Modeling multinucleon transfer reactions based on the Master Equation

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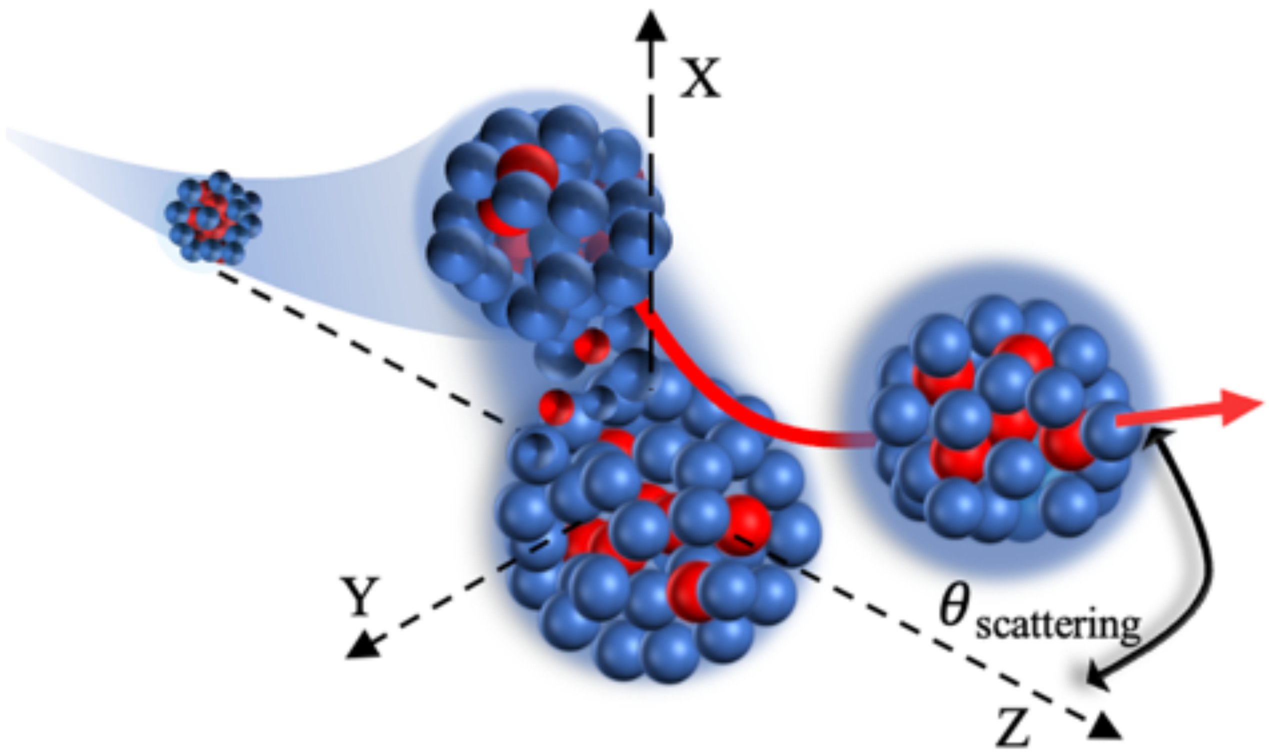
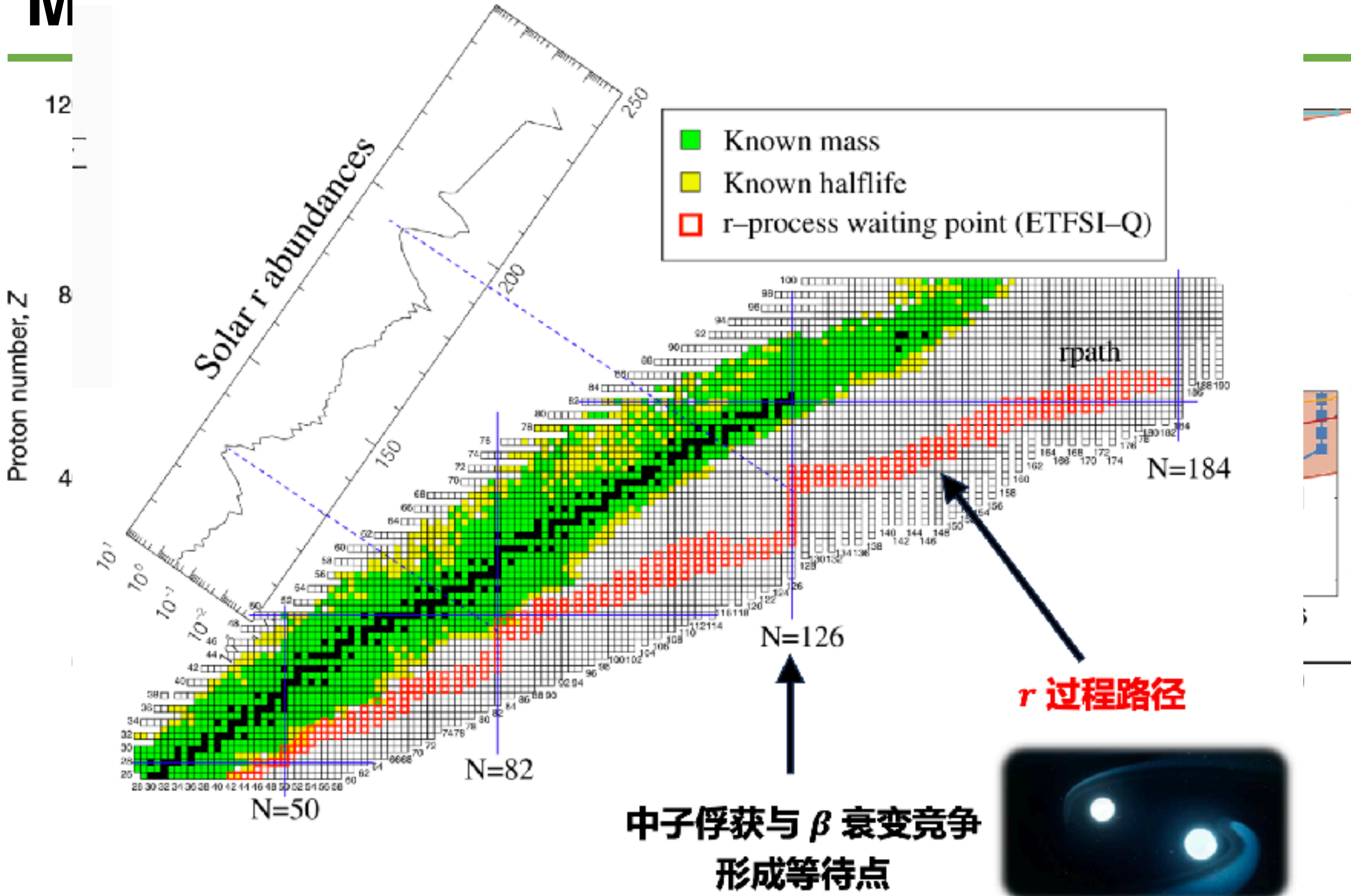
Content

