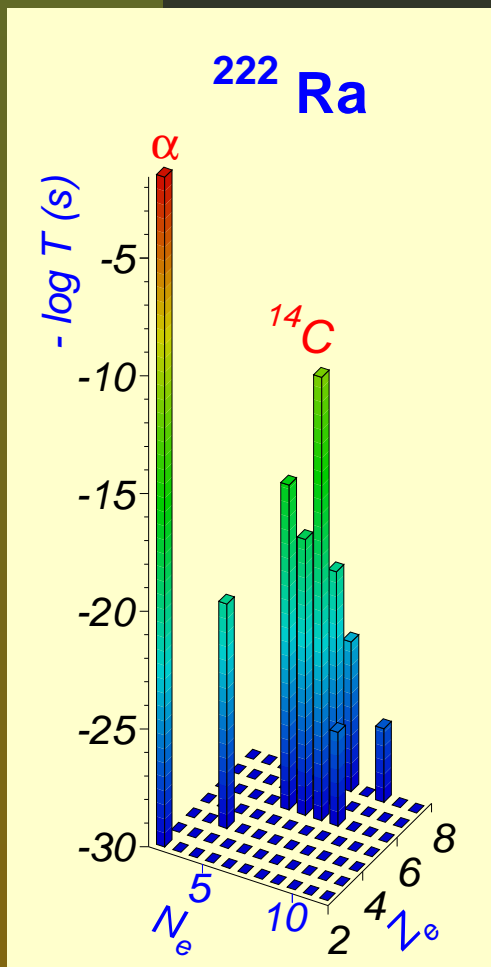
3290 nuclei,  
2377 measured  
and 913 det. from  
Systematics.

G. Audi, W.  
Meng, Private  
communication  
2011.



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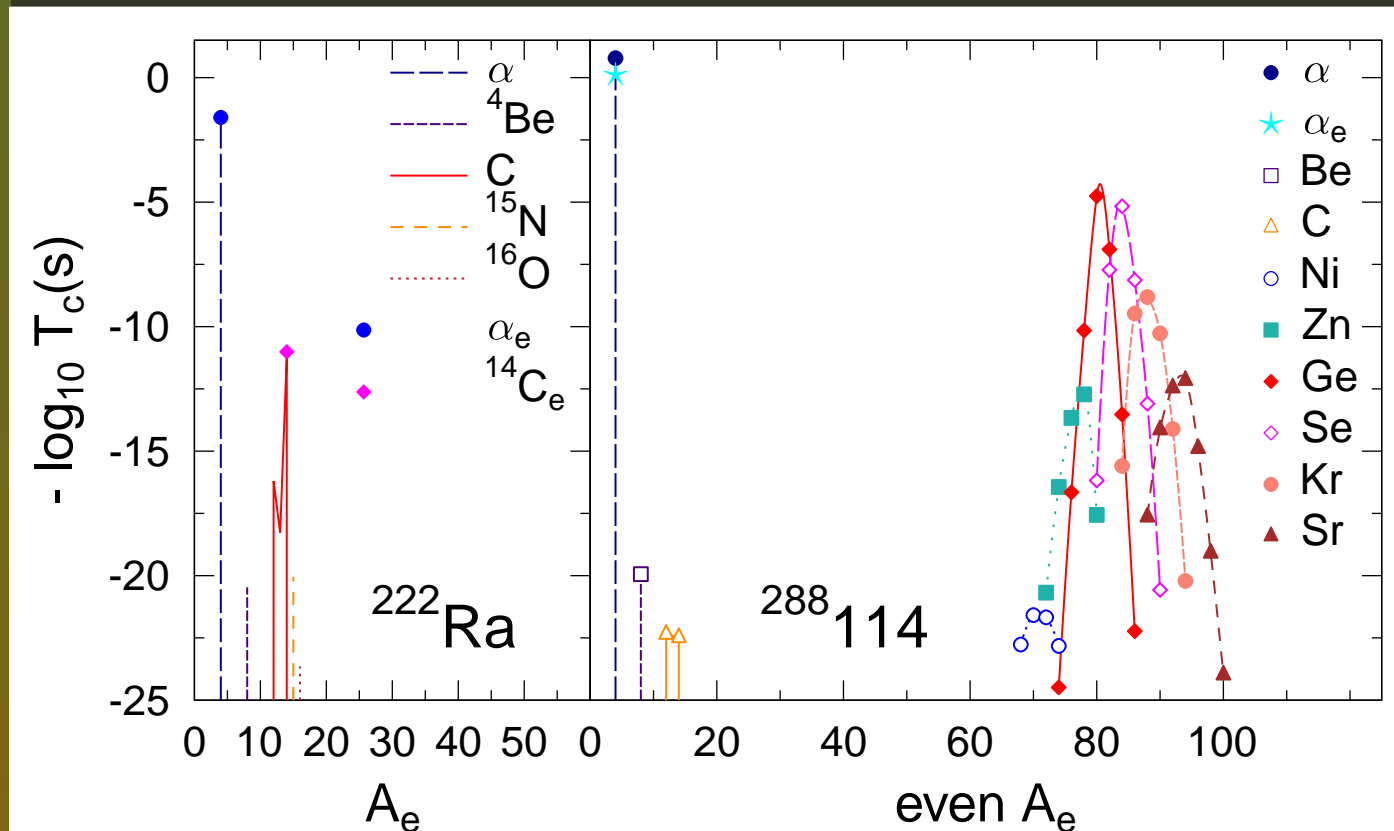
# Examples of time spectra (I)



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# Examples of time spectra (II)

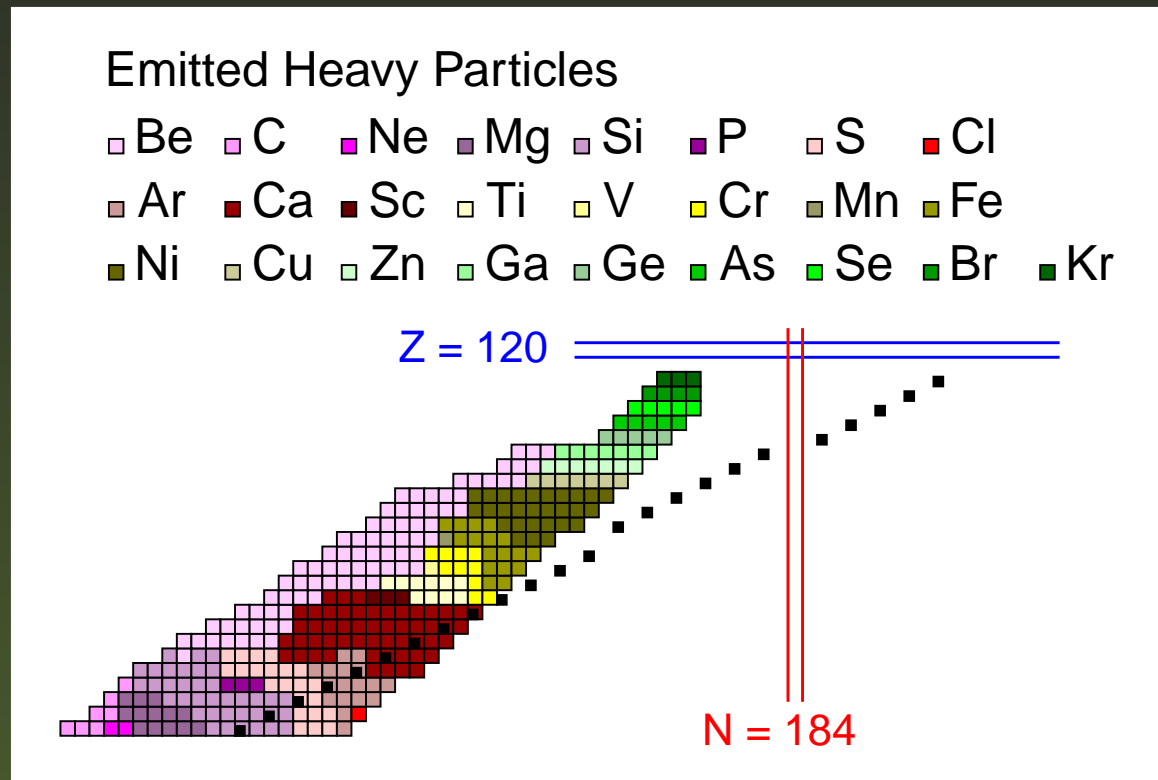


${}^{288}114 \rightarrow {}^{80}\text{Ge} + {}^{208}\text{Pb}$ . D.N. Poenaru, R.A. Gherghescu, W. Greiner, *Phys. Rev. Lett.* **107** (2011) 062503.



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# Superheavies as cluster emitters



New concept: for  $Z > 110$   $Z_e > 28$  to get a daughter around  $^{208}\text{Pb}$ .

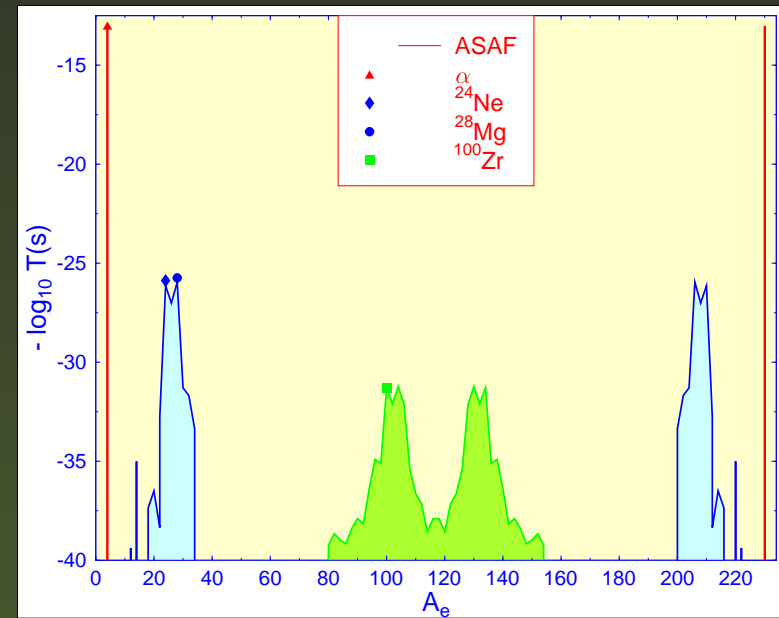
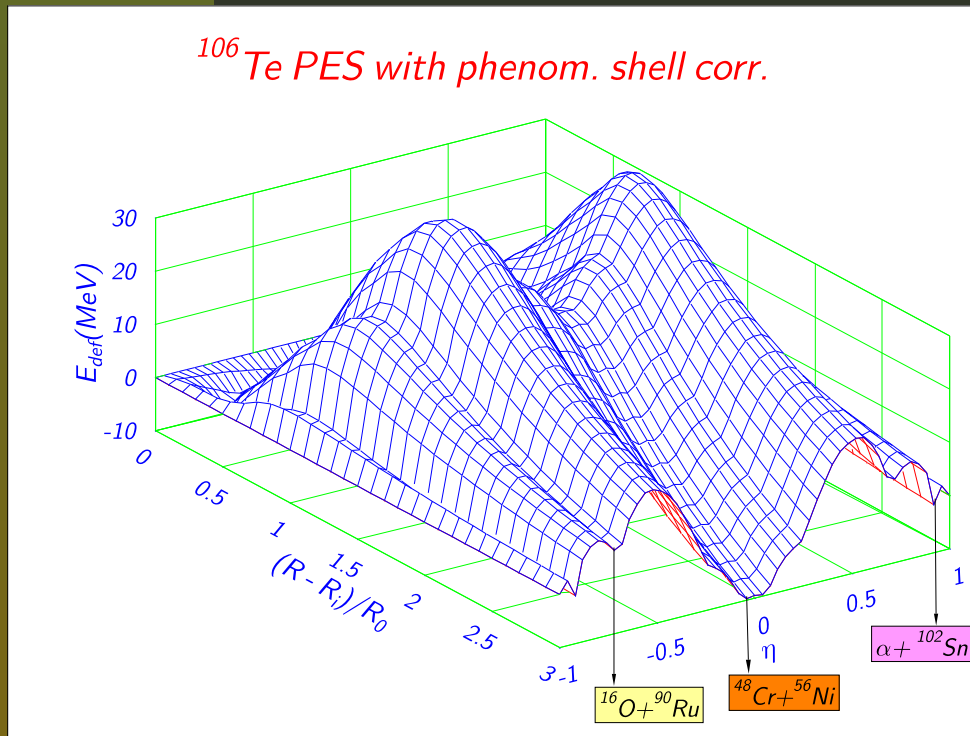
D.N. Poenaru, R.A. Gherghescu, W. Greiner,

*Phys. Rev. Lett.* **107** (2011) 062503.



