Warsaw macro-micro model and random walk method for calculating the fusion probability of superheavy elements **Aleksander Augustyn** Michał Kowal, Tomasz Cap, Krystyna Siwek-Wilczyńska



CENTRE FOR NUCLEAR RESEARCH ŚWIERK

Graduate Physics Seminar 26 October 2023

Superheavy elements

- Only man-made
- Z>103 (transactinides)
- Produced in nuclear reactions:
 - Cold fusion
 - Hot fusion

H He hydrogen 1.0080 ±0.0002 helium 4.0026 16 17 2 Key: 13 14 15 ±0.0001 atomic number 5 7 9 10 3 4 6 8 Ě Li Be в Ċ Ν ο Ne Symbol carbon 12.011 ± 0.002 nitrogen 14.007 ± 0.001 neon 20.180 ± 0.001 lithium beryllium name 10.81 ± 0.02 0xygen 15.999 ± 0.001 fluorine abridged standard atomic weight 9.0122 ± 0.0001 18.998 ± 0.001 6.94 ±0.06 11 12 13 14 15 16 17 18 Ρ Mg 1005 1002 AI Si S CI Na Ar sodium aluminium silicon phosphorus sulfur chlorine argon 39.95 ± 0.16 32.06 ± 0.02 22.990 ±0.001 26.982 ± 0.001 28.085 ± 0.001 30.974 ± 0.001 35.45 ±0.01 3 4 6 10 11 12 19 K 21 22 Ti 23 V 24 25 26 27 28 29 30 32 33 34 35 36 20 31 Ča Ĉr М'n Ñi Ğa Se Br Кř Ge Sc Fe Co Cu Zn As potassium 39.098 ±0.001 calcium 40.078 ± 0.004 scandium titanium 47.867 ±0.001 vanadium chromium manganese 54.938 ±0.001 iron 55.845 ± 0.002 cobalt 58.933 ±0.001 nickel 58.693 ± 0.001 copper 63.546 ± 0.003 zinc 65.38 ± 0.02 gallium 69.723 ± 0.001 germanium 72.630 ± 0.008 arsenic 74.922 ± 0.001 selenium 78.971 ± 0.008 79.904 ± 0.003 krypton 83.798 ± 0.002 44.956 ± 0.001 50.942 ± 0.001 51.996 ± 0.001 47 49 54 37 38 39 **Y** 40 41 42 43 44 45 46 48 50 51 52 53 Rb Sr Zr Nb Мо Tc Ru Rh Pd Ag silver 107.87 ± 0.01 Cd In Sn Sb Te Хе rubidium strontium yttrium 88.906 ±0.001 zirconium niobium olybdenur technetium ruthenium rhodium palladium cadmium indium tin 118.71 ± 0.01 antimony 121.76 ± 0.01 tellurium iodine 126.90 ± 0.01 xenon 131.29 ± 0.01 85.468 ± 0.001 91.224 ±0.002 92.905 ± 0.001 95.95 ± 0.01 101.07 ± 0.02 106.42 ± 0.01 112.41 ±0.01 127.60 ± 0.03 87.62 ± 0.01 102.91 ±0.01 114.82 ± 0.01 [97] 55 56 57-71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 Hg mercury 200.59 ± 0.01 Hf ΤI Pb Bi w Cs Ва Та Re Os Ir Pt Au Po At Rn lanthanoids tantalum 180.95 ± 0.01 rhenium 186.21 osmium 190.23 ± 0.03 platinum 195.08 ± 0.02 thallium 204.38 ± 0.01 132.91 ± 0.01 barium 137.33 ± 0.01 hafnium 183.84 ± 0.01 iridium 192,22 gold 196.97 ± 0.01 lead 207.2 # 1.1 208.98 ± 0.01 polonium astatine radion 178.49 ±0.01 [209] +0.01 + 0.01 [210] [222] 87 114 116 117 118 88 89-103 104 105 106 107 108 109 110 111 112 113 115 Rf Rg FI Fr Ra Db Sg seaborgium Bh Hs Mt Ds Cn Nh Mc Ts Og Lv actinoids dubnium bohrium armstadtium flerovium francium radium utherfordium hassium meitnerium roentgeniun operniciur nihonium moscovium ivermorium tennessine oganessor [223] 12261

IUPAC Periodic Table of the Elements



INTERNATIONAL UNION OF PURE AND APPLIED CHEMISTRY

la	57 La anthanum 138.91 ± 0.01	58 Ce cerium 140.12 ± 0.01	59 Pr praseodymium 140.91 ±0.01	60 Nd neodymium 144.24 ±0.01	61 Pm promethium (145)	62 Sm samarium 150.36 ± 0.02	63 Eu europium 151.96 ± 0.01	64 Gd gadolinium 157.25 ± 0.03	65 Tb terbium 158.93 ± 0.01	66 Dy dysprosium 162.50 ± 0.01	67 Ho holmium 164.93 ±0.01	68 Er erbium 167.26 ± 0.01	69 Tm thulium 168.93 ± 0.01	70 Yb ytterbium 173.05 ± 0.02	71 Lu Iutetium 174.97 ± 0.01
	89 Ac actinium [227]	90 Th thorium 232.04 ± 0.01	91 Pa protactinium 231.04 ±0.01	92 U uranium 238.03 ±0.01	93 Np neptunium garj	94 Pu plutonium [244]	95 Am americium [243]	96 Cm curium paŋ	97 Bk berkelium [247]	98 Cf californium [251]	99 Es einsteinium [252]	100 Fm fermium (257)	101 Md mendelevium [258]	102 No nobelium	103 Lr Iawrencium [262]

For notes and updates to this table, see www.iupac.org. This version is dated 4 May 2022. Copyright © 2022 IUPAC, the International Union of Pure and Applied Chemistry.

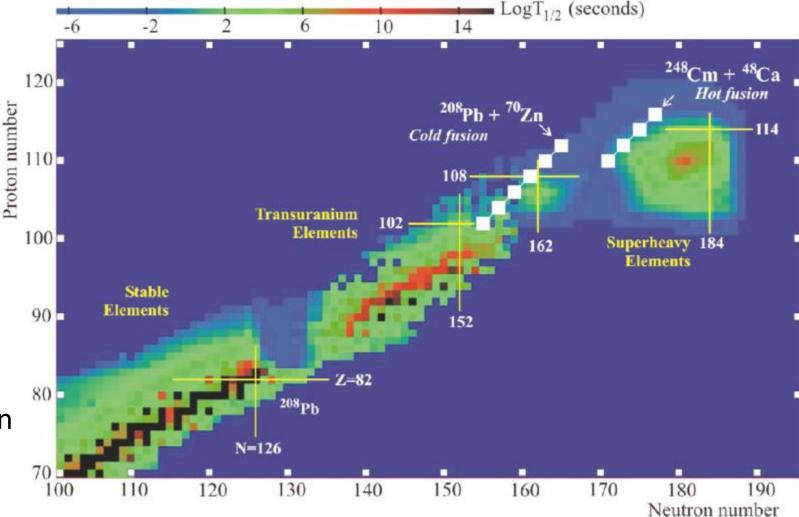
18



Superheavy elements

- Only man-made
- Z>103 (transactinides)
- Produced in nuclear reactions: ¹¹⁰
 - Cold fusion
 - Hot fusion
- Expensive process:
 - Target/Projectile production
 - Length of irradiation

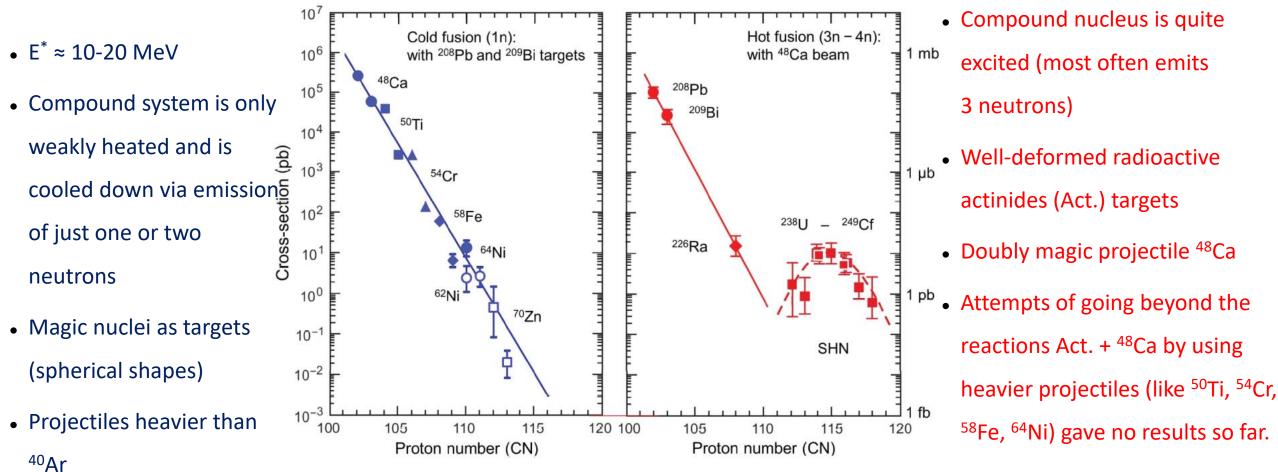
Oganessian, Yu. (2006). Synthesis and decay properties of superheavy elements. Pure and Applied Chemistry - PURE APPL CHEM. 78. 889-904. 10.1351/pac200678050889.





Cold and hot fusion

• E* = 30-40 MeV



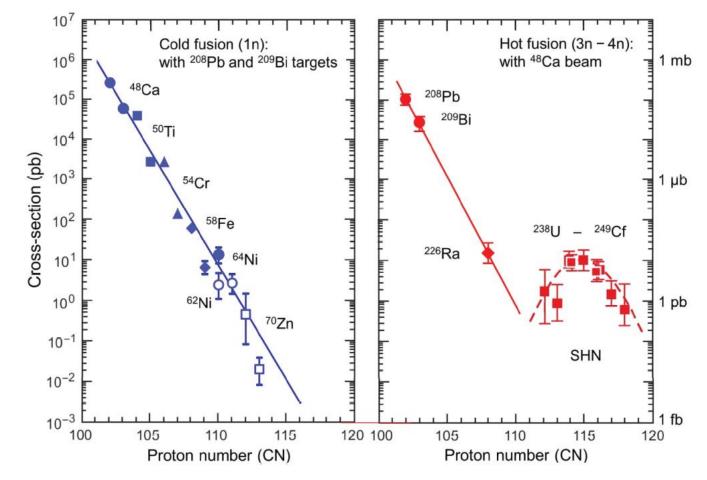
Sigurd Hofmann, Sergey N. Dmitriev, Claes Fahlander, Jacklyn M. Gates, James B. Roberto and Hideyuki Sakai Report of the 2017 Joint Working Group of IUPAC and IUPAP, Pure Appl. Chem. 2020; 92(9): 1387–1446