- Background
  - Multinucleon transfer reaction (MNT)
- Modeling MNT with the master equation

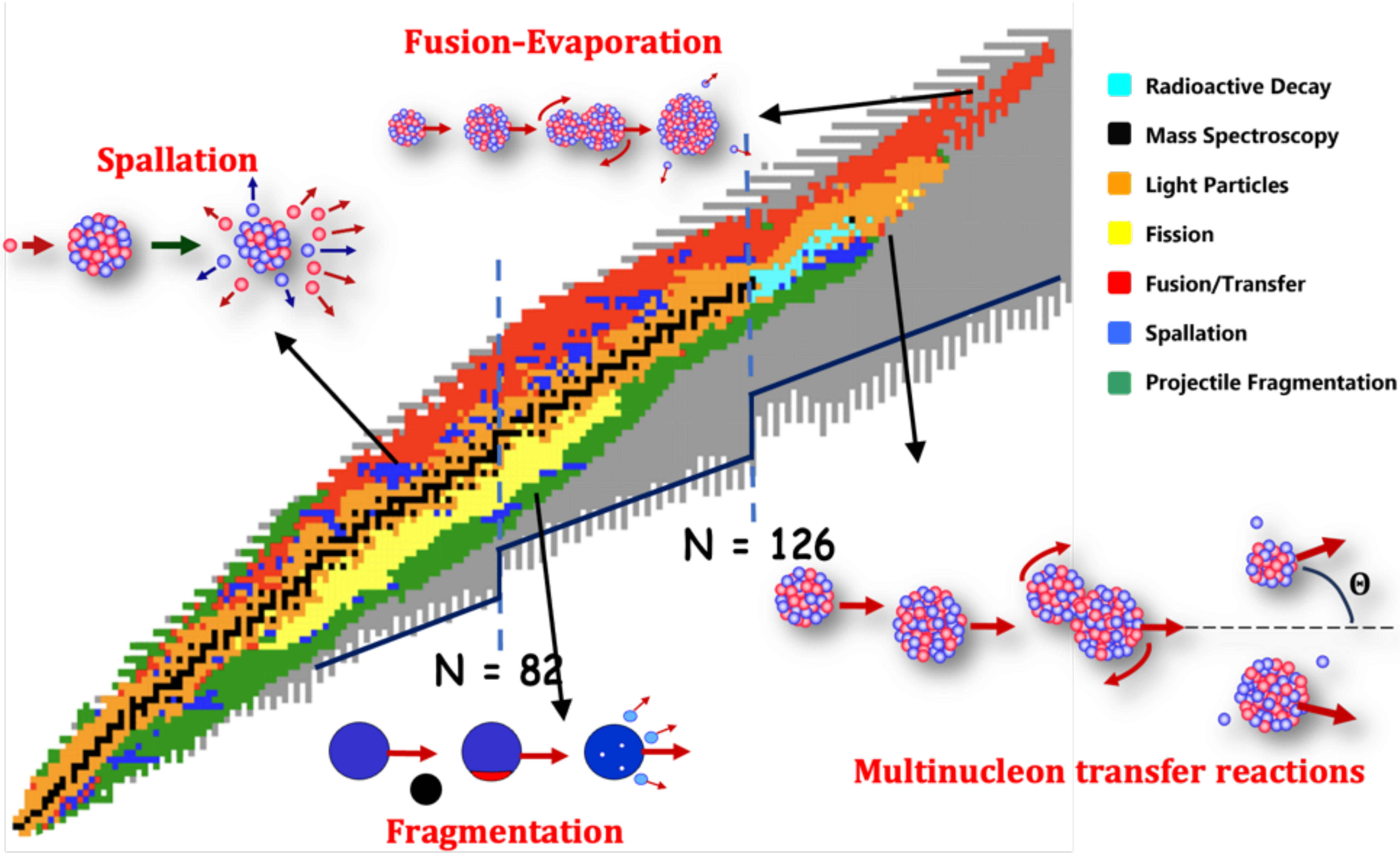




T to
the question about the origin of elements in the universe.