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# Unified approach: CF; HPR, and $\alpha$ -d



Three valleys: cold-fission (almost symmetrical);  $^{16}\text{O}$  radioactivity, and  $\alpha$ -decay

$^{234}\text{U}$  half-lives spectrum  
(short T up)



# Experimental confirmations

Rare events in a strong background of  $\alpha$  particles

Detectors:

- Semiconductor telescope + electronics
- Magnetic spectrometers (SOLENO, Enge split-pole)
- Solid state nuclear track det. (SSNTD). Cheap and handy. Need to be chemically etched then follows microscope scanning

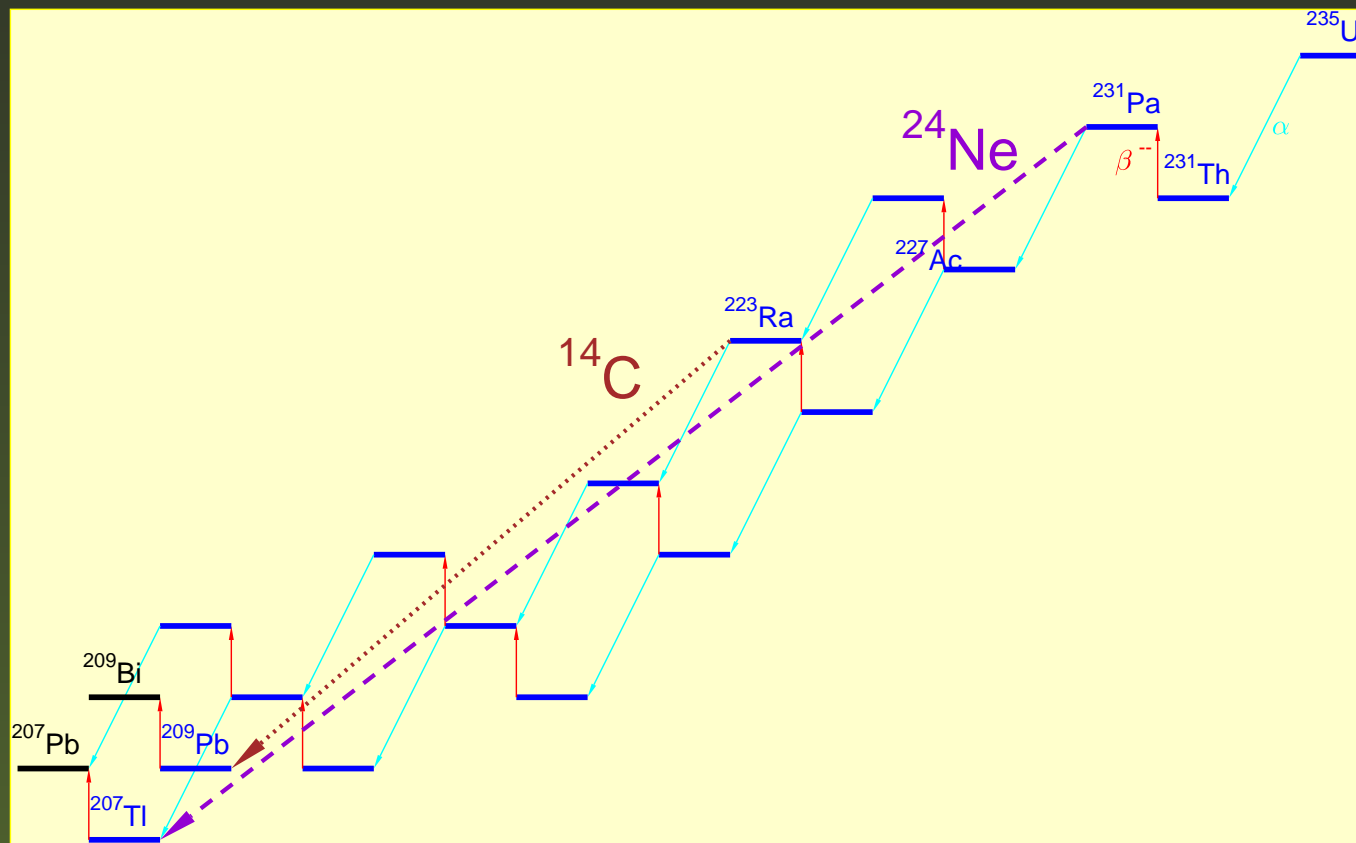
Experiments performed in Universities and Research Institutes from: Oxford; Moscow; Orsay; Berkeley; Dubna; Argonne; Livermore; Geneva; Milano; Vienna, and Beijing. Table: **R. Bonetti** and A. Guglielmetti, Rom. Rep. Phys. **59** (2007) 301.



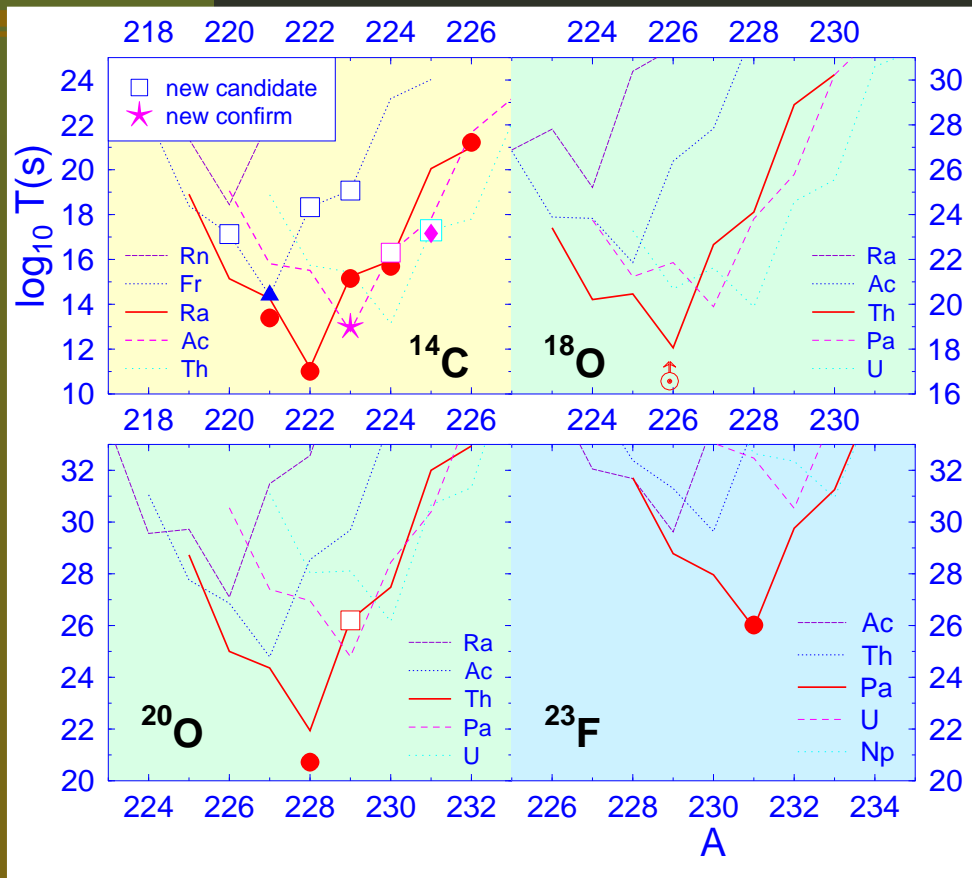
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# Natural radioactive family

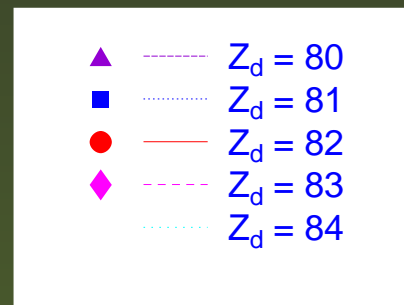
Compare  $\alpha$  and  $\beta^-$  to  $^{14}\text{C}$  and  $^{24}\text{Ne}$  decays