 Heavier actinides with Z>98 too short-lived to be used as targets



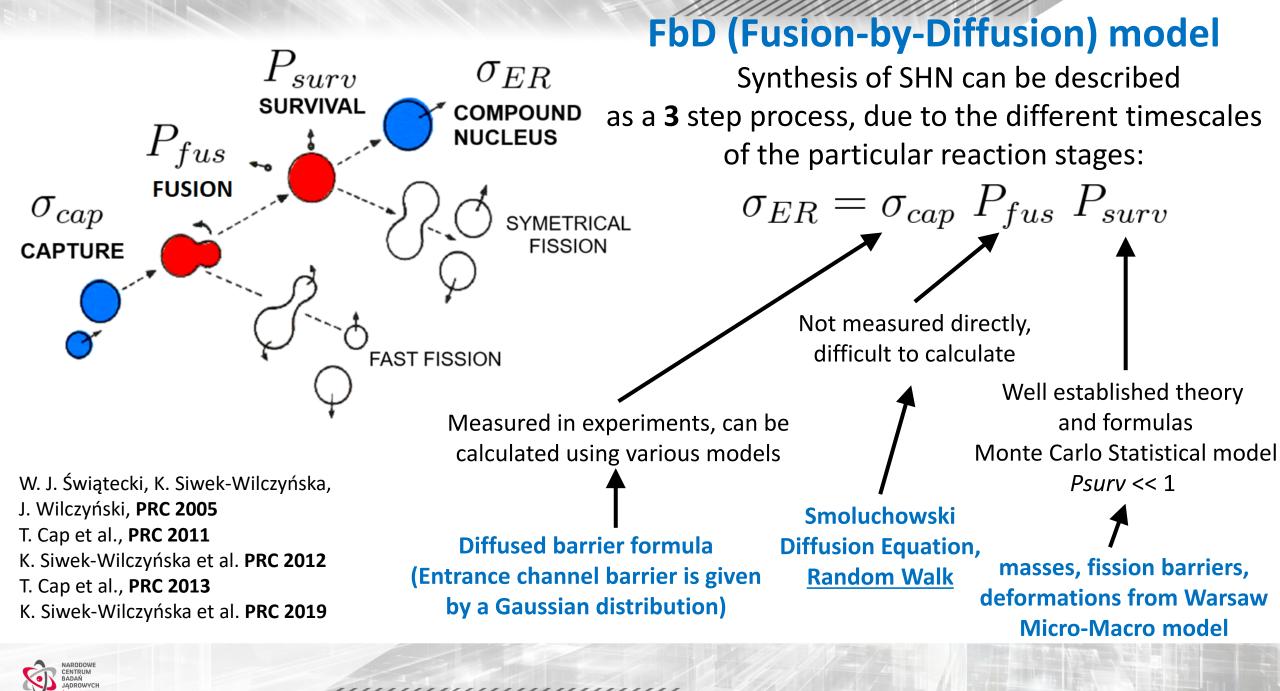
Motivation

- Experiments require theory to determine the optimal reactions and bombarding energies
- A way to calculate P_{fus} would be very helpful in the search for the new elements 119 and 120
- We wanted to use the micro-macro model with the inclusion of rotational energy and a random walk method on potential energy surfaces (PES) to calculate the probability of fusion, while describing the fusion process
- The model is first tested on cold fusion reactions with near spherical projectiles: ⁴⁸Ca+²⁰⁸Pb, ⁵⁰Ti+²⁰⁸Pb and ⁵⁴Cr+²⁰⁸Pb



Sigurd Hofmann, Sergey N. Dmitriev, Claes Fahlander, Jacklyn M. Gates, James B. Roberto and Hideyuki Sakai Report of the 2017 Joint Working Group of IUPAC and IUPAP, Pure Appl. Chem. 2020; 92(9): 1387–1446

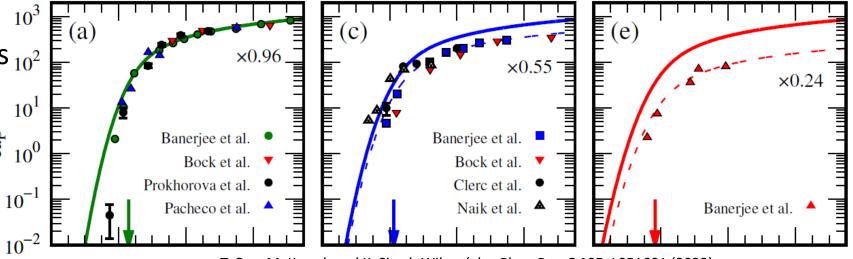




Capture cross section σ_{cap}

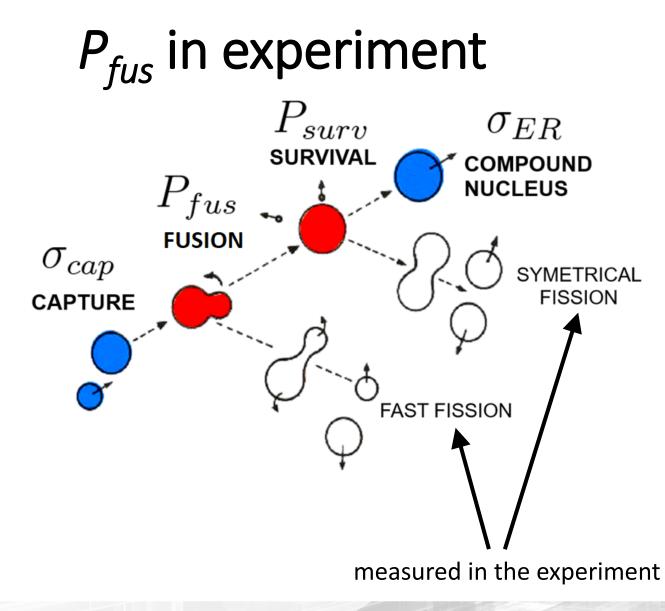
- The entrance channel barrier is described by a distribution that can be approximated by a Gaussian function
- The formula for the capture cross section is derived by folding the Gaussian barrier distribution with the classical 10^3 expression for the fusion cross 10^2 section $(10^3 - 10^3)$

$$\sigma_{cap} = \pi R^2 \frac{\omega}{E_{c.m.}\sqrt{2\pi}} \Big[X\sqrt{\pi}(1 + \operatorname{erf}(X)) + \exp(-X^2) \Big] = \pi \lambda^2 (2l_{max} + 1)^2, \text{ where } X = \frac{E_{c.m.} - B_0}{\omega\sqrt{2}}$$

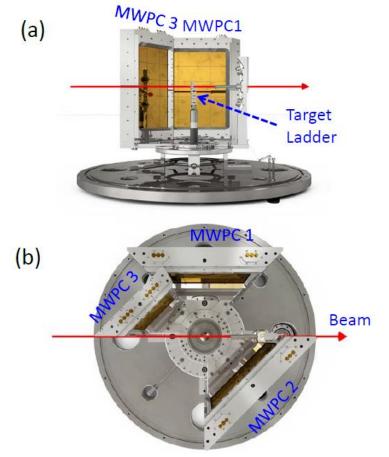


T. Cap, M. Kowal, and K. Siwek-Wilczyńska, Phys. Rev. C 105, L051601 (2022)

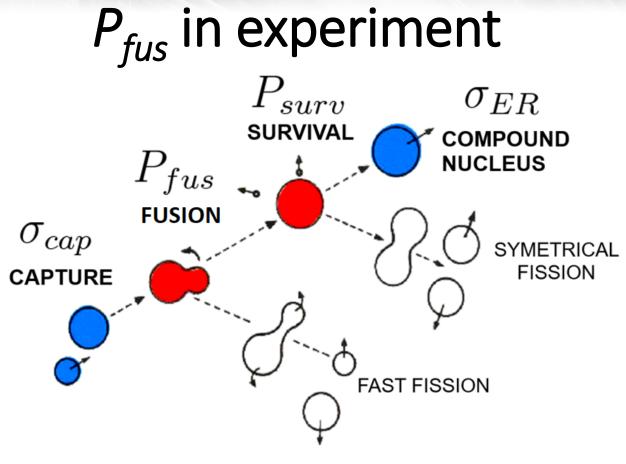




Mechanisms Suppressing Superheavy Element Yields in Cold Fusion Reactions Banerjee *et al.*, PRL 122, 232503 (2019)



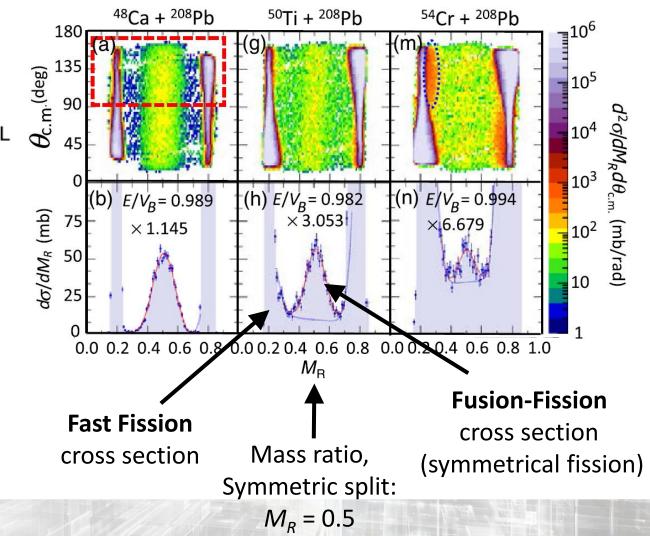




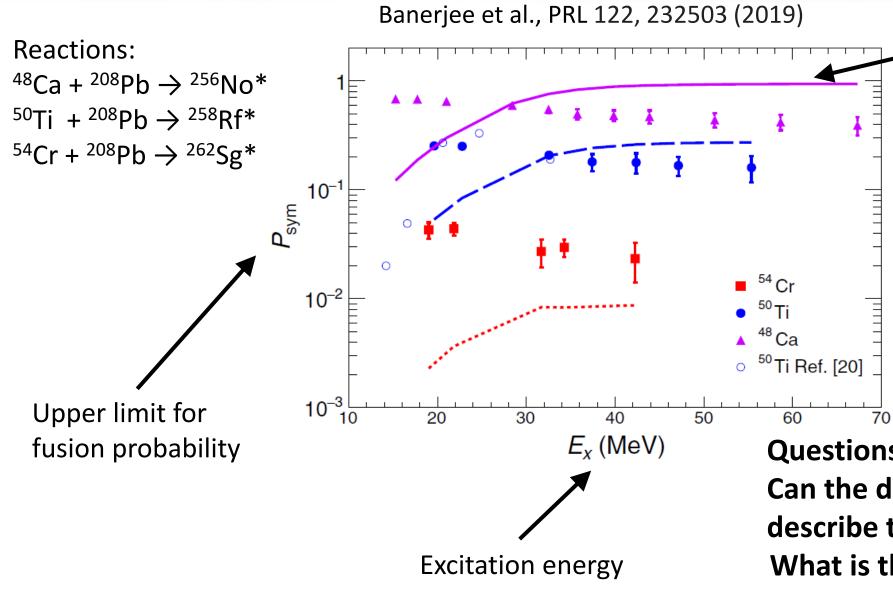
 P_{fus} can be experimentally estimated:



Mechanisms Suppressing Superheavy Element Yields in Cold Fusion Reactions Banerjee *et al.*, PRL 122, 232503 (2019)







Diffusion model calculations by V. Zagrebaev and W. Greiner PRC 78, 034610 (2008).