https://people.nscl.msu.edu/~thoennes/isotopes/

MNT · How to expand the nuclide chart



https://people.nscl.msu.edu/~thoennes/isotopes/

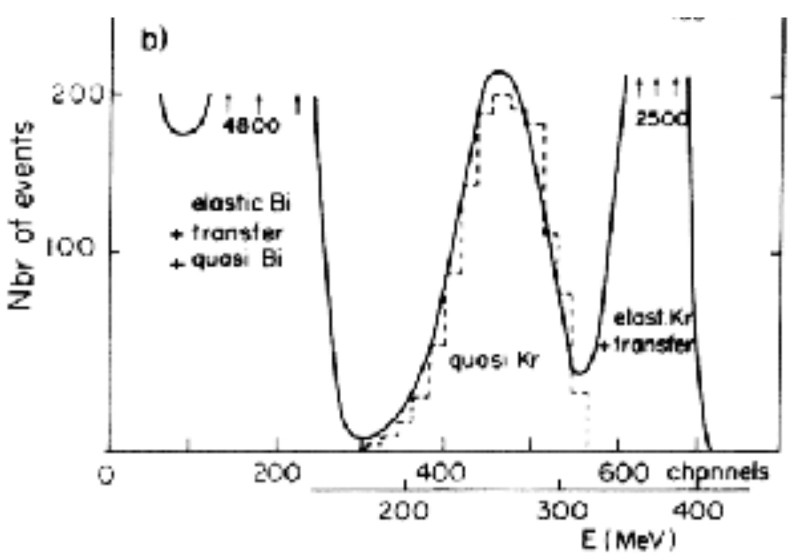
MNT · Origin

- A new type of reaction has been observed at **Dubna**, and independently at **Orsay** and **Berkeley**.

H. M. Devaraja et al., Phys. Part. Nucl. Lett. 19, 693 (2022).

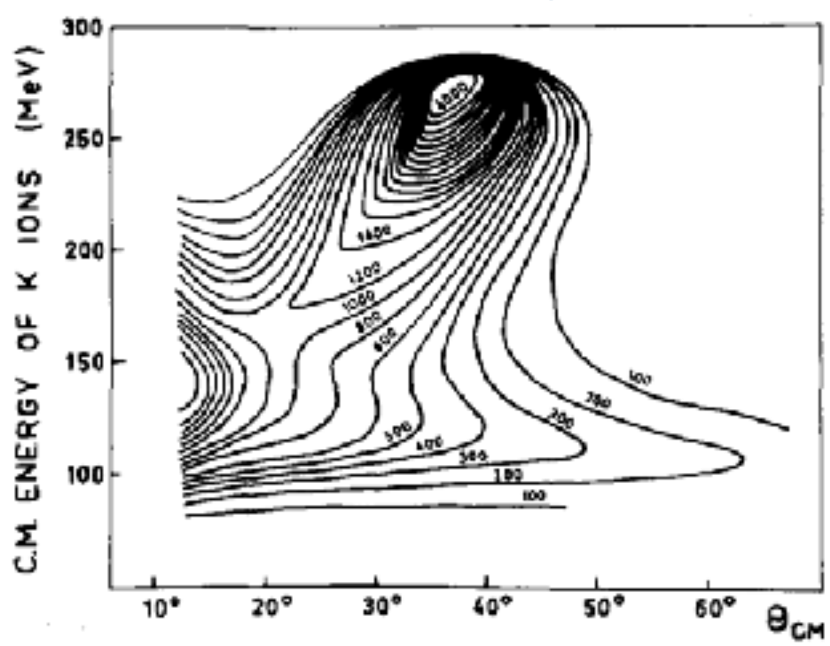
Laboratory	Reactions	Beam energy, MeV (E_B/B_{int})	Identified nuclides	Methods employed
Dubna	$^{18}O + {}^{232}Th$ [54]	122 (1.4) (38.4%)	^{18}C, ^{20}N, ^{22}O	Bρ-ΔE–E
	$^{22}Ne + {}^{232}Th$ [55], [56]	174 (1.6) (57.2%)	$^{23,24}F$, $^{25,26}Ne$, ^{21}N, $^{23,24}O$, ^{25}F	
	$^{40}Ar + {}^{232}Th$ [57]	290 (2.6) (162%)	$^{29,30}Mg$, $^{31-33}Al$, $^{33-36}Si$, $^{35-38}P$, $^{39,40}S$, $^{41,42}Cl$	
Orsay France	$^{40}Ar + {}^{238}U$ [58]	263 (1.3) (26.6%)	^{37}Si, ^{40}P, $^{41,42}S$	TOF-ΔE–E
	$^{40}Ar + {}^{238}U$ [59]	340 (1.6)(64%)	^{54}Ti, ^{56}V, $^{58,59}Cr$, ^{61}Mn, $^{63,64}Fe$	
LBNL Berkeley	$^{56}Fe + {}^{238}U$ [60]	465 (1.5) (50%)	$^{52,53}Sc$, ^{56}Ti, $^{57,58}V$, ^{60}Cr, ^{55}Ti	TOF-ΔE–E