# Systematics $T_{1/2}$ : $^{14}\text{C}$ , $^{18,20}\text{O}$ , $^{23}\text{F}$ rad.

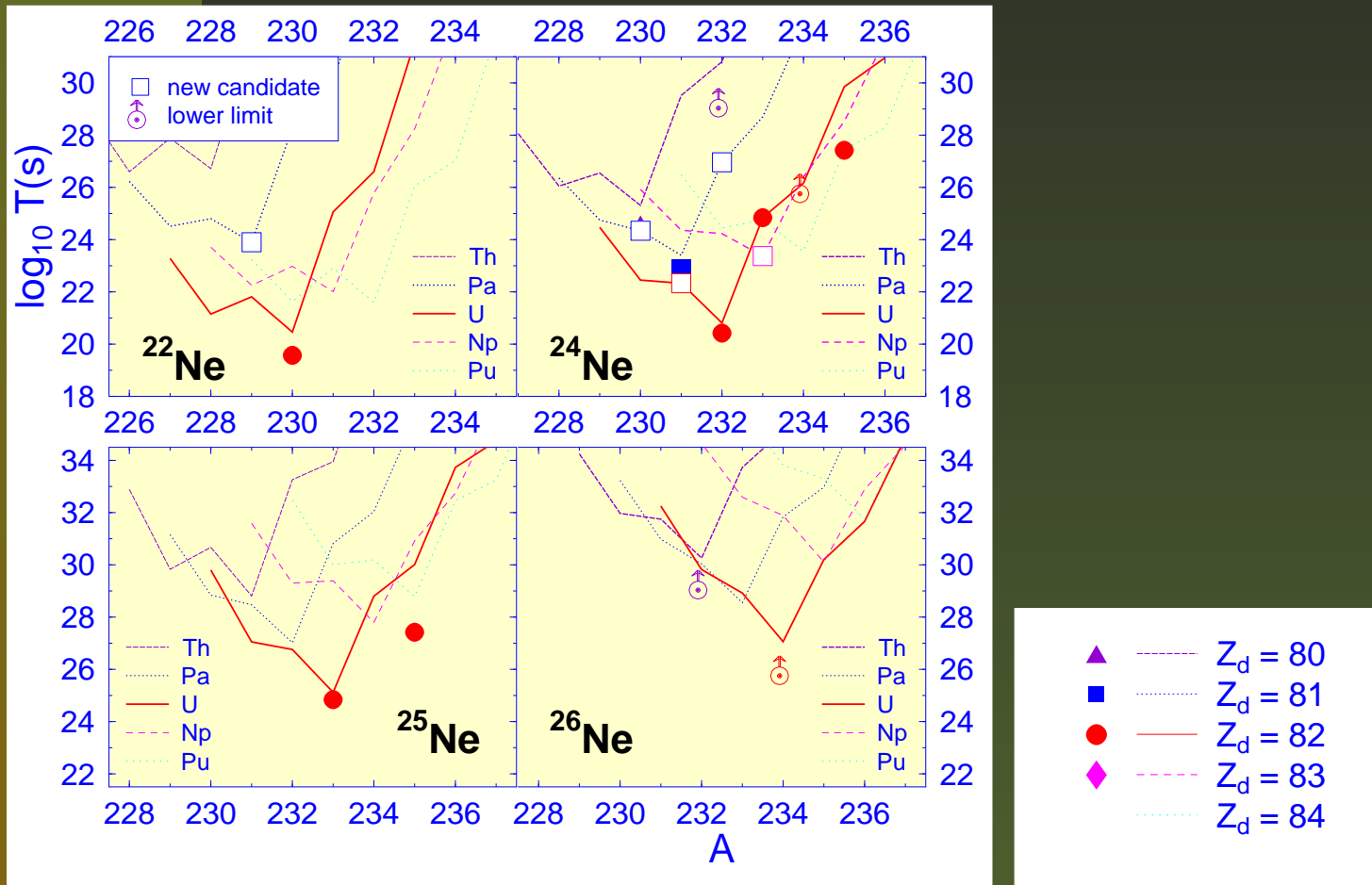


Calculated lines  
within ASAF model  
and exp. points



new confirm — A. Guglielmetti et al., J Phys: Conf Ser **111** (2008) 012050  
 One of the new candidates from our paper: Poenaru, Nagame, Gherghescu, W. Greiner *Phys. Rev. C* **65** (2002) 054308.

# Systematics $T_{1/2}$ : $^{22,24,25,26}\text{Ne}$ rad.

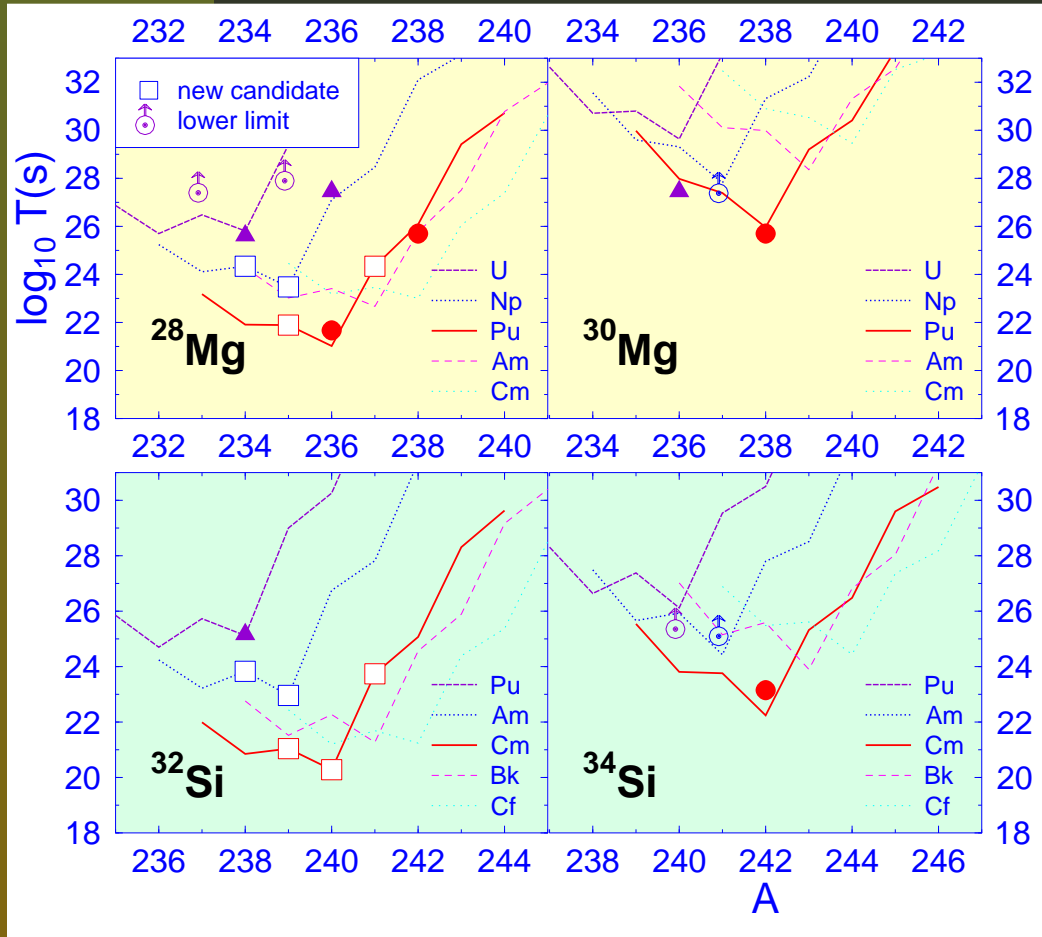


Only lower limits for  $^{18}\text{O}$  and  $^{26}\text{Ne}$



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# Systematics $T_{1/2}$ : $^{28,30}\text{Mg}$ , $^{32,34}\text{Si}$ rad.



Minima at  $N_d = 126$   
 Strong shell effect  
 Even-odd staggering



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# Universal curves (I)

Approximations:  $\log S = [(A_e - 1)/3] \log S_\alpha$ ,  
 $\nu(A_e, Z_e, A_d, Z_d) = \text{constant}$ . From fit to  $\alpha$  decay:  
 $S_\alpha = 0.0160694$  and  $\nu = 10^{22.01} \text{ s}^{-1}$ .

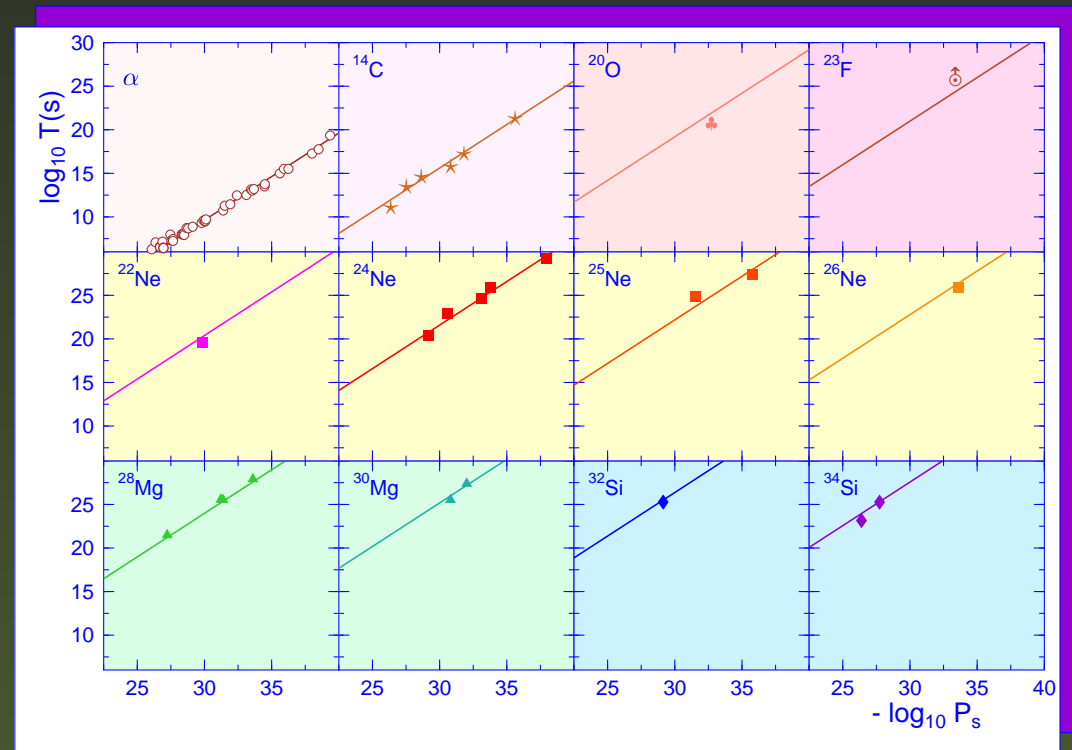
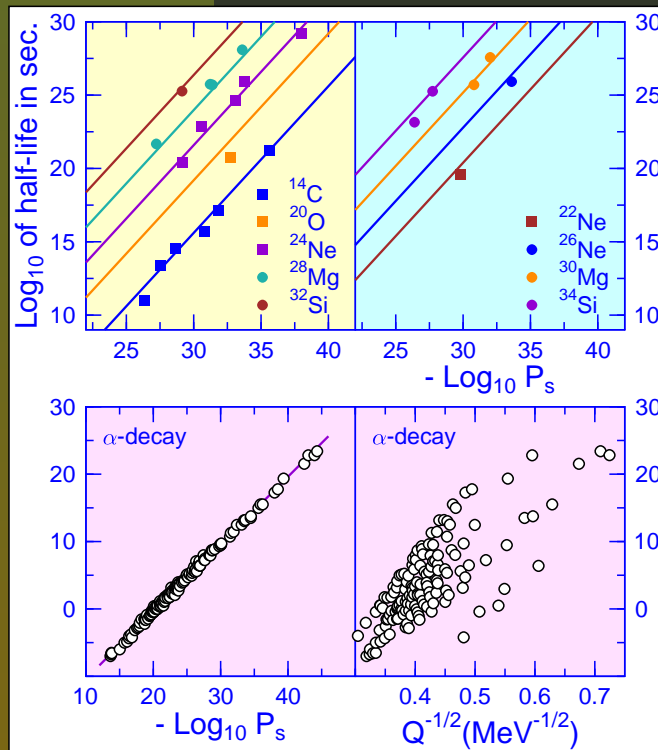
$$\log T = -\log P - 22.169 + 0.598(A_e - 1)$$

$$-\log P = c_{AZ} \left[ \arccos \sqrt{r} - \sqrt{r(1-r)} \right]$$
$$c_{AZ} = 0.22873(\mu_A Z_d Z_e R_b)^{1/2}, \quad r = R_t/R_b, \quad R_t =$$
$$1.2249(A_d^{1/3} + A_e^{1/3}), \quad R_b = 1.43998 Z_d Z_e / Q, \quad \text{and } \mu_A =$$
$$A_d A_e / A.$$

DN Poenaru, W Greiner, *Physica Scripta* **44** (1991) 427.