The experimental trends are different than the model predictions for all 3 reactions.

The conclusion was that diffusion is not the main mechanism responsible for the synthesis of SHN.

Questions:

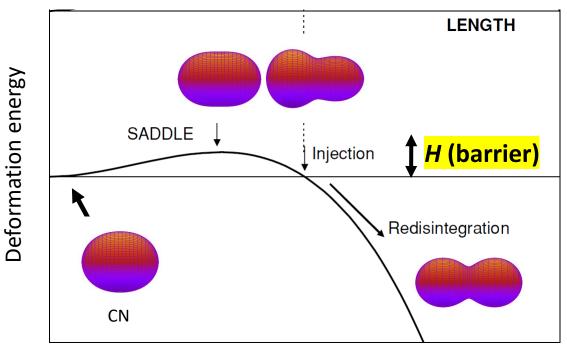
Can the diffusion approach (FbD) describe the experimental results? What is the fusion mechanism?



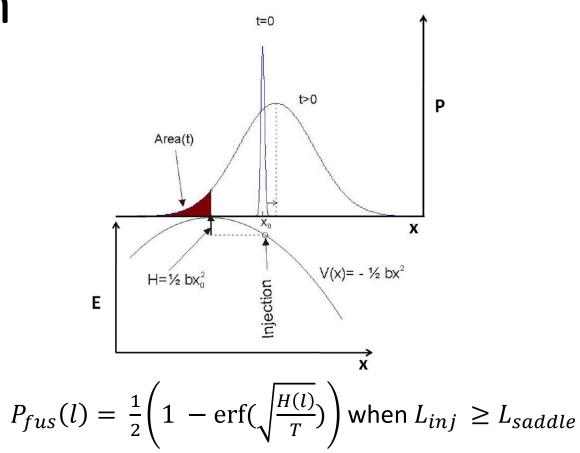
P_{fus} is calculated by solving 1D Smoluchowski Diffusion Equation

P_{fus} in Fusion by Diffusion

1D motion approximation The system must overcome an internal barrier **H** to fuse.



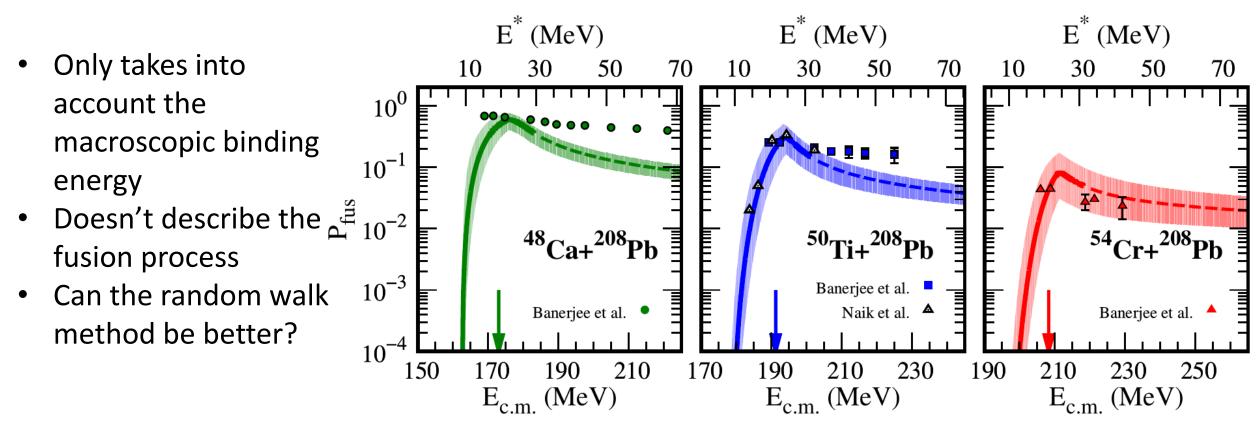
L is the effective elongation (along the fusion path)



H(l) – the function of angular momentum
and bombarding energy *T* – the temperature depends on available energy



Fusion probability from FbD model

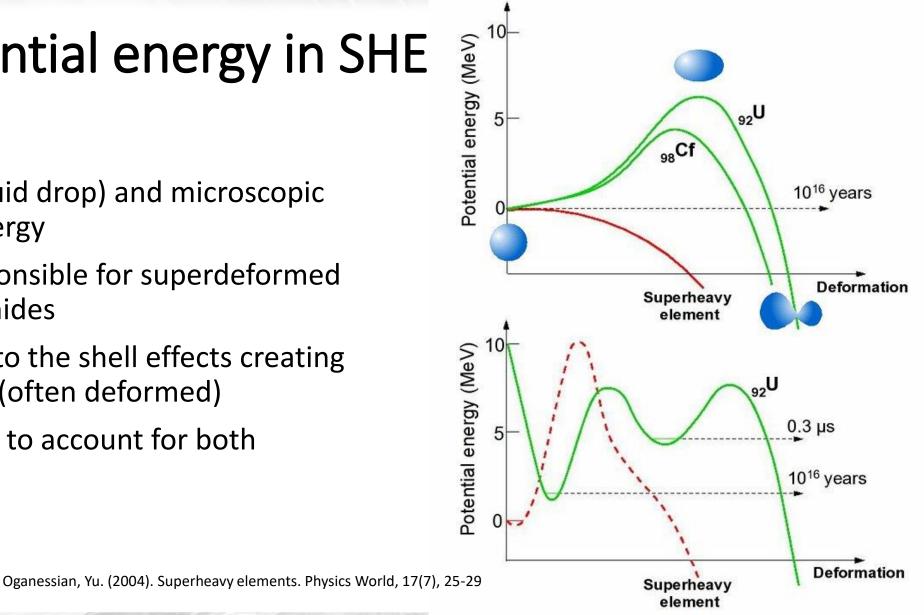


T. Cap, M. Kowal, and K. Siwek-Wilczyńska, Phys. Rev. C 105, L051601 (2022)



Binding/Potential energy in SHE

- Macroscopic (liquid drop) and microscopic (shell effects) energy
- Shell effects responsible for superdeformed minimum in actinides
- SHE exist thanks to the shell effects creating the ground state (often deformed)
- The model needs to account for both energies





Atomic Data and Nuclear Data Tables 138 (2021) 101393



Check fo

Properties of heaviest nuclei with $98 \le Z \le 126$ and $134 \le N \le 192$

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Ground-state and saddle-point shapes and masses for 1305 heavy and superheavy nuclei

including odd-A and odd–odd systems. Static fission barrier heights, one- and two-nucleon separation energies, and $Q\alpha$ values.

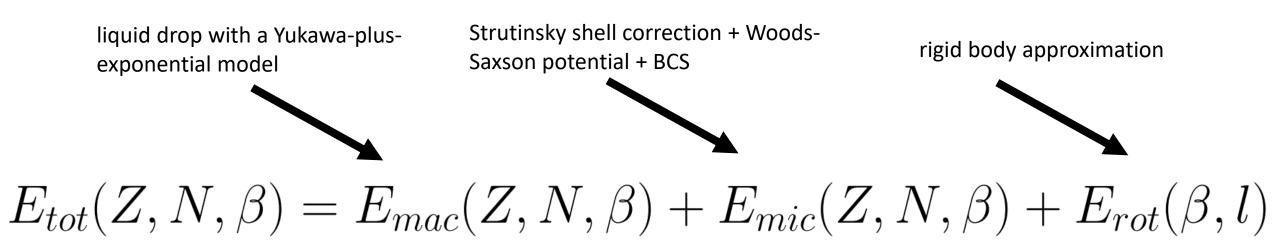
Microscopic—macroscopic method with the deformed Woods—Saxon single-particle potential and the Yukawa-plus-exponential macroscopic energy taken as the smooth part.

Ground-state shapes and energies are found by the minimization over seven axially-symmetric deformations. A search for saddle-points was performed by using the "imaginary water flow" method in three consecutive stages, using five- (for nonaxial shapes) and seven-dimensional (for reflection-asymmetric shapes) deformation spaces.

Good agreement with the experimental data for actinides.



Warsaw macro-micro model with rotational energy



 <u>Allows to obtain the binding energy for a given nuclear shape β and angular</u> <u>momentum *I*.
</u>



Macroscopic energy E_{mac}

- Liquid drop model
- Responsible for the majority of energy/mass
- Dependence on shape in the surface and Coulomb term
- Most often normalized with respect to the sphere

 $E_{mac}^{gs} = E_{mac}^{gs}(deformation) - E_{mac}^{gs}(sphere)$

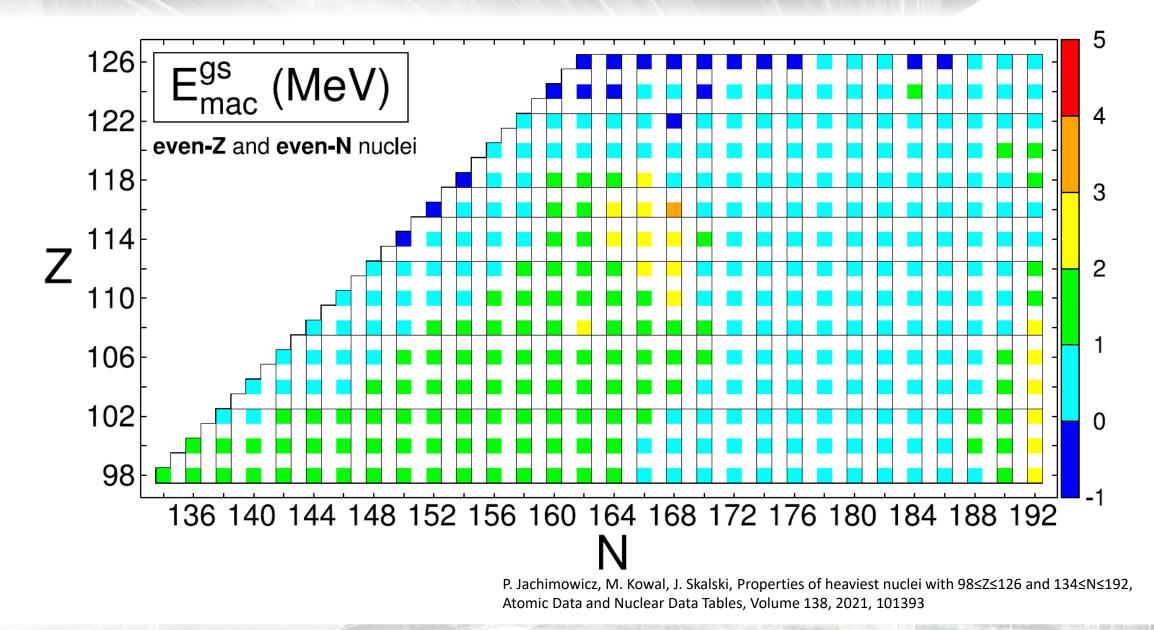
$$E_{mac}(Z,N,\beta) = -a_{v}(1-\kappa_{v}I^{2})A + a_{s}(1-\kappa_{s}I^{2})A^{2/3}B_{s}(\beta)$$

of energy/mass
surface and
$$-f(k_{F}r_{p})Z^{2}A^{-1} + \bar{\Delta}_{mac}$$

$$B_{S} = \frac{A^{-2/3}}{8\pi^{2}r_{0}^{2}a^{4}} \int \int_{V} \left(2 - \frac{r_{12}}{a}\right) \frac{e^{-r_{12}/a}}{r_{12}/a} d^{3}r_{1}d^{3}r_{2},$$

$$B_C = \frac{15}{32\pi^2} \frac{A^{-5/3}}{r_0^5} \int \int_V \frac{1}{r_{12}} \left[1 - \left(1 + \frac{1}{2} \frac{r_{12}}{a_{den}} \right) e^{-r_{12}/a_{den}} \right] d^3 r_1 d^3 r_2$$