$^{209}Bi + {}^{84}Kr$ at energy 525 MeV



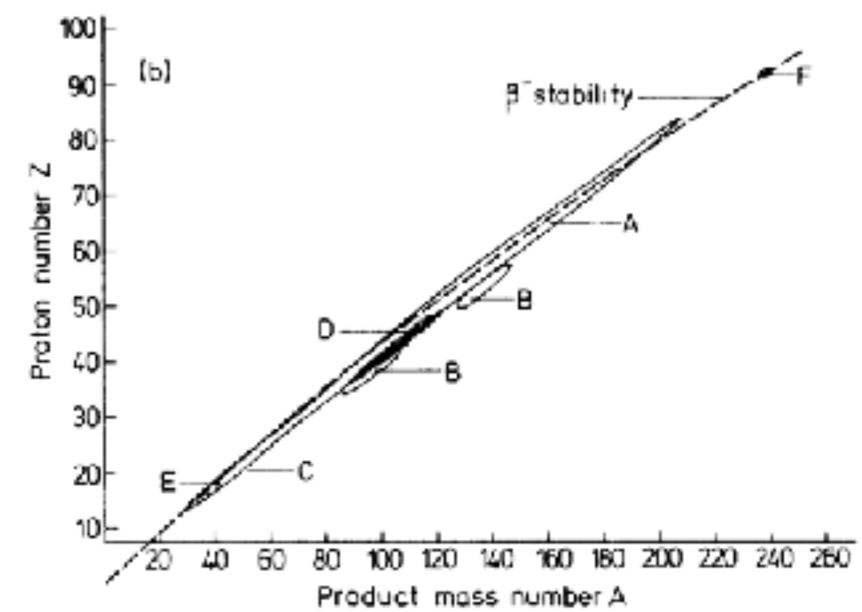
Phys. Rev. Lett. 32. 738 (1974)