# Universal curves (II)



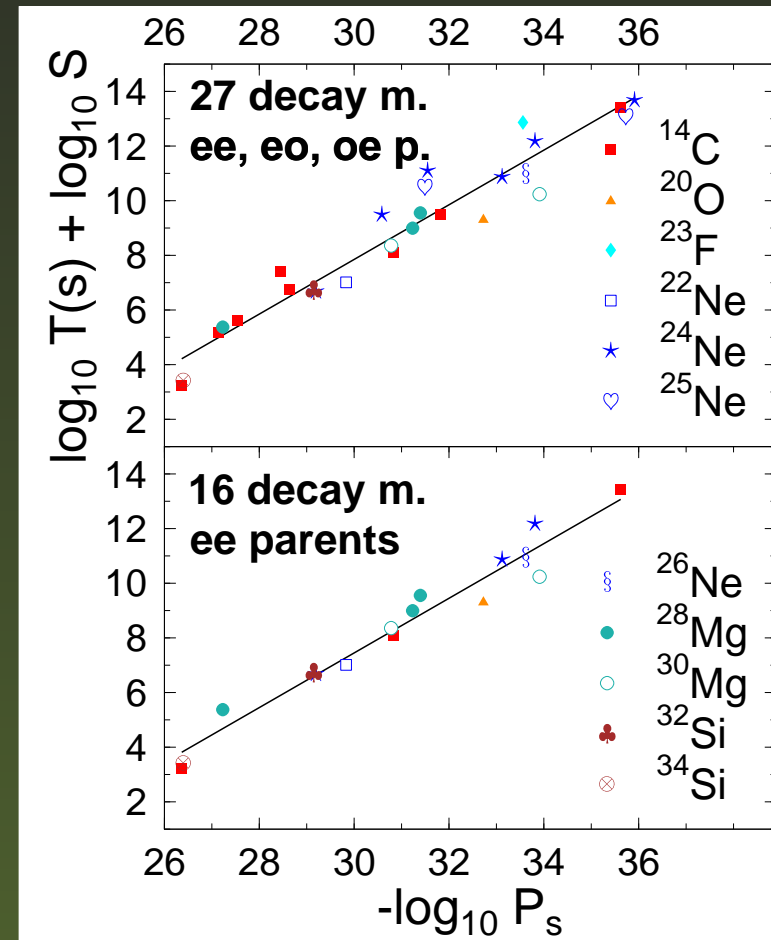
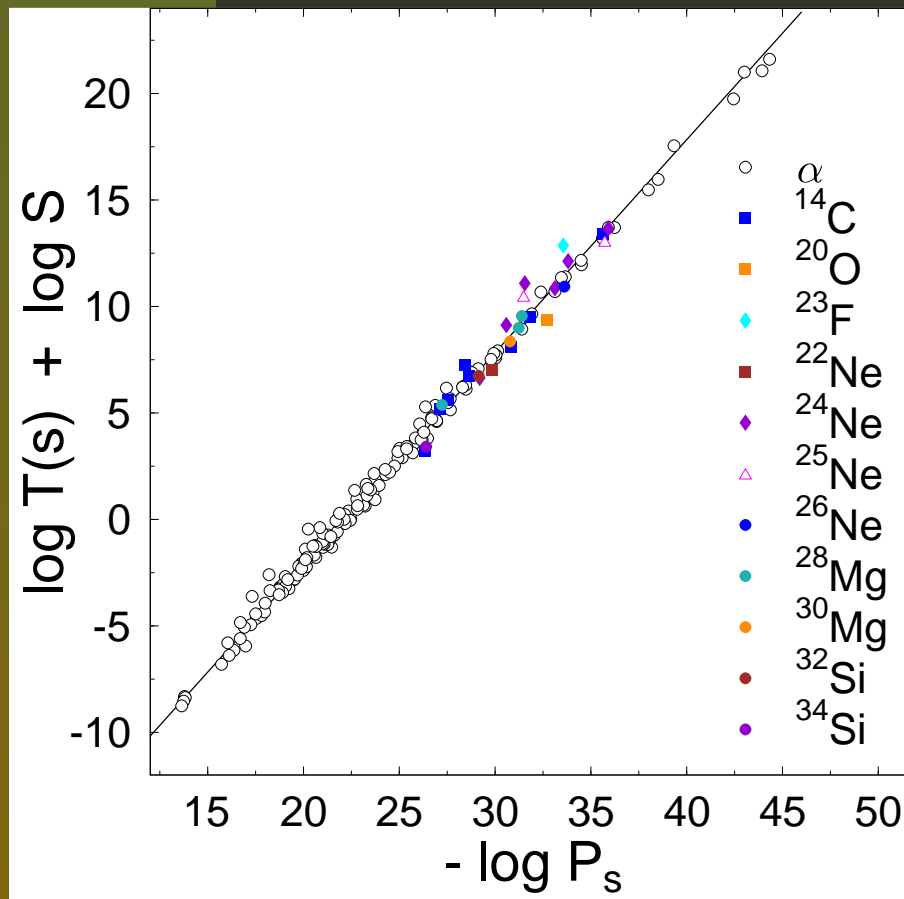
Geiger-Nuttal plot  $T_\alpha = f(\text{range of } \alpha \text{ in air})$

$\log T = f(1/Q^{-1/2})$



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# Single Universal curve: $\alpha$ and HIR

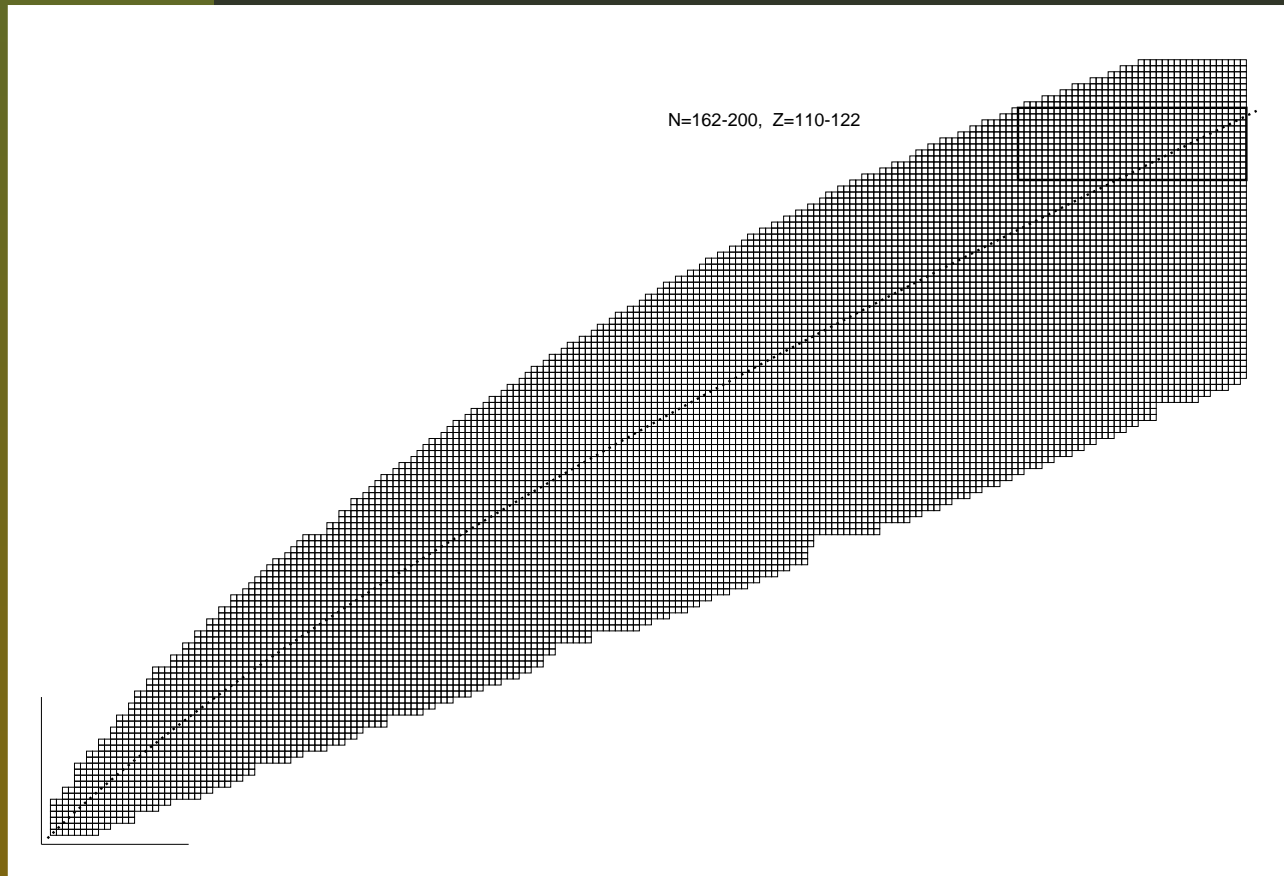


D.N. Poenaru, R.A. Gherghescu, W. Greiner, Phys. Rev. C, **83** (2011) 014601.



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# KTUY05 Calculated Masses

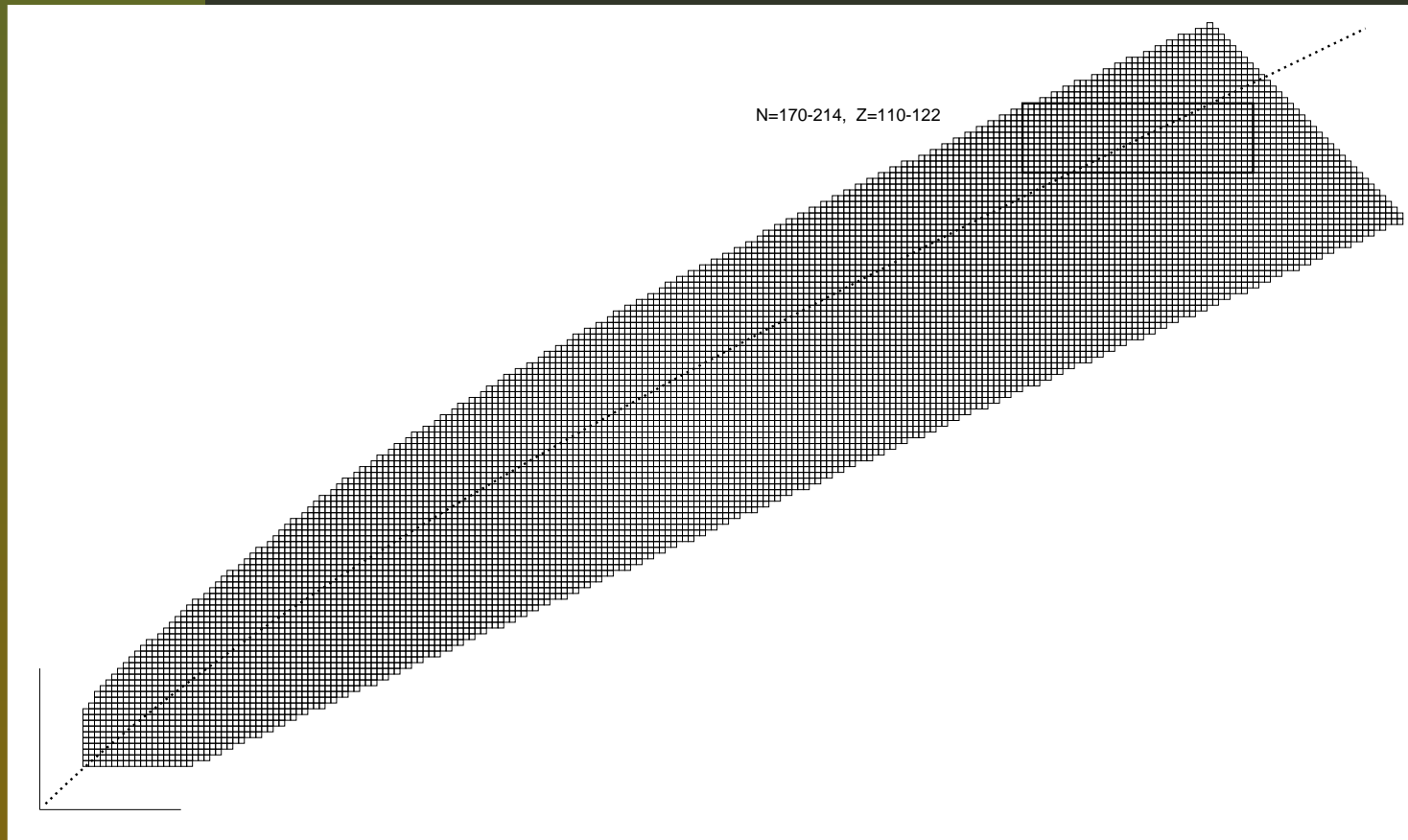


9441 nuclei with  $Z=2-130$  and  $N=2-200$ . H. Koura, T. Tachibana, M. Uno and M. Yamada, *Prog. Theor. Phys.* **113** (2005) 305.