Microscopic energy E_{mic}

$$E_{mic}(Z,N,\beta) = E_{corr}^{sh}(Z,N,\beta) + E_{corr}^{pair}(Z,N,\beta)$$

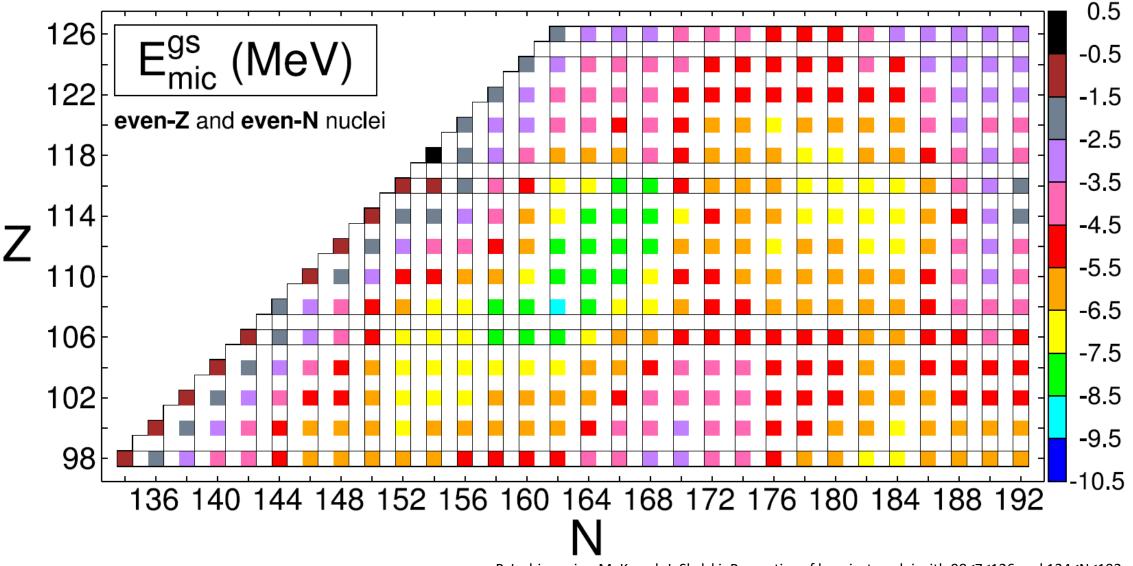
• Strutinsky shell correction based on the deformed Woods–Saxon singleparticle potential $V_{WS}(\vec{r})$

$$(\vec{r}) = -rac{V}{1+e^{d(\vec{r},\beta)/a_{ws}}}$$

- The single-particle potential is diagonalized in the deformed harmonic-oscillator basis
- Pairing energy, pair correlation from Bardeen–Cooper–Schrieffer (BCS) theory

Jachimowicz, P. & Kowal, M. & Skalski, J.. (2014). Q α values in superheavy nuclei from the deformed Woods-Saxon model. Physical Review C. 89. 10.1103/PhysRevC.89.024304.





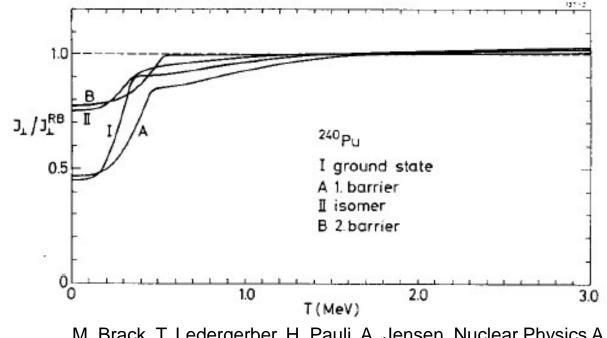
P. Jachimowicz, M. Kowal, J. Skalski, Properties of heaviest nuclei with 98≤Z≤126 and 134≤N≤192, Atomic Data and Nuclear Data Tables, Volume 138, 2021, 101393



Rotational energy E_{rot}

- Rigid body approximation
- Moment of inertia calculated analytically

$$E_{rot} = l(l+1)\frac{(\hbar c)^2}{2I(\beta)} \quad \mathbf{I} = \begin{pmatrix} I_{\perp} & \\ & I_{\perp} \\ & & I_{\parallel} \end{pmatrix}$$

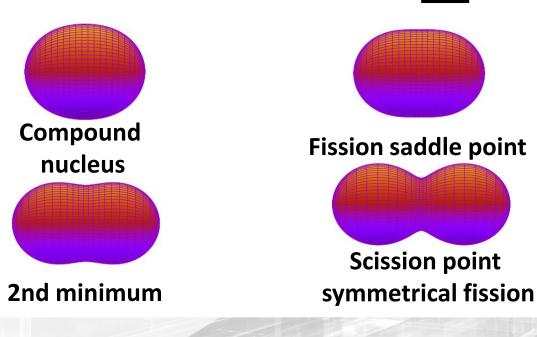


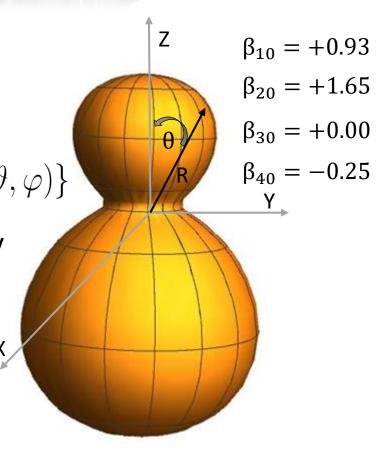
M. Brack, T. Ledergerber, H. Pauli, A. Jensen, Nuclear Physics A **234**, 185–215 (1974)

$$I_{\perp} = \frac{1}{5}\rho R_0^5 \int \sin(\theta) (\pi \sin^2(\theta) + 2\pi \cos^2(\theta)) (1 + \frac{1}{2}\sqrt{\frac{3}{\pi}}\beta_{10}\cos(\theta) + \frac{1}{4}\sqrt{\frac{5}{\pi}}\beta_{20}(3\cos^2(\theta) - 1)) (1 + \frac{1}{4}\sqrt{\frac{7}{\pi}}\beta_{30}(5\cos^3(\theta) - 3\cos(\theta)) + \frac{1}{16}\sqrt{\frac{9}{\pi}}\beta_{40}(35\cos^4(\theta) - 30\cos^2(\theta) + 3))^5 d\theta$$



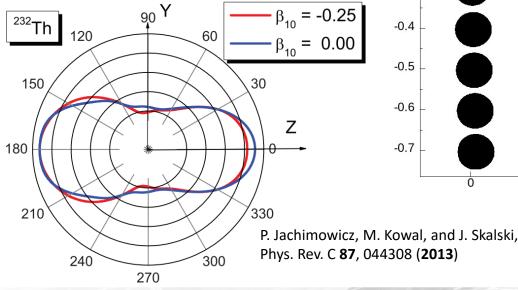
- An expansion of the nuclear radius R(θ , ϕ) onto spherical harmonics Y_{$\lambda\mu$}(θ , ϕ) is used: $R(\vartheta, \varphi) = cR_0\{1 + \sum_{\lambda=1}^{\infty} \beta_{\lambda 0} Y_{\lambda 0}(\vartheta, \varphi)\}$
- For now, shapes in random walk method are limited to axially symmetrical ($\mu = 0$) and depend only on β_{10} , β_{20} , β_{30} and β_{40} .

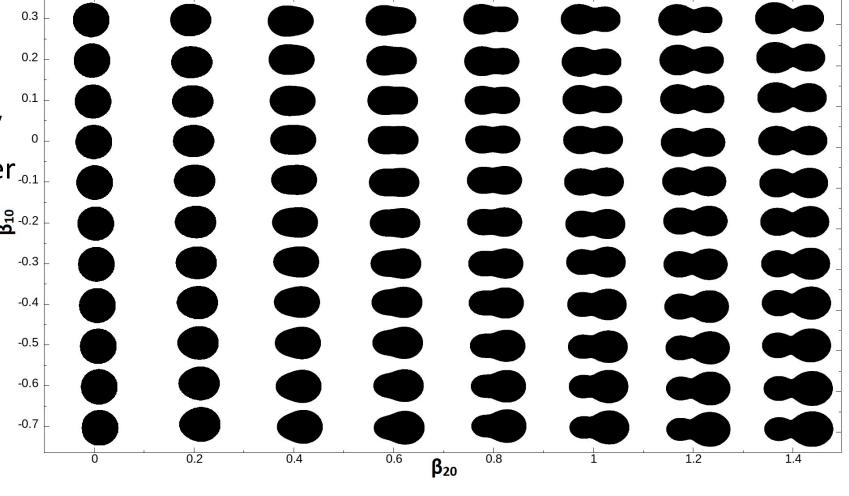






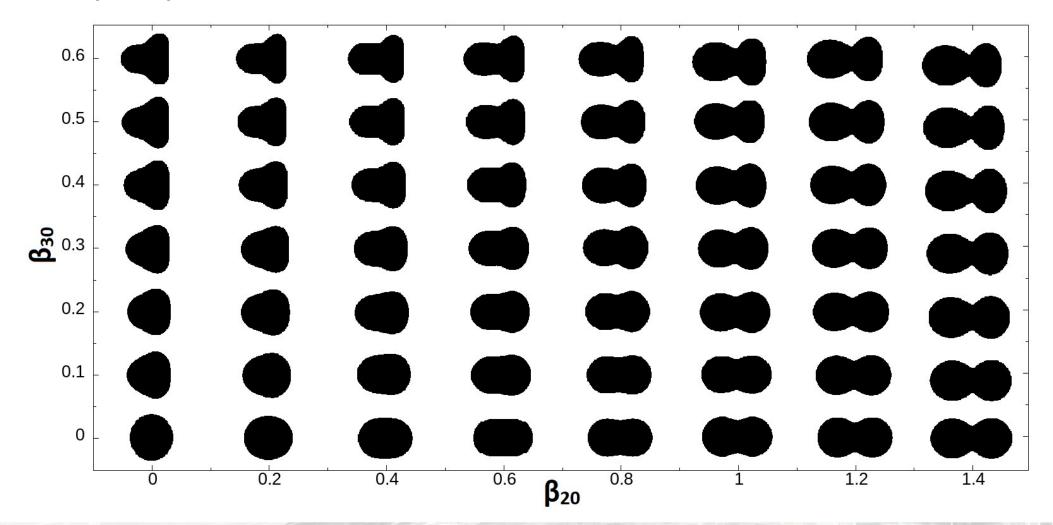
- β_{10} dipole
- β_{20} quadrupole/elongation
- β_{30} hexadecople/asymmetry
- β_{40} octupole/neck parameter -0.1
- β_{10} used as an actual shape β_{10} -0.2 parameter