$^{232}Th(^{40}Ar, K)$ at energy 388 MeV



Phys. Lett . 47B (1973) 484

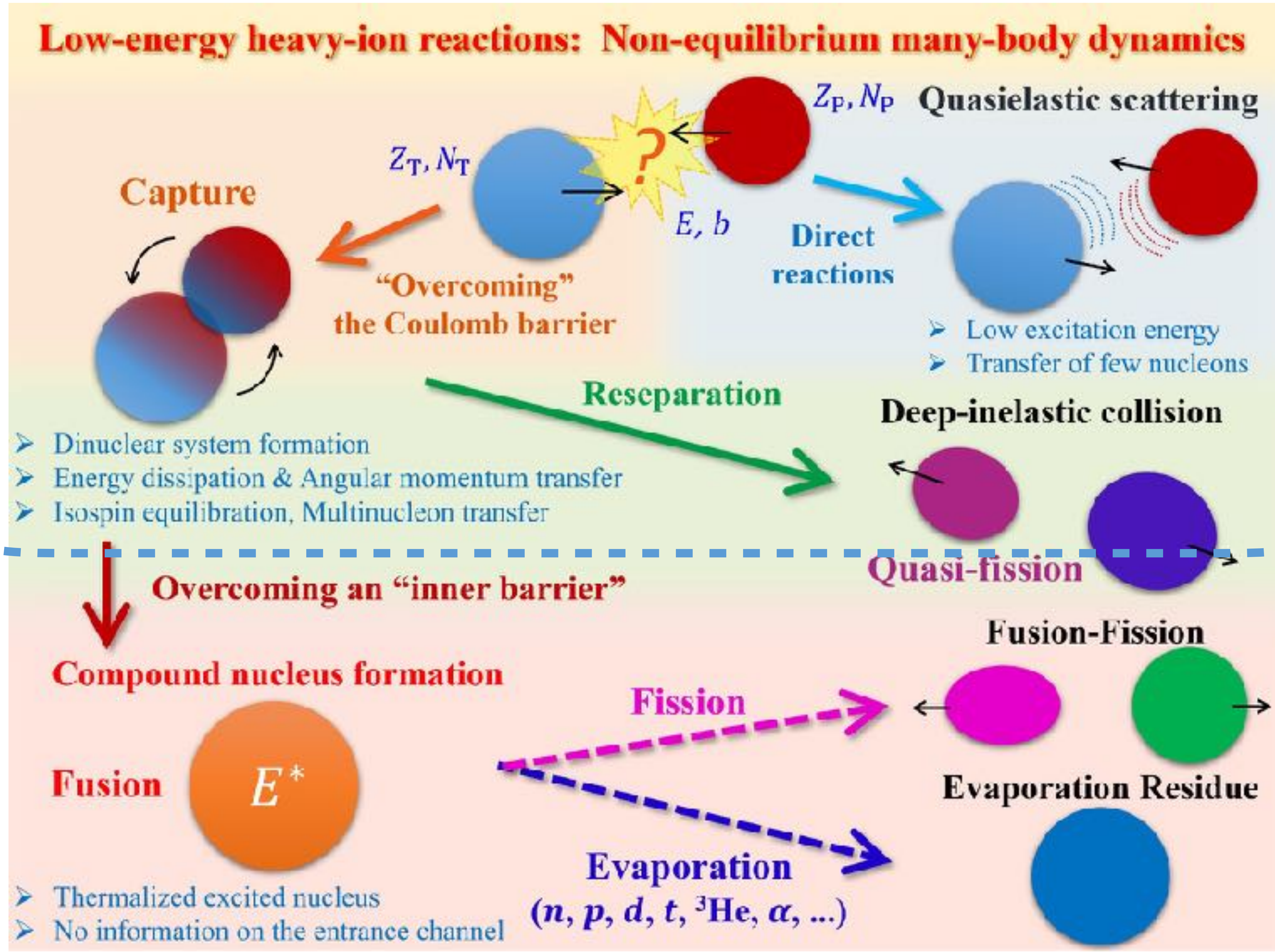
$^{238}U + {}^{40}Ar$ at energy 288 MeV



Phys. RevC. 13. 2347 (1976)

- At different laboratories it was called in different ways: **deep inelastic transfers**, **quasi-fission**, **relaxation phenomena**, and **strongly damped collisions**.

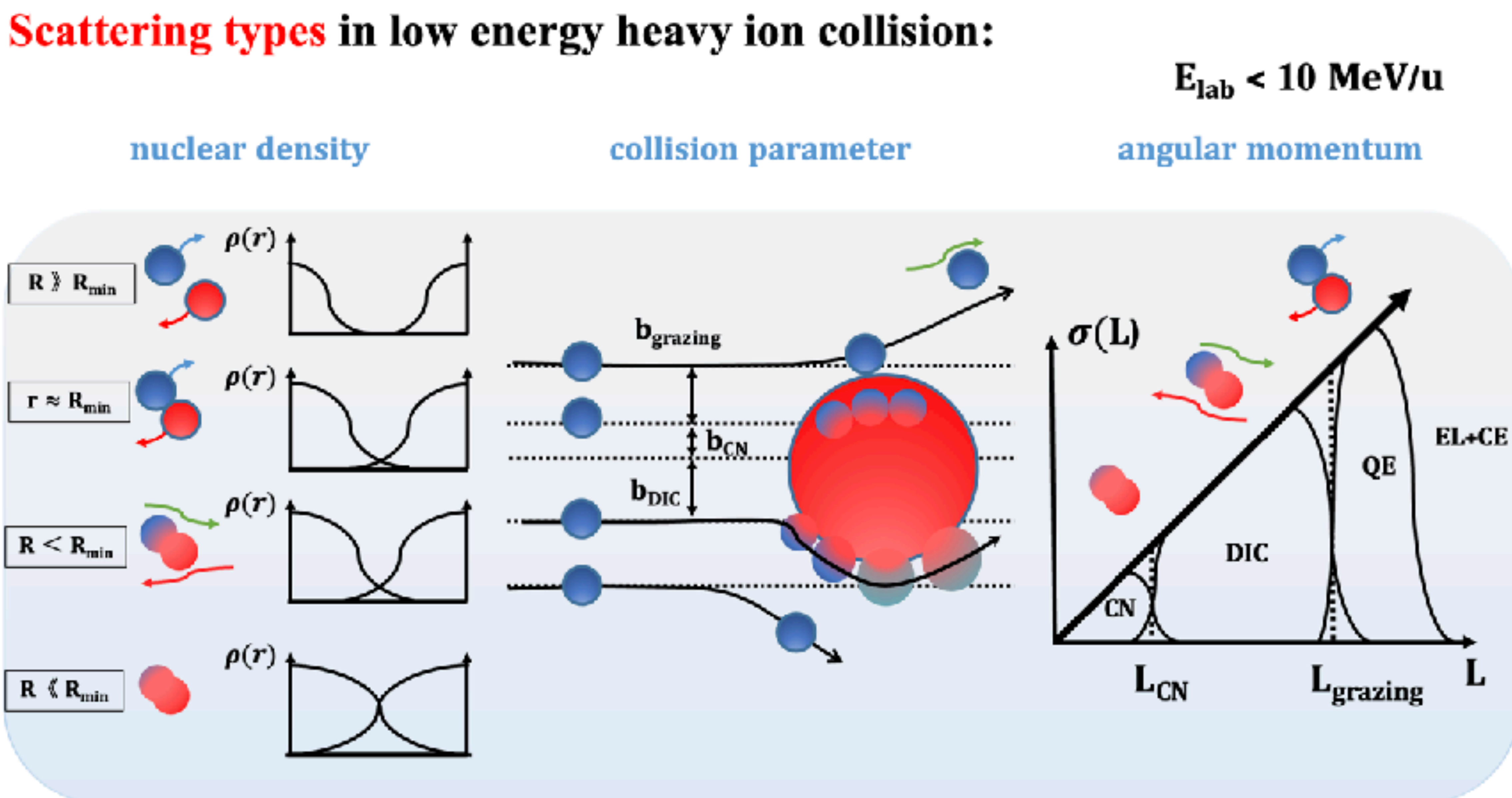
MNT · Low-energy heavy-ion reaction



Complex quantum non-equilibrium evolution process with multiple reaction mechanisms and wide time scale