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# FRDM95 Calculated Masses

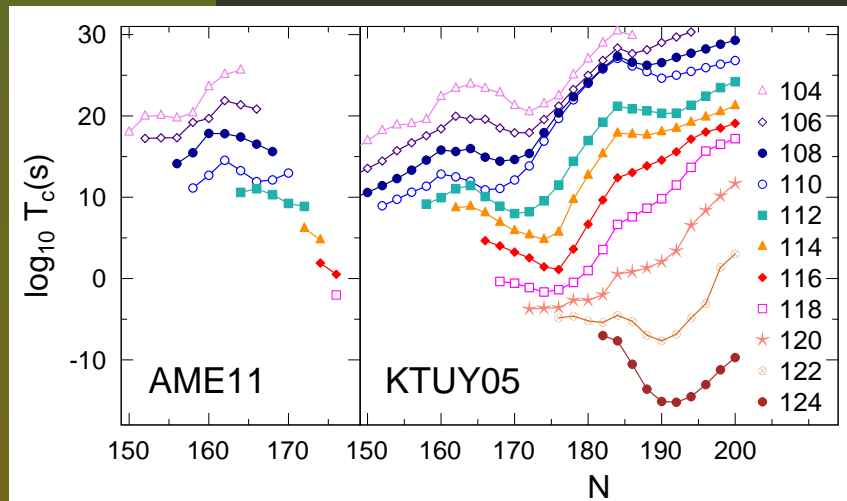


8979 nuclei with  $Z=8-136$  and  $N=8-236$ . P. Möller, J.R. Nix, W.D. Myers, W.J. Swiatecki, *At. Data Nucl. Data Tables* **59** (1995) 185.

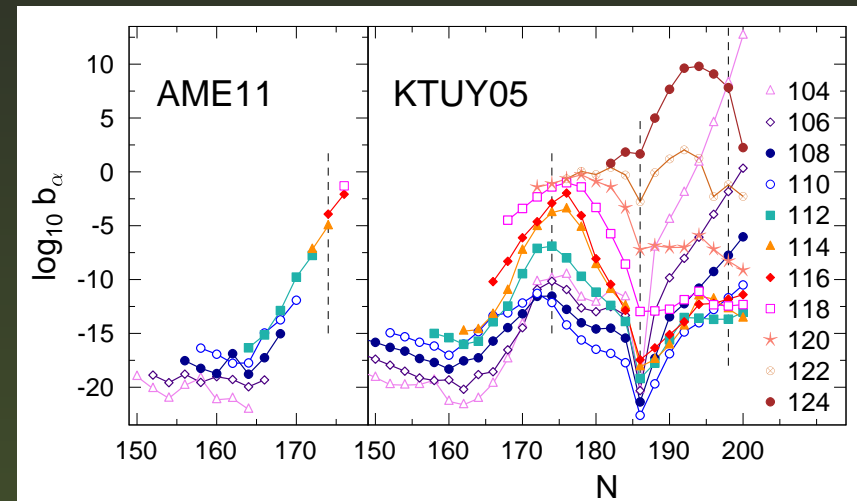


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# HIR of SH nuclei



Half-lives



Branching ratios

Branching ratio with respect to  $\alpha$  decay:  $b_\alpha = T_\alpha/T_c$ .

Usually  $b_\alpha \ll 1$ .

Trend: shorter  $T_c$  and larger  $b_\alpha$ .

For larger  $Z > 120$  there are SHs with  $T_c < 1$  ns and  $b_\alpha > 1$ .

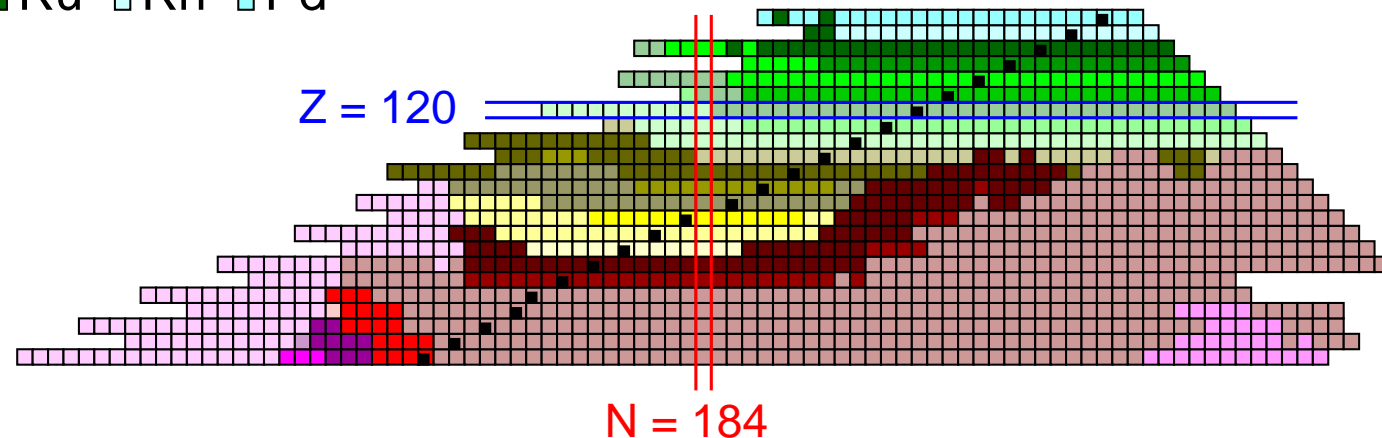


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# SH nuclei as cluster emitters

## EMITTED CLUSTERS

■ Be ■ Ar ■ Ca ■ Ti ■ V ■ Cr ■ Mn ■ Fe  
■ Ni ■ Cu ■ Zn ■ Ga ■ Ge ■ As ■ Se ■ Br  
■ Kr ■ Rb ■ Sr ■ Y ■ Zr ■ Nb ■ Mo ■ Tc  
■ Ru ■ Rh ■ Pd



FRDM95  $Z_e \leq Z - 80$  (freq. daughter around  $^{208}\text{Pb}$ )

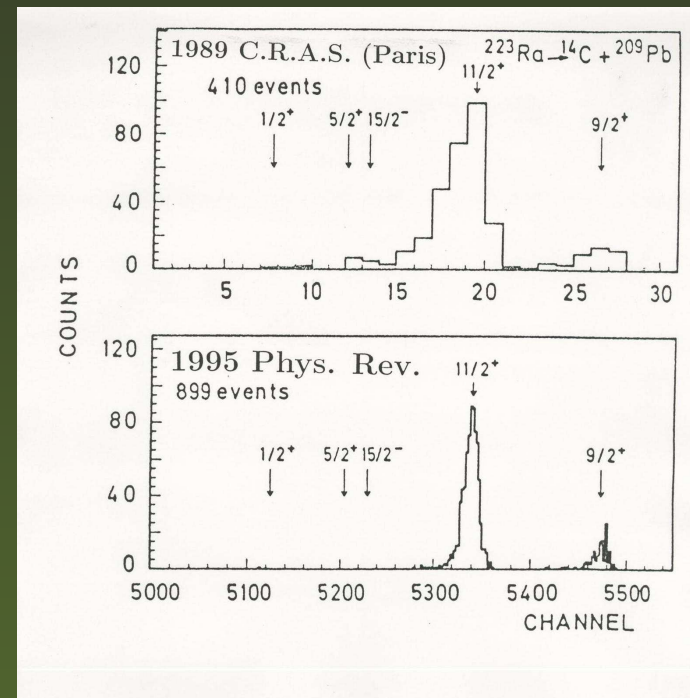
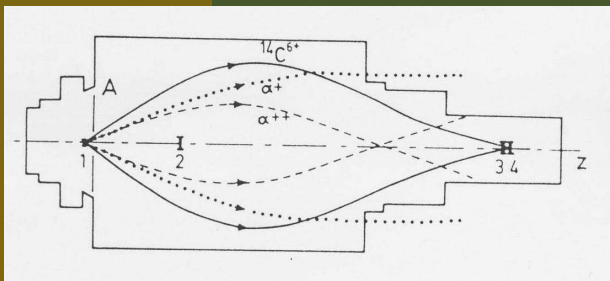
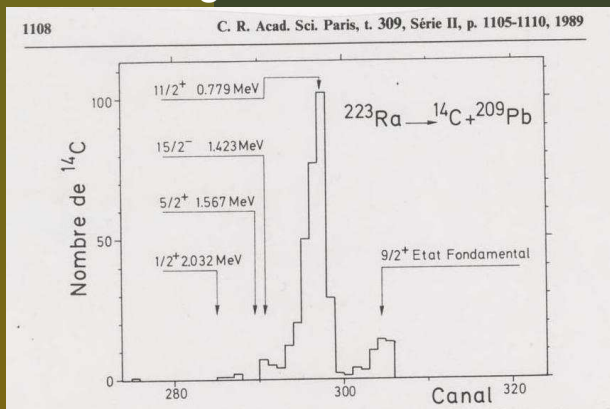
Most probable emitted clusters with different colors.



# Fine structure of $^{14}\text{C}$ radioactivity

Martin Greiner and Werner Scheid, Radioactive decay into excited states via heavy ion emission, *J. Phys. G: Nucl. Phys.* **12** (1986) L229.