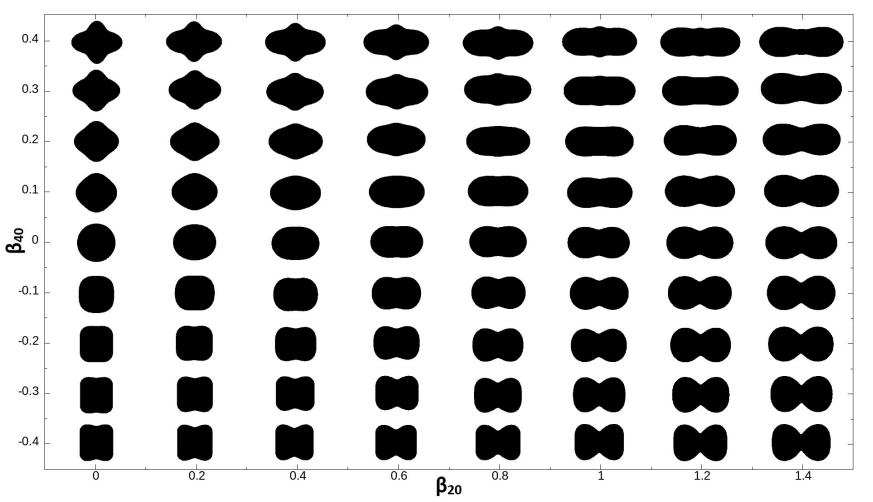
Phys. Rev. C 87, 044308 (2013)







 β₄₀ is crucial in the beginning of the fusion process and during scission

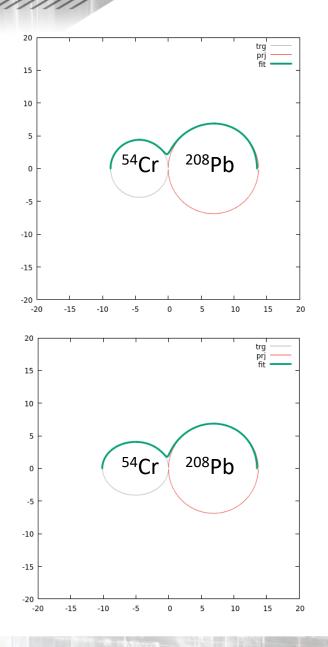




Starting point parametrization

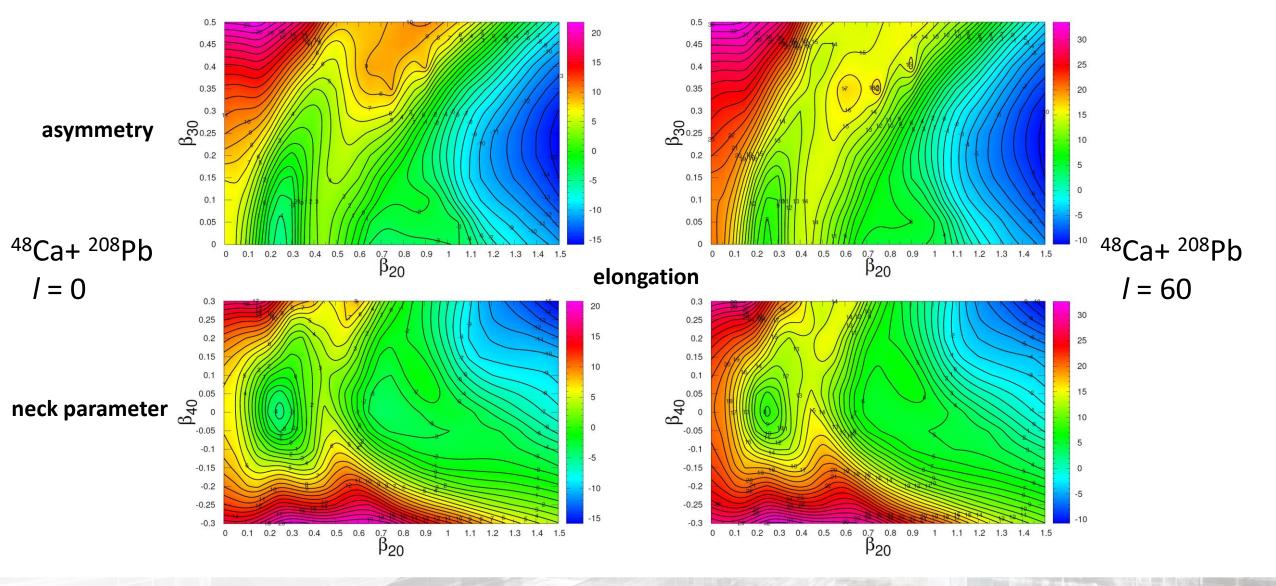
- After overcoming the entrance channel barrier, the projectile and the target are assumed to be in a touching configuration.
- The spherical harmonic parametrization is fitted, with the origin situated in the neck, giving the β parameters for the starting point configuration.
- The shapes are asymmetrical, but β_{30} is near to 0, thanks to the inclusion of $\beta_{10}.$
- Calculations were done for ⁵⁴Cr spherical and deformed (least optimal for fusion).

System	β_{10}	β_{20}	β_{30}	β_{40}
⁴⁸ Ca+ ²⁰⁸ Pb	0.93	1.65	0.0	-0.24
$^{50}{ m Ti}{+}^{208}{ m Pb}$	0.92	1.75	0.01	-0.30
⁵⁴ Cr(spherical)+ ²⁰⁸ Pb	-0.87	1.75	-0.01	-0.30
54 Cr(deformed) $+^{208}$ Pb	-0.94	1.83	0.18	-0.16



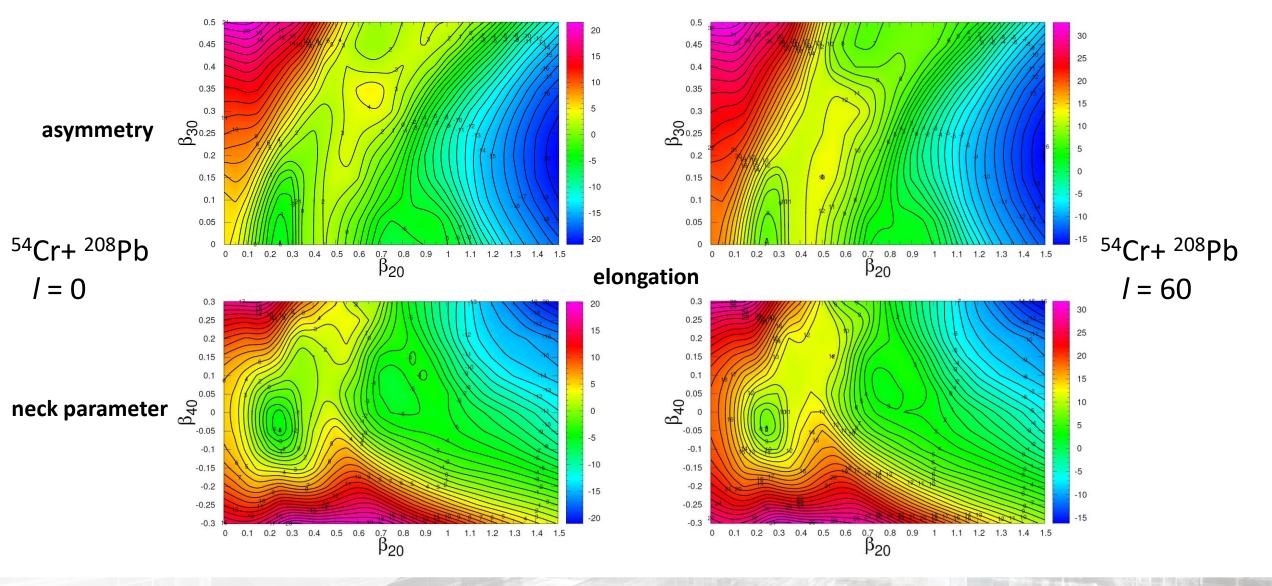


Potential energy surfaces





Potential energy surfaces



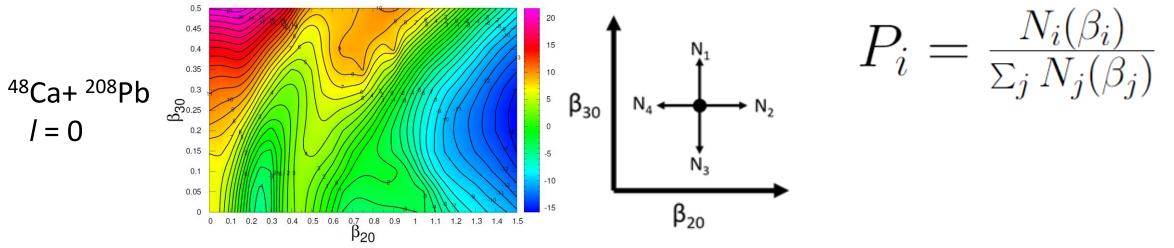


Random walk method

• The probability of transitioning from one shape to another is determined by the number of available energy levels for a given shape β.

 $N_i(\beta_i) \propto \exp\left(2\sqrt{a(E^*(\beta_i) - E_{rot}(\beta_i))}\right)$ a – constant density parameter

 Only one β parameter changes at a time, by a step of 0.05, giving 8 possible directions of movement.



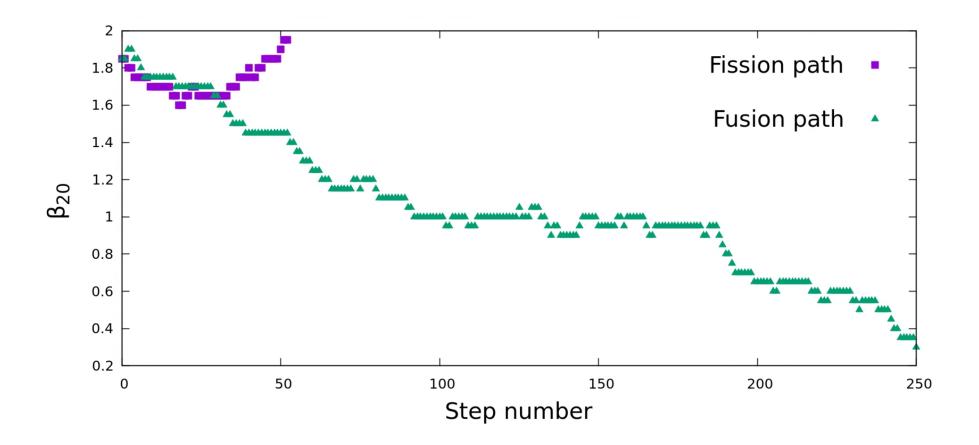
• Fusion: $\beta_{20} \le 0.3$, $\beta_{30} \le 0.2$, $\beta_{40} \le 0.2 \rightarrow$ compound nucleus.