Sekizawa's talk

MNT · Low-energy heavy-ion reaction



For multi-nucleon transfer reactions or fusion reactions (high excitation energy, and multiple reaction channels) , the macroscopic master equation seems to be a more reasonable choice

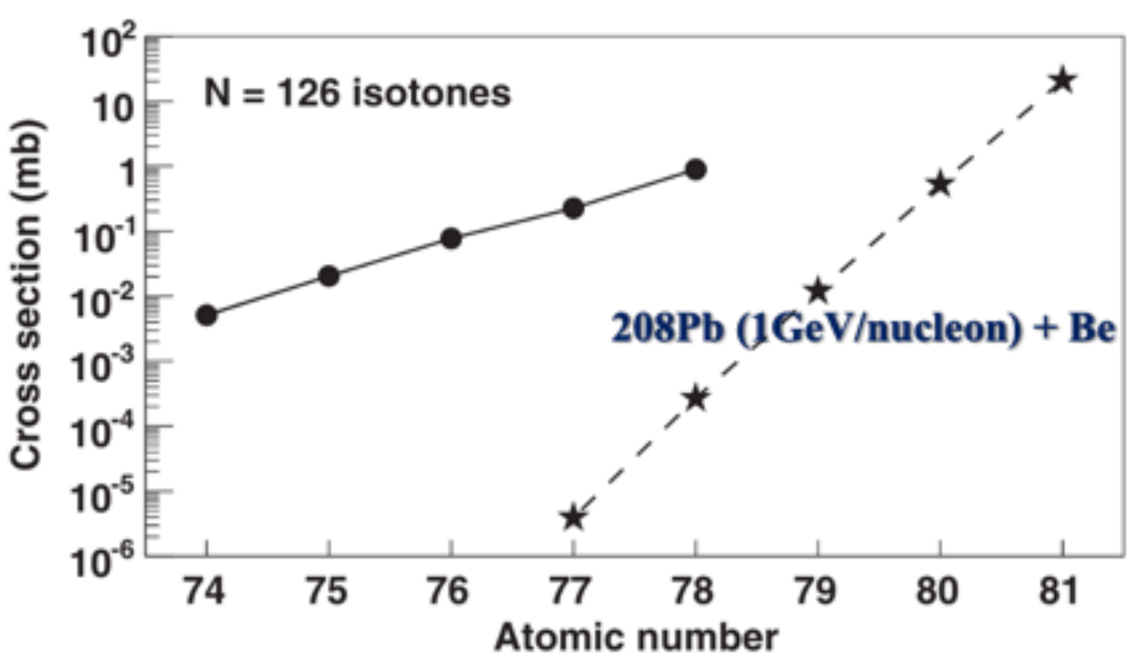
N =126 nuclei ・MNT

PRL **115,** 172503 (2015) PHYSICAL REVIEW LETTERS week ending 23 OCTOBER 2015

Pathway for the Production of Neutron-Rich Isotopes around the $N = 126$ Shell Closure

Y. X. Watanabe,[1,*] Y. H. Kim,[2,3,†] S. C. Jeong,[1,‡] Y. Hirayama,[1] N. Imai,[1,§] H. Ishiyama,[1,‡] H. S. Jung,[1] H. Miyatake,[1] S. Choi,[2,3] J. S. Song,[2,3,4] E. Clement,[5] G. de France,[5] A. Navin,[5,‖] M. Rejmund,[5] C. Schmitt,[5] G. Pollarolo,[6] L. Corradi,[7] E. Fioretto,[7] D. Montanari,[8] M. Niikura,[9,¶] D. Suzuki,[9,**] H. Nishibata,[10] and J. Takatsu[10]

[1]*Institute of Particle and Nuclear Studies, High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan*