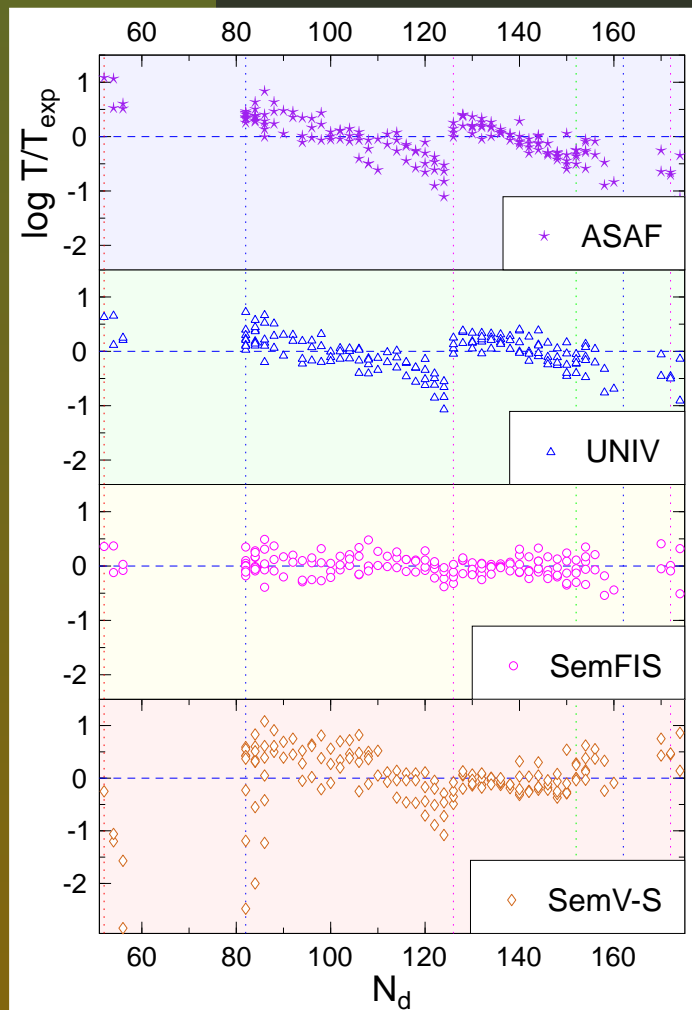
Experiments with SOLENO spectrometer at Orsay, France: E. Hourany, M. Hussonnois *et al.*, *C. R. Acad. Sci. Paris* **309** (1989) 1105. E. Hourany *et al.*, *Phys. Rev. C* **52** (1995) 267: the transition from the gs of  $^{223}\text{Ra}$  to the first excited state of the daughter  $^{209}\text{Pb}$  is stronger than that to its gs. A transition with an excited state of  $^{14}\text{C}$  was not observed.



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# $\alpha$ -decay: ASAF, semFIS & UNIV



## STANDARD DEVIATION

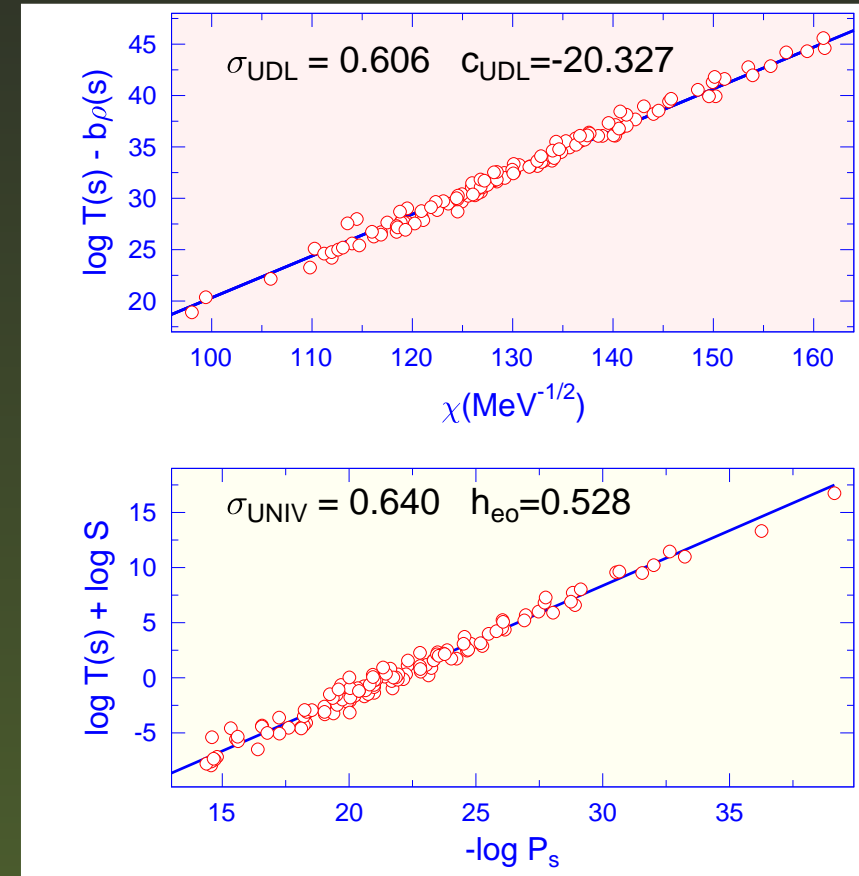
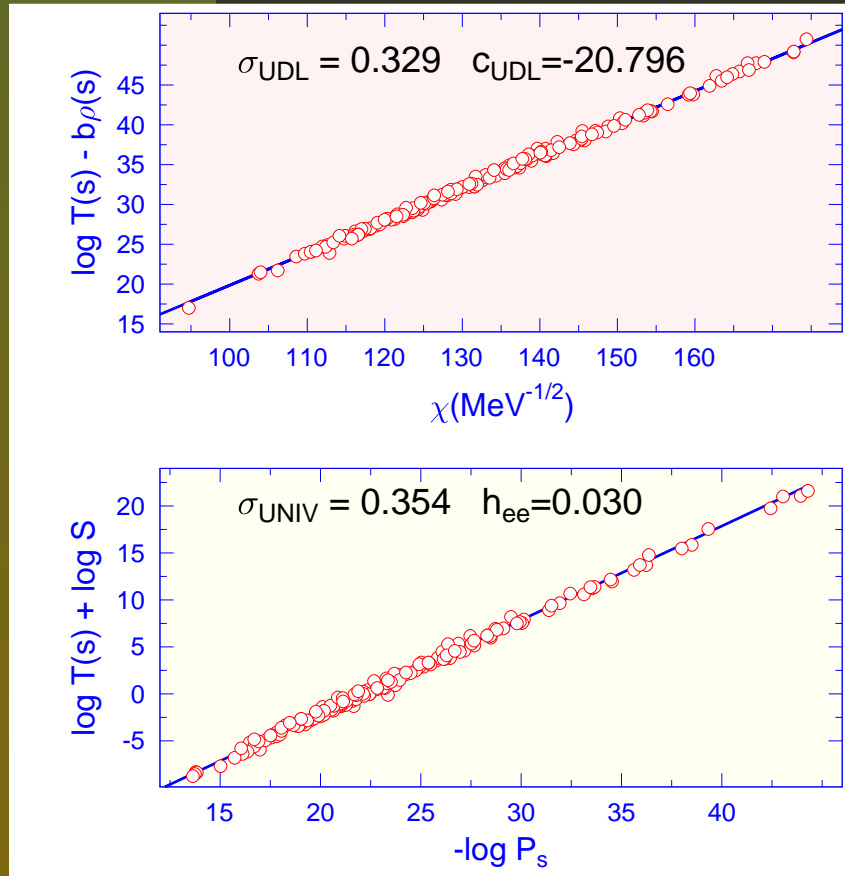
Group	$\sigma$ -ASAF	$\sigma$ -UNIV	$\sigma$ -semFIS
47 e-e	0.402	0.267	0.164
45 e-o	0.615	0.554	0.507
25 o-e	0.761	0.543	0.485
25 o-o	0.795	0.456	0.451

Poenaru, D.N., Plonski, I.H.,  
Gherghescu, R.A., Greiner, W.,  
*J. Phys. G* 32 (2006) 1223



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# UNIV and UDL curve for $\alpha$ $Z_{even}$



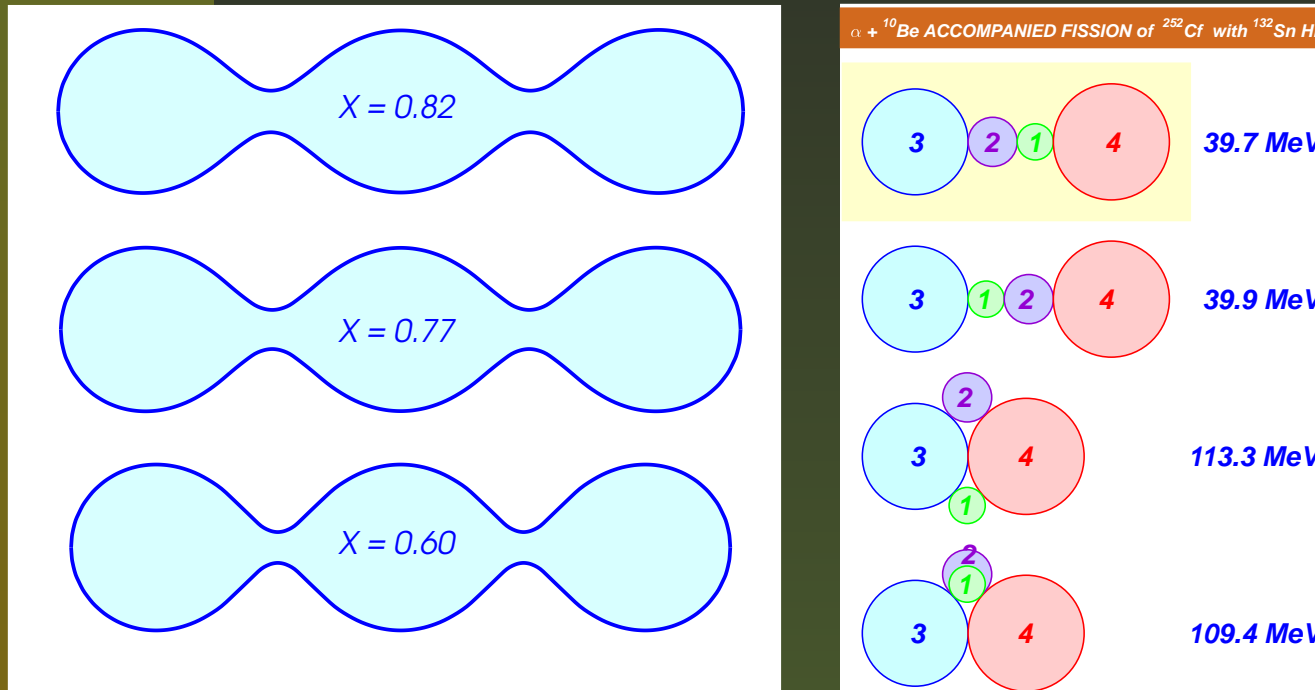
UDL: C. Qi, F.R. Xu, R.J. Liotta, R. Wyss Phys. Rev. Lett. **103** (2009) 072501.