 Fission: when the thickness of the neck connecting the fission fragments is less than 4.0 fm.



Example of a paths

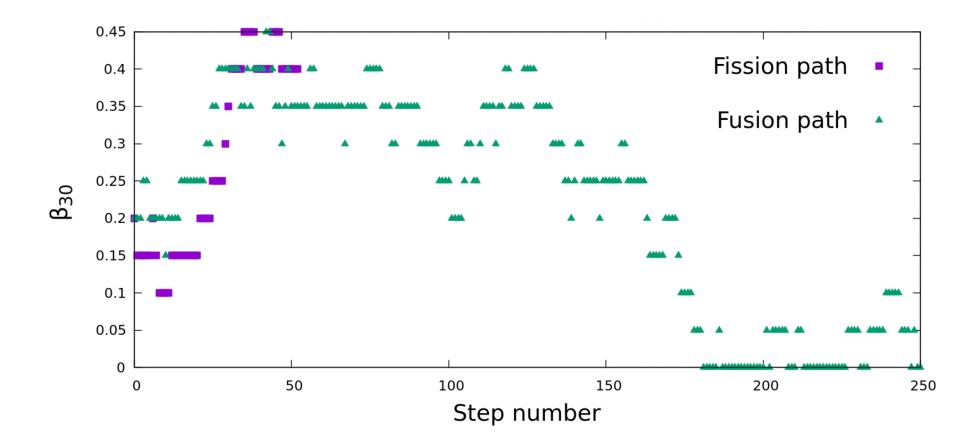
⁵⁴Cr+ ²⁰⁸Pb *E** = 50 MeV, *I* = 40





Example of a paths

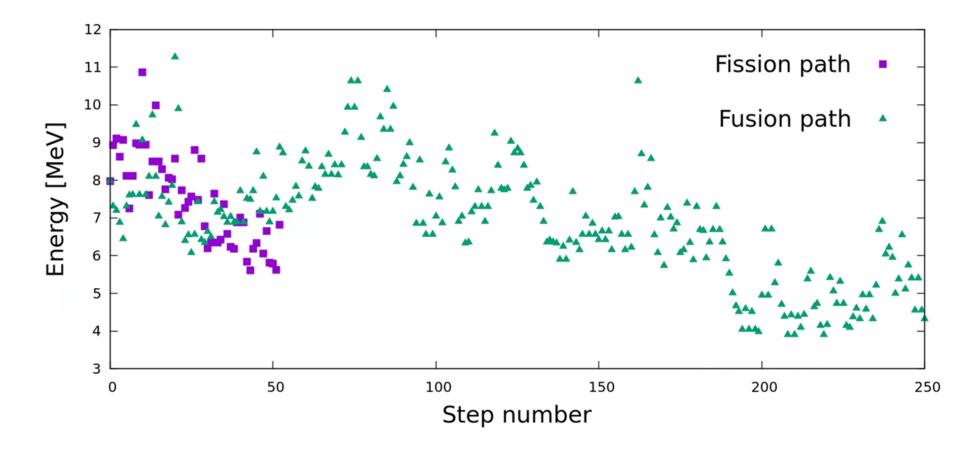
⁵⁴Cr+ ²⁰⁸Pb *E** = 50 MeV, *I* = 40





Example of a paths

⁵⁴Cr+ ²⁰⁸Pb *E** = 50 MeV, *I* = 40





Random walk method

 Calculations were done for excitation energies from 20 to 70 MeV with 1 MeV step. 10000 paths were calculated for a given energy and *l*-value from 0 to *l*_{max}. P_{fus}(E_{cm}, *l*) is given as a ratio of the number of paths that lead to fusion to the total number of paths

 $P_{fus}(E_{cm'} |) = \frac{\text{paths which ended in fusion}}{10000}$

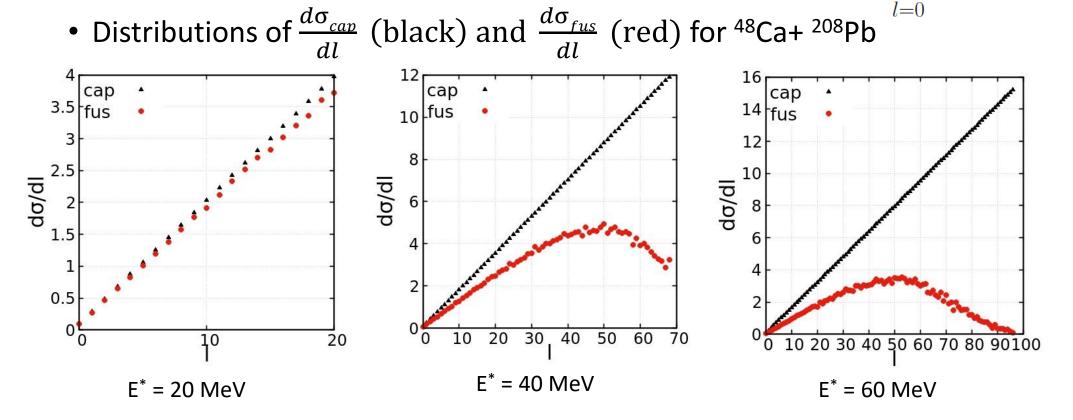
- ~ 3600 E^{*} and *I* combinations \rightarrow ~36 million paths per reaction
- Average over *I* to get P_{fus} dependent on E_{cm} :

$$P_{fus}(E_{cm}) = \frac{\sum_{l=0}^{lmax}(2l+1)P_{fus}(l)}{(2l_{max}+1)^2} \text{ fusion probability averaged over } l$$

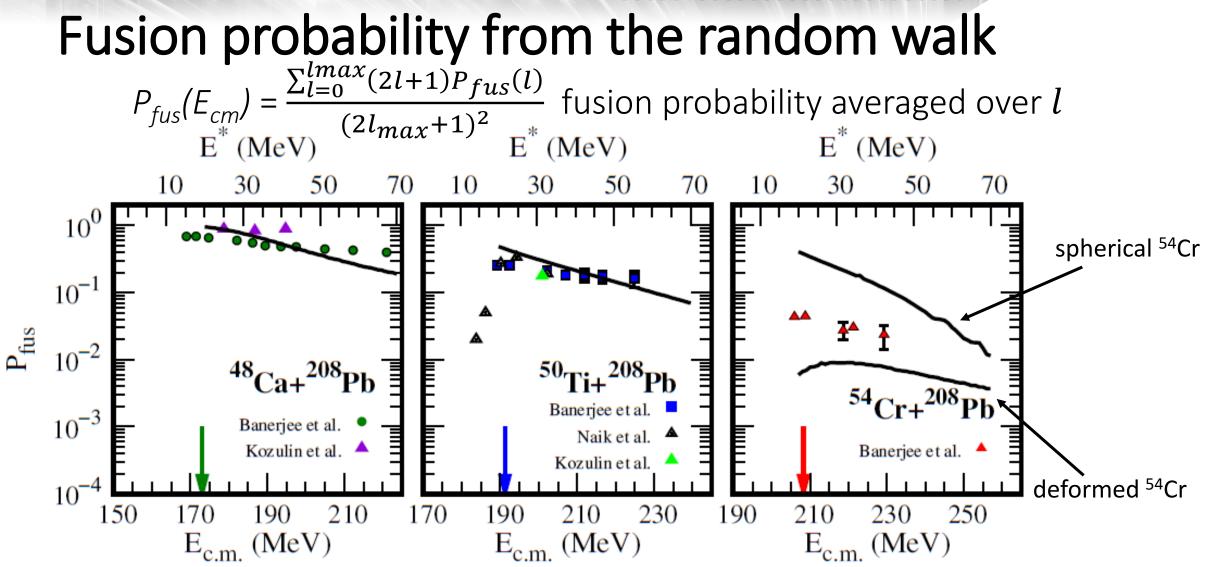


Capture and fusion cross section

- Capture cross section calculated from FbD model
- Fusion cross section from the random walk method $\sigma_{fus} = \pi \lambda^2 \sum (2l+1)T(l)P_{fus}(l) = \sigma_{cap} \times P_{fus}(l)$



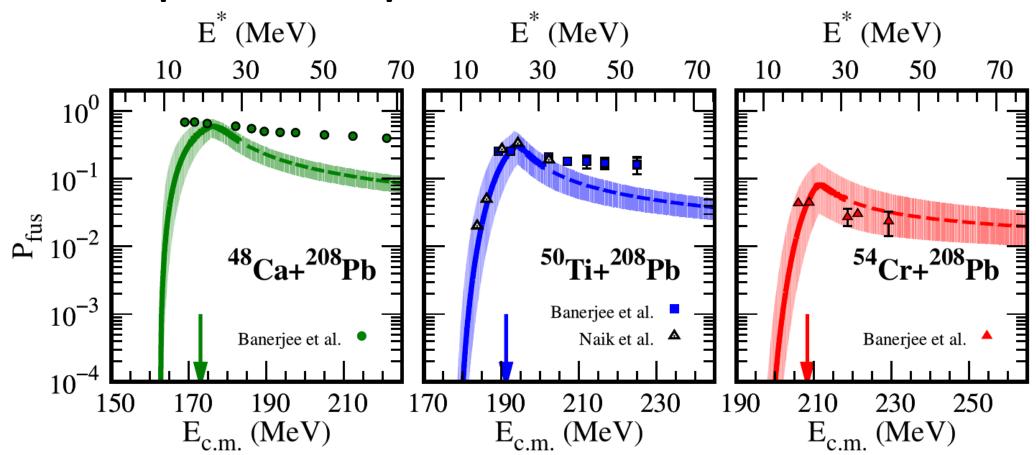




The averaged fusion probability, shown as solid, black lines, calculated with the random walk method, for the reactions 48Ca+208Pb, 50Ti+208Pb and 54Cr+208Pb. For comparison, experimental results obtained in [K. Banerjee et al., PRL 122, 232503 (2019)] and [E. M. Kozulin et al., NPA 802, 45 (2008)] are also shown. The arrows indicate the value of the mean entrance channel barrier for each reaction



Fusion probability from FbD model

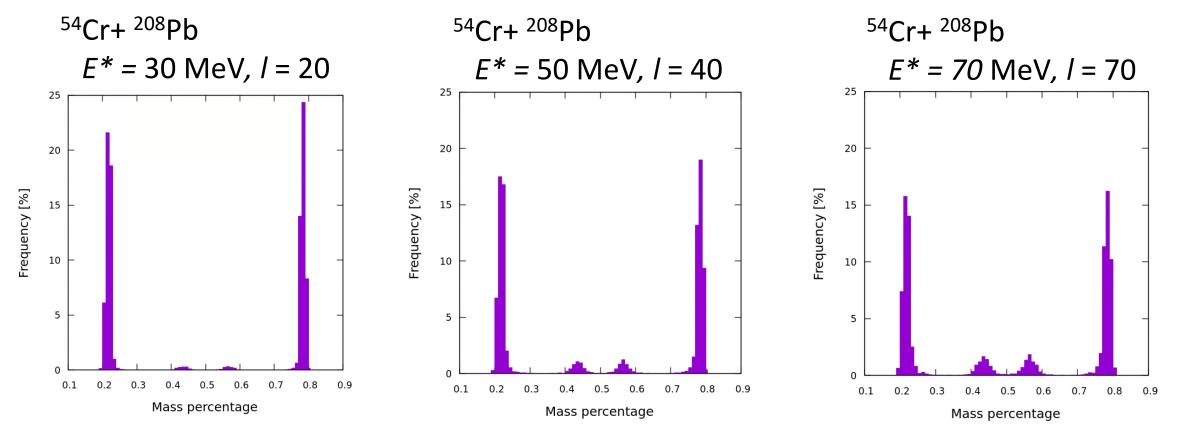


T. Cap, M. Kowal, and K. Siwek-Wilczyńska, Phys. Rev. C 105, L051601 (2022)



Mass distribution of fission fragments

• The final fission shapes can be divided, and their volumes compared, giving the mass distribution of fission fragments, for each E^{*} and *I*.