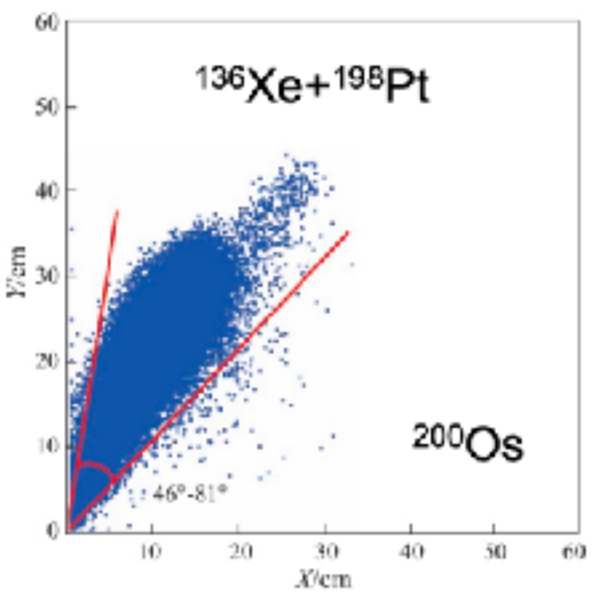
platinum

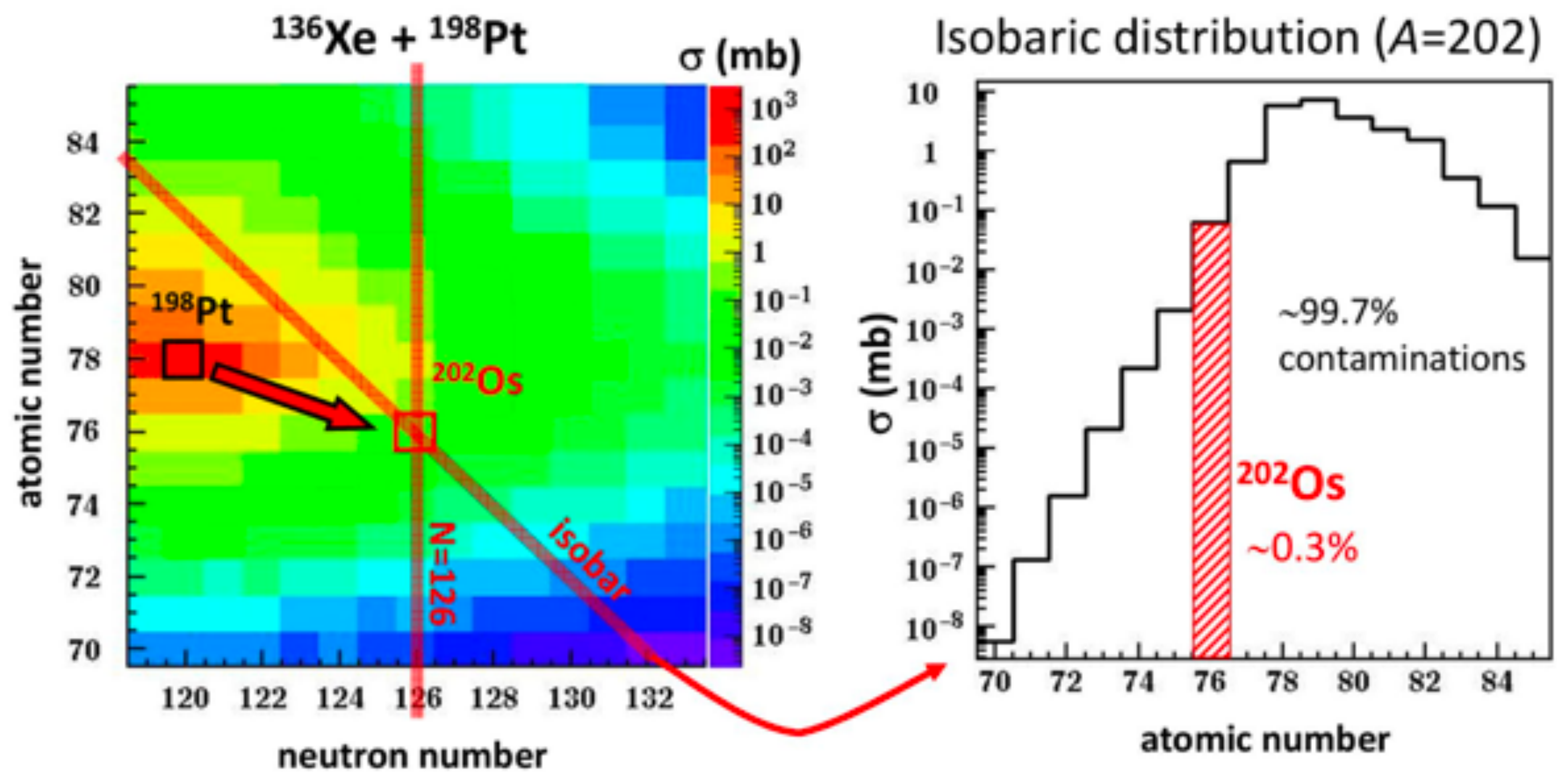
In conclusion, the promised potential of the production of new isotopes around and beyond the neutron shell $N = 126$ by multinucleon transfer reactions was established for the first time in the $^{136}Xe + ^{198}Pt$ system at an energy above the Coulomb barrier. The absolute cross

Y. X. Watanabe et al., PRL 115, 172503 (2015)

Y. X. Watanabe et al.,
Nucl. Instrum. Methods Phys. Res., Sect. B 317, 752 (2013).

MNT ·





Wenxue Huang et al. Nucl. Phys. Rev. (2017)