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# Multicluster fission (I)

True-ternary and 2 particle-accompanied fission (quaternary)



D.N. Poenaru and W. Greiner, *J. Phys. G: Nucl. Part. Phys.* **25** (1999) L7

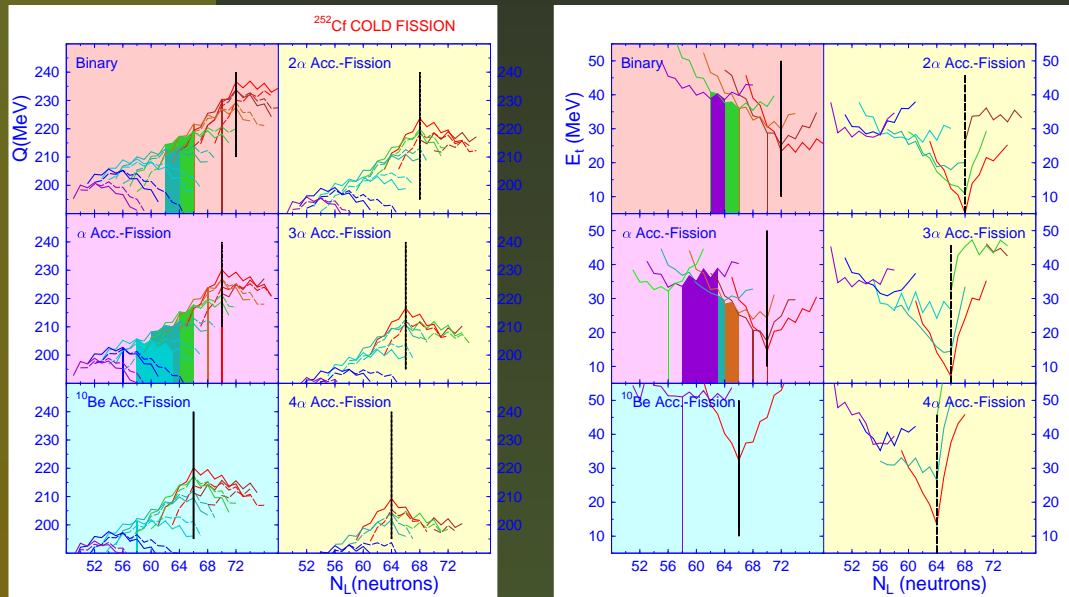
D.N. Poenaru, W. Greiner, J.H. Hamilton, A.V. Ramayya, E. Hourany and R.A. Gherghescu, *Phys. Rev. C* **59** (1999) 3457



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# Multicluster fission (II)

Good chance to be detected:  $2\alpha$ -,  $3\alpha$ -, and  $4\alpha$ -accompanied fission.  $Q$ -value and pot. barrier of  $2\alpha$ -accompanied fission is similar to  $^8\text{Be}$ -accompanied fission.



EXPERIMENTS: F. Gönnerwein, P. Jesinger, M. Mutterer, W.H. Trzaska, G. Petrov, A.M. Gagarski, V. Nesvizhevski and P. Geltenbort, *Heavy Ion Physics* **18** (2003) 419.  
F. Gönnerwein, M. Mutterer and Yu. Kopatch, *Europhysics News* **36** (2005) 11.  
Yu.V. Pyatkov, D.V. Kamanin, W.H. Trzaska, W. von Oertzen *et al.* *Rom. Rep. Phys.* **59** (2007) 569.



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# Applications: Atomic clusters

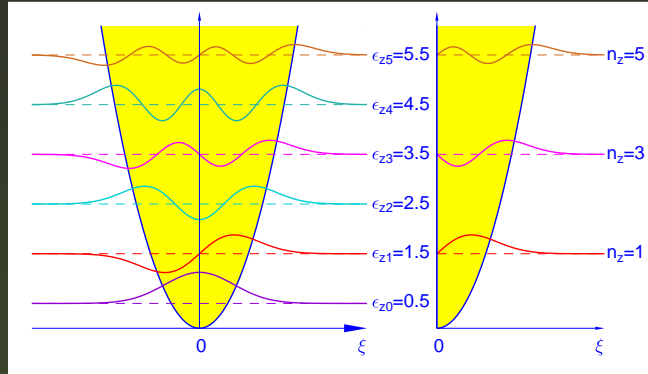
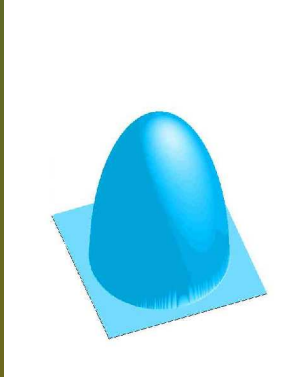
The delocalized conduction (valence) electrons of a metallic cluster form a Fermi liquid like the nucleons in an atomic nucleus, hence one can extend the MMM to atomic clusters.

## *Some publications*

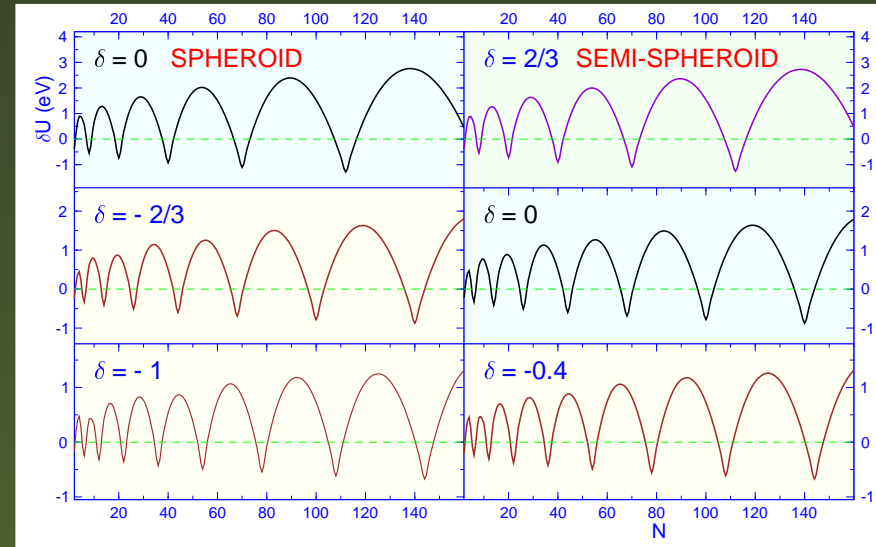
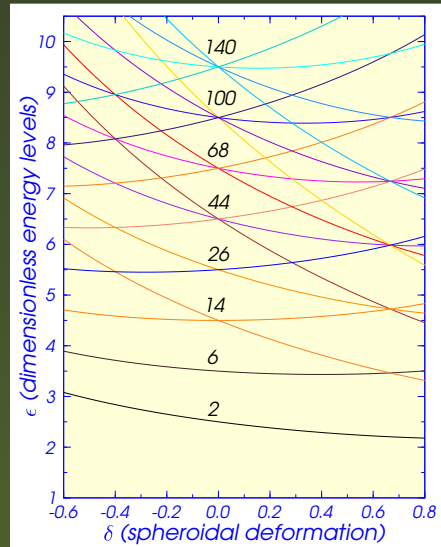
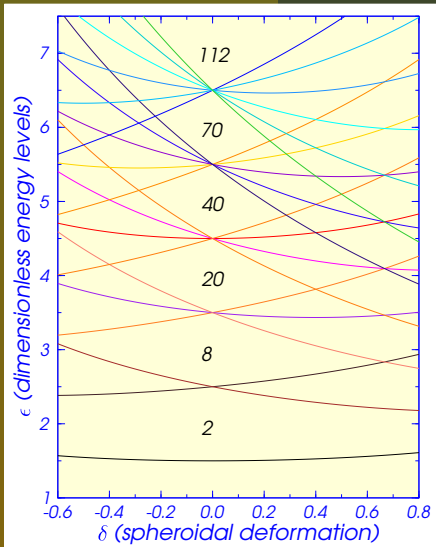
- D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner, *EPL* **79** (2007) 63001.
- D.N. Poenaru, R.A. Gherghescu, I.H. Plonski, A.V. Solov'yov, W. Greiner, *Europ. Phys. J. D* **47** (2008) 379-393. HIGHLIGHT PAPER.
- D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner, *Phys. Lett. A* **372** (2008) 5448-5451.
- R.A. Gherghescu, D.N. Poenaru, A.V. Solov'yov, W. Greiner, *Int. J. Mod. Phys. B* **22** (2008) 4917-4935.



# Hemispheroidal atomic cluster (I)



Hemispheroidal atomic cluster  
 $a^2 c = 1$  — volume conservation  
 $a = [(2 - \delta)/(2 + \delta)]^{1/3}$   
 New shell model with striking properties of symmetry.  
 Maximum degeneracy at  $\delta = 2/3$

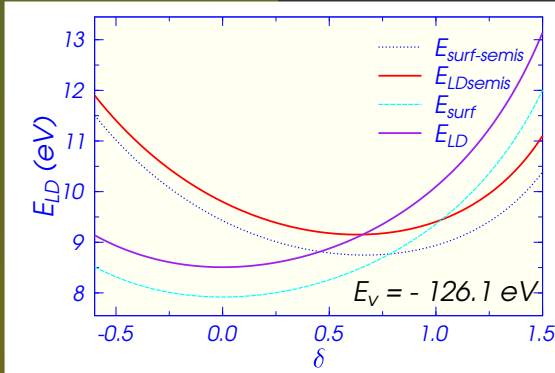


D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner, Phys. Lett. A 372 (2008) 5448;  
 EPL 79 (2007) 63001; 88 (2009) 23002

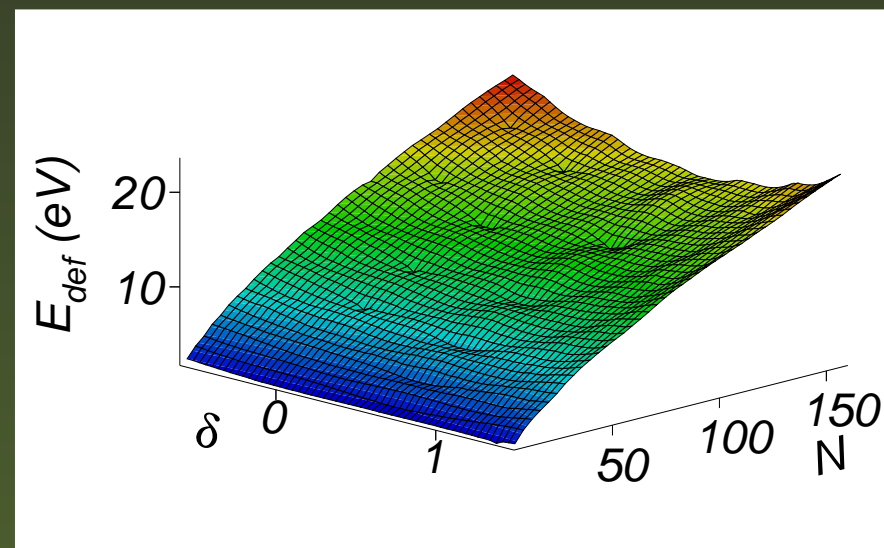
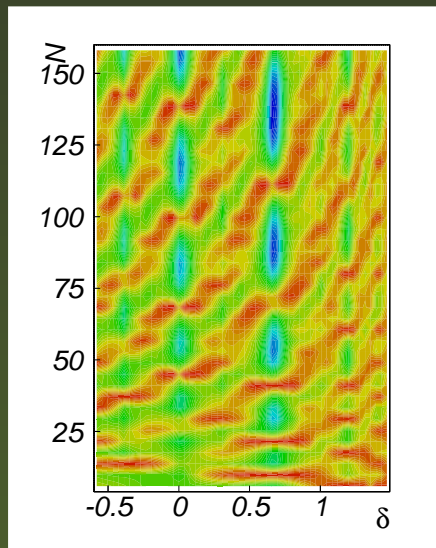
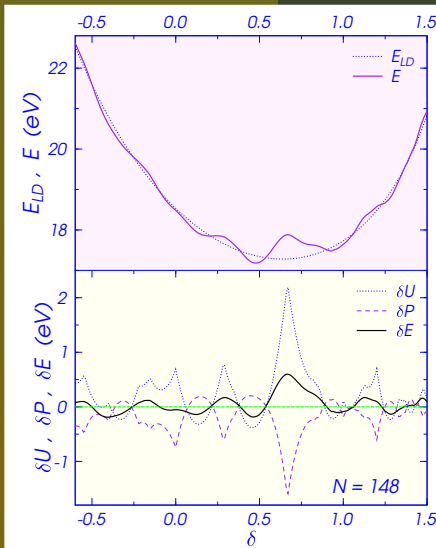


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# Hemispheroidal atomic cluster (II)



Figures, TOP: LDM (surface + curvature) energy of  $\text{Na}_{56}$  semi-spheroidal cluster compared to the spheroidal one. BOTTOM:  $\text{Na}_{148}$  cluster, pairing corrections, total deformation energy (LDM + shell and pairing corrections). Within LDM the most stable shape is a superdeformed prolate semi-spheroid ( $\delta \approx 0.66$ ).