Summary

- The random walk method reproduces experimental results for spherical pairs of targets and projectiles. For deformed systems, limited to only axially symmetric shapes, the model gives a lower limit of P_{fus} which is an order of magnitude smaller than the data. To properly describe such systems, the method would have to be expanded to non-axially symmetric shapes and would have to incorporate multiple possible starting points depending on the orientation of the target and the projectile.
- The random walk method looks to be a promising direction of study to better understand the fusion probability in reactions leading to the synthesis of superheavy nuclei.

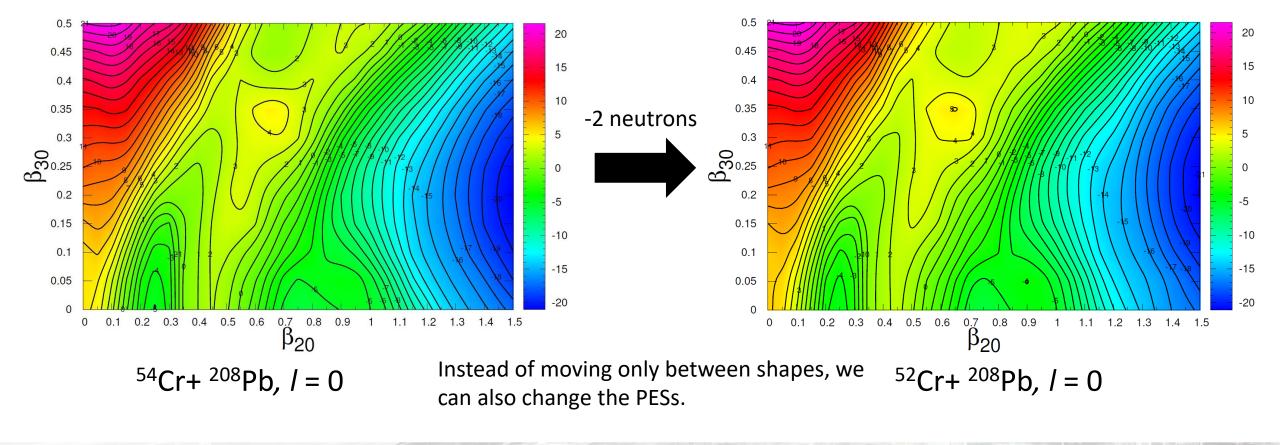


Next steps

- Expand to 8 $\beta_{\lambda 0}$ dimensions
- Determine optimal step size for each β parameter
- Expand to non-axially symmetric shapes ($\beta_{\lambda\mu}$) and incorporate multiple possible starting points depending on the orientation of the target and the projectile
- Introduce a density parameter beyond Fermi gas model
- Incorporate shell-correction damping
- Allow for the emission of neutrons, protons and alfa particles during the random walk
- Calculate P_{fus} for other cold fusion reactions
- Calculate P_{fus} for hot fusion reactions, both already performed and planned experiments



Emission of neutrons, protons and alfa particles during the random walk





Thank you for your attention!



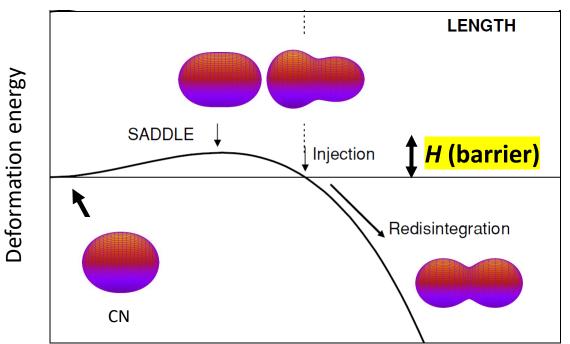
www.ncbj.gov.pl

BONUS SLIDES



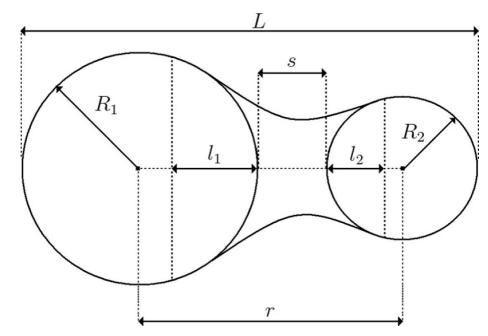
In the **FBD** model, we use 1D motion approximation

The system must overcome an internal barrier **H** to fuse.



L is the effective elongation (along the fusion path)

Macroscopic deformation energies are calculated using the parameterization of the nuclear shapes by two spheres joined smoothly by a third quadratic surface of revolution.

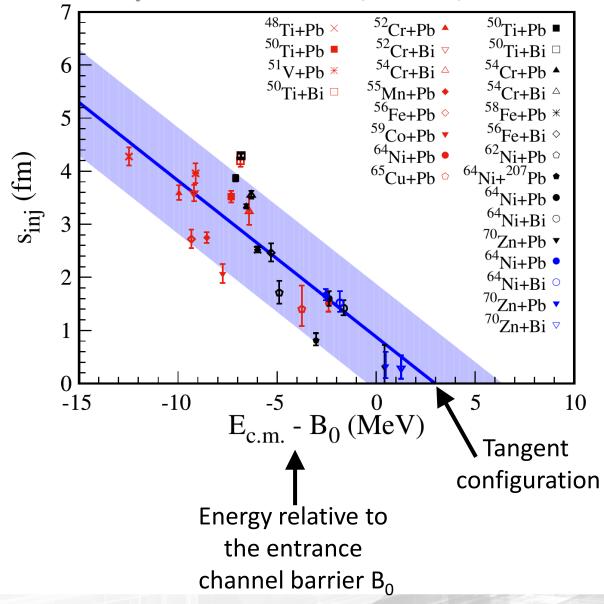


The distance between the nuclear surfaces of two colliding nuclei at the **injection point** s_{inj} is the only adjustable parameter of the model.

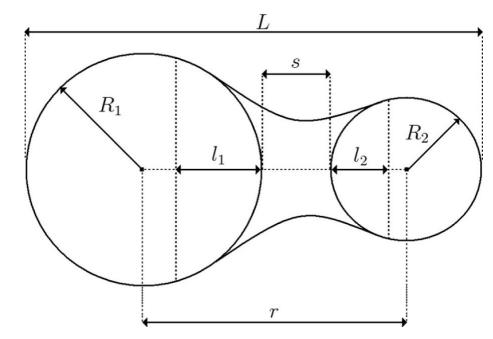
 s_{inj} distance was parametrized by analyzing 27 cold fusion reactions.



 $s_{inj} = 0.878 \text{ fm} - 0.294 \times (E_{c.m.} - B_0) \text{ fm/MeV}$



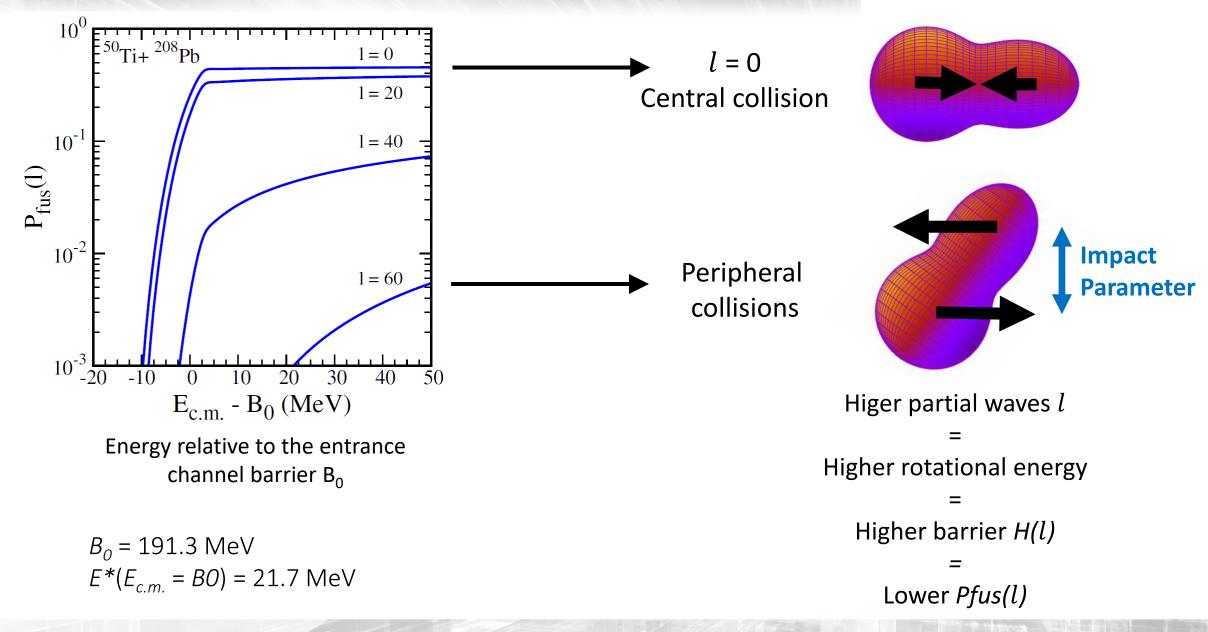
Macroscopic deformation energies are calculated using the parameterization of the nuclear shapes by two spheres joined smoothly by a third quadratic surface of revolution.



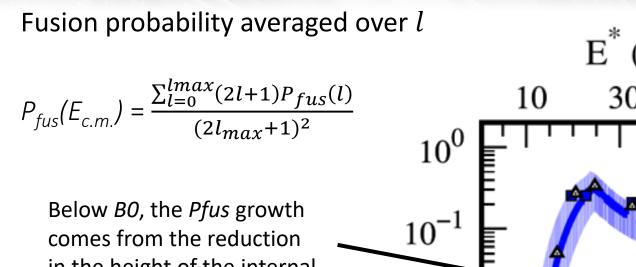
The distance between the nuclear surfaces of two colliding nuclei at the **injection point** s_{inj} is the only adjustable parameter of the model.

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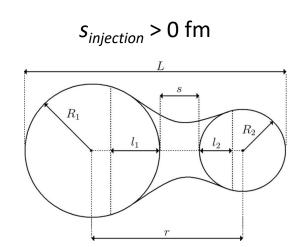


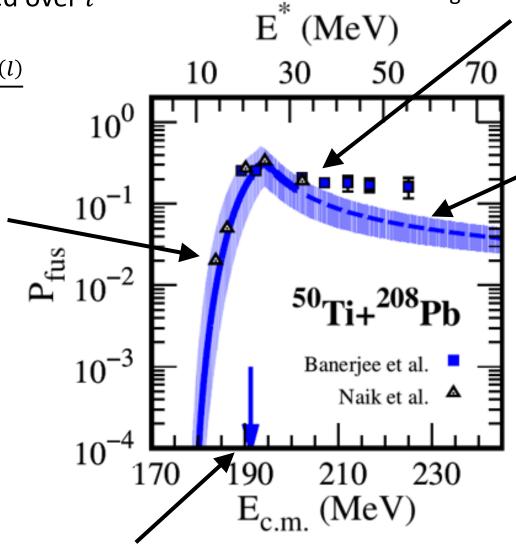






in the height of the internal barrier opposing fusion.





Tangent configuration of projectile and target

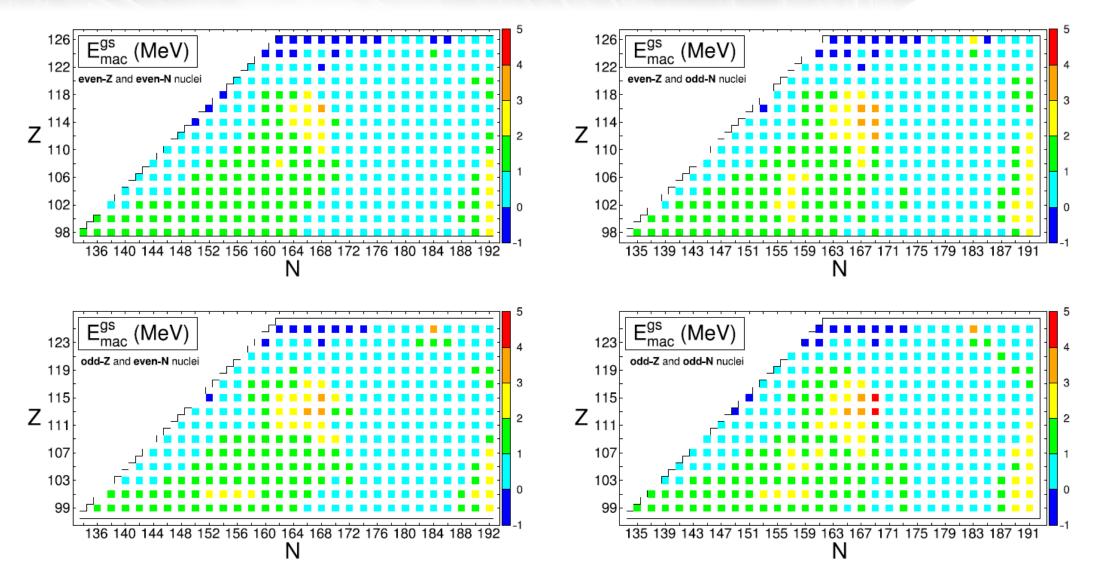
 $(s_{injection} = 0 \text{ fm})$

The *Pfus* saturation above *BO* results from suppression of the contributions from higher partial waves and can be linked to the critical angular momentum.

The difference between rotational energies in the fusion saddle and the contact (sticking) configuration plays a major role in CN formation at energies above *BO*.

B0 - entrance channel barrier (Coulomb+Nuclear potential)







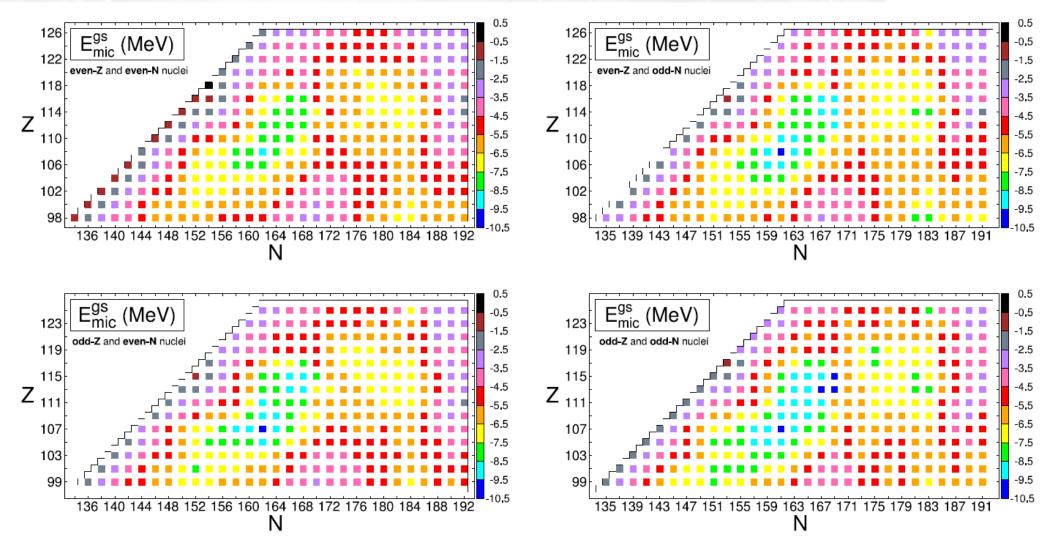


Fig. 2. Calculated microscopic component E_{mic}^{gs} of the ground state binding energy in 4 separate groups of nuclei.



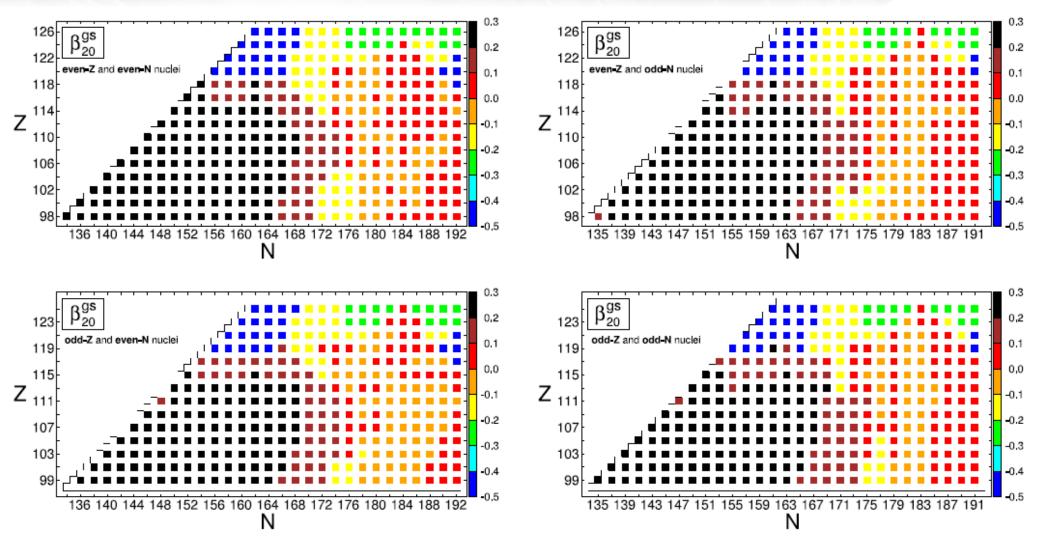


Fig. 4. Calculated ground-state quadrupole deformations β_{20}^{gs} in 4 separate groups of nuclei.



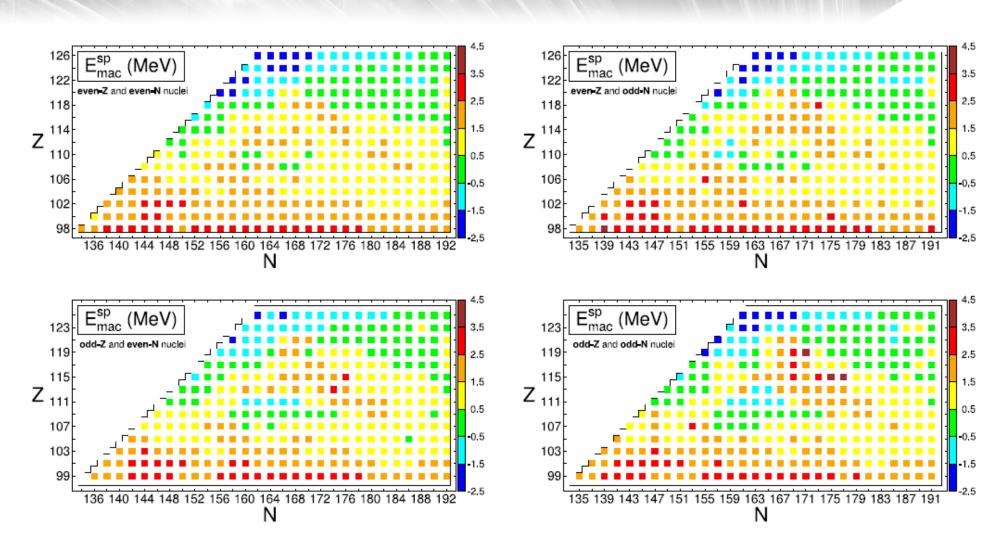


Fig. 12. As in Fig. 1, but for the calculated saddle points.



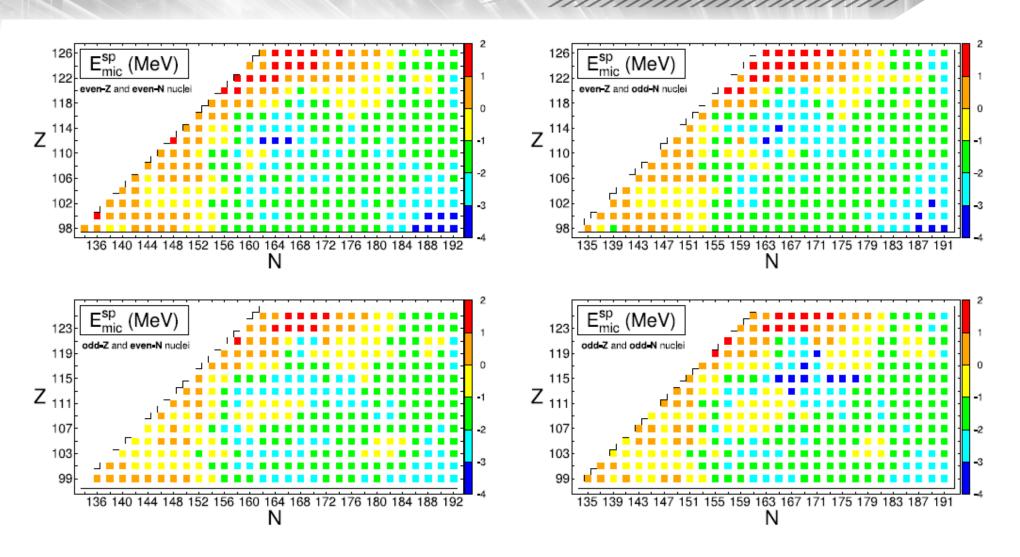


Fig. 13. As in Fig. 2, but for the calculated saddle points.