D.N. Poenaru, R.A. Gherghescu, A.V. Solov'yov, W. Greiner, EPJD 47 (2008) 379 → HIGHLIGHT PAPER; J. Phys. G: Nucl. Part. Phys. 36 (2009) 125101; 37 (2010) 085101; Nucl. Phys. A 834 (2010) 163c; Int. J. Mod. Phys. B 24 (2010) 3411



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# Liquid drop model of a charged cluster

$M_N^{z+}$  will have  $n_e = N - z$  delocalized electrons. Deformation energy

$$E_{LDM} = E - E^0 = (E_s - E_s^0) + (E_C - E_C^0) = E_s^0(B_s - 1) + E_C^0(B_C - 1)$$

Spherical shapes:  $E_s^0 = 4\pi R_0^2 \sigma = a_s n_e^{2/3} = 4\pi r_s^2 n_e^{2/3}$  ;

$E_{C-metal}^0 = z^2 e^2 / (2R_0) = z^2 e^2 / (2r_s n_e^{1/3})$  for a surface distrib. of charge. The ratio to  $E_C^0 = 3z^2 e^2 / (5R_0)$  for bulk homog. charge distr. is 5/6, i.e. 17 % smaller.

$\sigma$  – surface tension.  $r_s$  – Wigner-Seitz radius.

$$Fissility \ X = \frac{E_c^0}{2E_s^0} = \frac{e^2}{16\pi r_s^3 \sigma} \frac{z^2}{n_e} < 1 ; \quad n_e > n_c = \frac{e^2 z^2}{16\pi r_s^2 \sigma}$$

Within LDM VERY LIGHT CHARGED ATOMIC CLUSTERS ARE UNSTABLE.

For nuclear fission  $X = E_c^0 / (2E_s^0) \simeq [3e^2 / (10r_0 a_s)] (Z^2 / A) < 1$  leading to  $Z^2 / A < (Z^2 / A)_c \simeq 10r_0 a_s / (3e^2)$  SUPERHEAVY NUCLEI ARE UNSTABLE.

Tables of material properties ( $r_s$ ,  $\sigma$  or  $a_s$ ,  $a_v$ , etc):

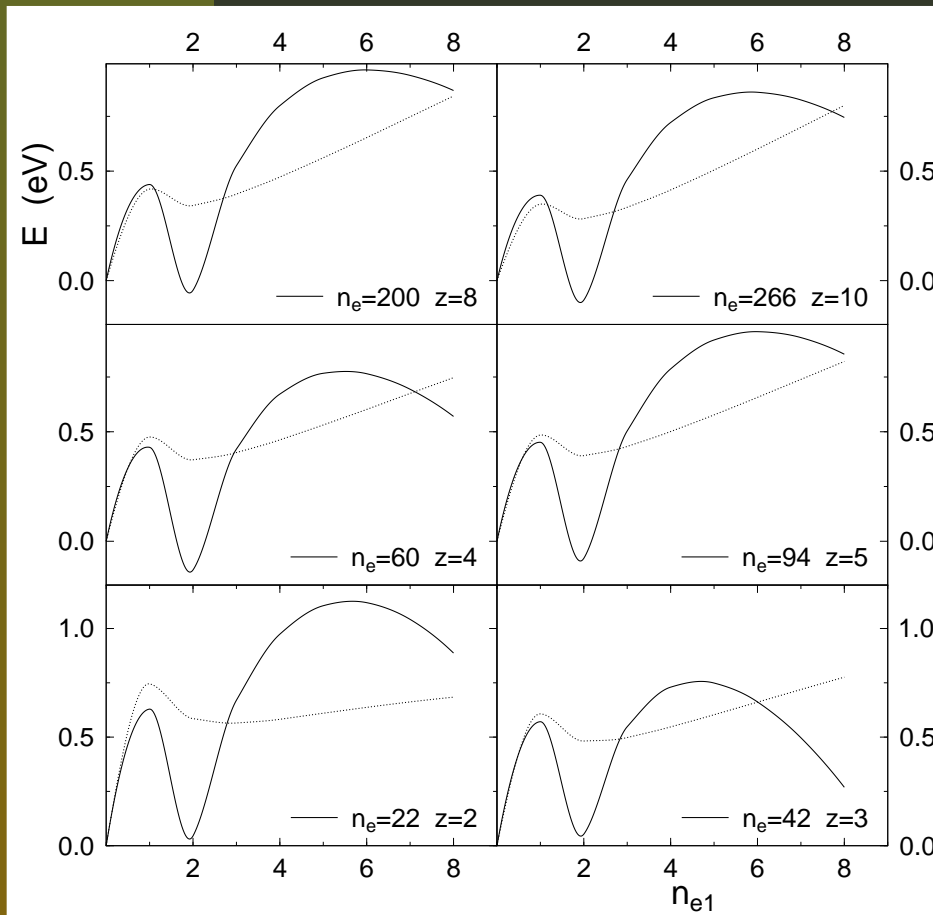
J.P. Perdew, Y. Wang, E. Engel, *Phys. Rev. Lett.* **66** (1991) 508.

U. Näher, S. Bjørnholm, S. Frauendorf, F. Garcias, C. Guet, *Phys. Rep.* **285** (1997) 245.





# Cs as an Ideal Emitter of a trimer $M_3^+$

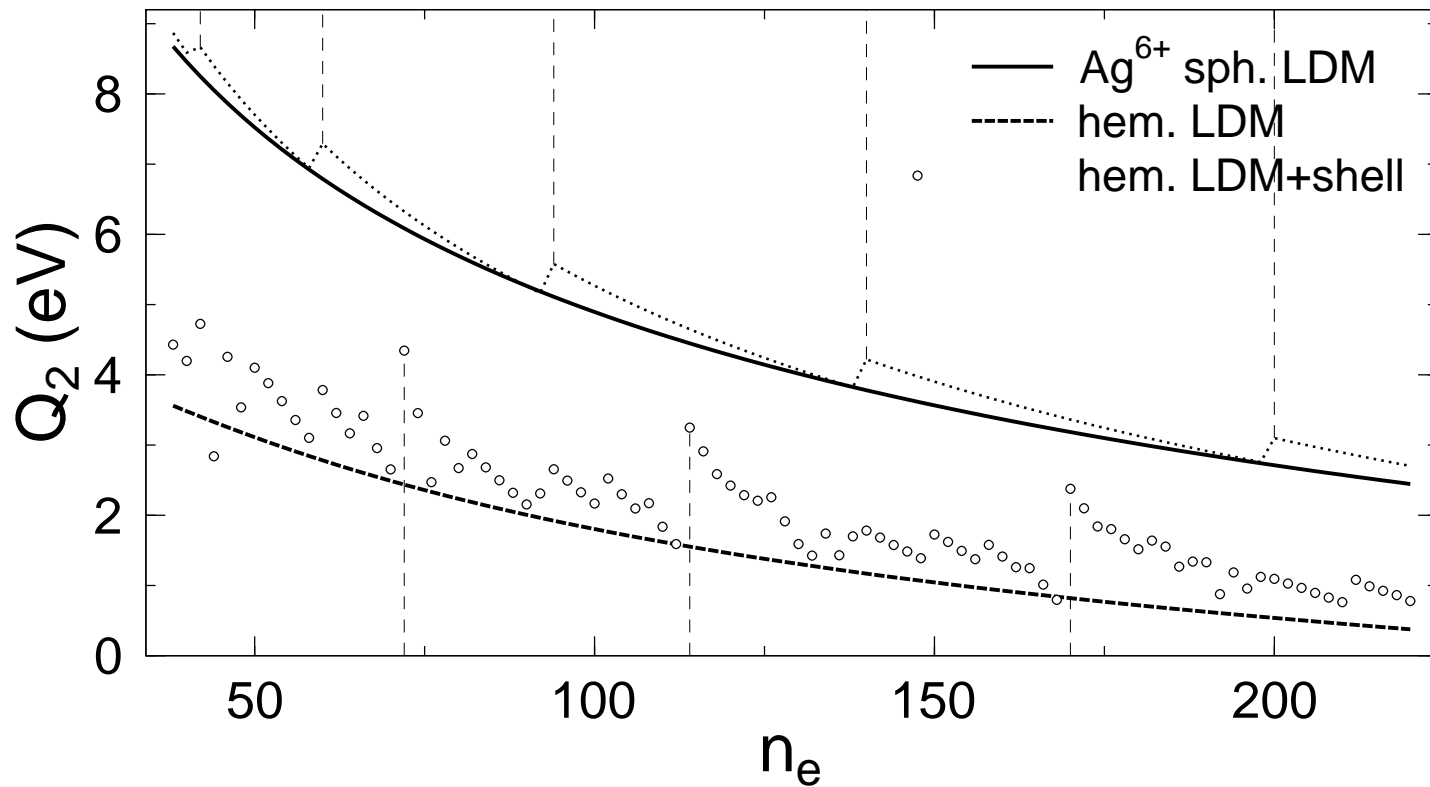


Details of the large asymmetry part of scission point deformation energies  $E_{LDM}$  (dotted line) and  $E_{LDM} + \delta E$  (full line) for different values  $n_e$ . Both  $E_{LDM}$  and  $\delta E$  have minima at  $n_e = 2$ .



$\text{Ag}^{6+}_{n_e+6}$

# Spheres and Hemispheroids



Max.  $Q$ -value for two fragments with magic numbers of electrons  $n_e = 2 + n_{d-magic}$ . For spherical shape  $n_{d-magic} = 40, 58, 92, 136, 198$ .

For a superdeformed hemispheroid  $n_{d-magic} = 70, 112, 168$ .



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# Summary

- ASAF model predictions have been confirmed for parent nuclei with  $Z = 87 - 96$
- The magicity of the daughter  $^{208}\text{Pb}$  was not fully exploited: new experimental searches can be performed
- The ASAF, semFIS and universal curves UNIV and UDL provide good estimation of half-lives
- For some superheavies HIR half-lives could be shorter than that of  $\alpha$  decay
- One can obtain saddle-point shapes by solving an integro-differential equation
- Experimental evidence for multicluster fission:  $2\alpha$ -accompanied fission and collinear cluster tri-partition
- Nanophysics of atomic clusters
  - The maximum degeneracy of the new hemispheroidal shell model occurs at a superdeformed prolate semi-spheroidal shape
  - LDM equilibrium shapes: superdeformed prolate hemispheroid; oblate shapes obtained by simulating the interaction with the substrate
  - Charged clusters are ideal “alpha”(singly ionized trimer fragment) emitters