H. Ikezoe et al., Nucl. Instrum. Meth. A 376, 420 (1996).

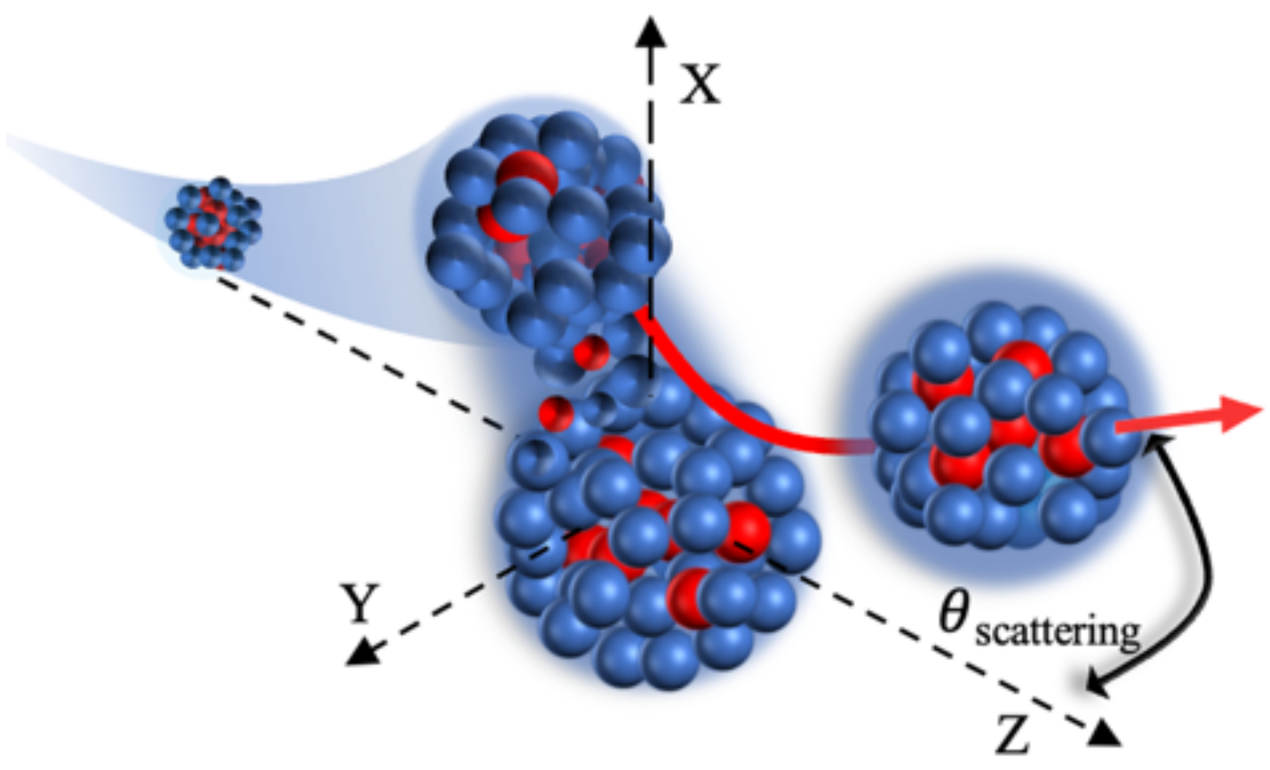
osmium

- **The emission of MNT reaction products in the laboratory system is not in the forward direction near 0°, but covers a wide range of cone angles.**

- **This brings great difficulties to the collection and separation of the Multinucleon transfer reaction products that we are interested in, and requires theoretical support.**

Content

- **Background**
  - **Multinucleon transfer reaction (MNT)**
- **Modeling MNT with the master equation**

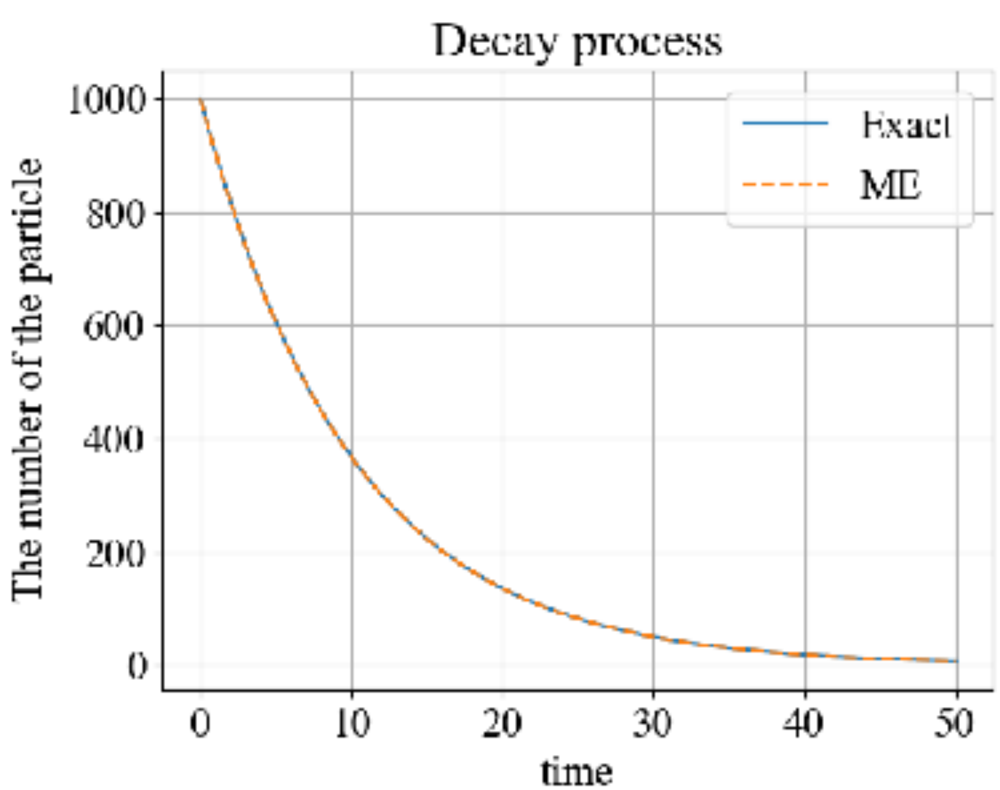


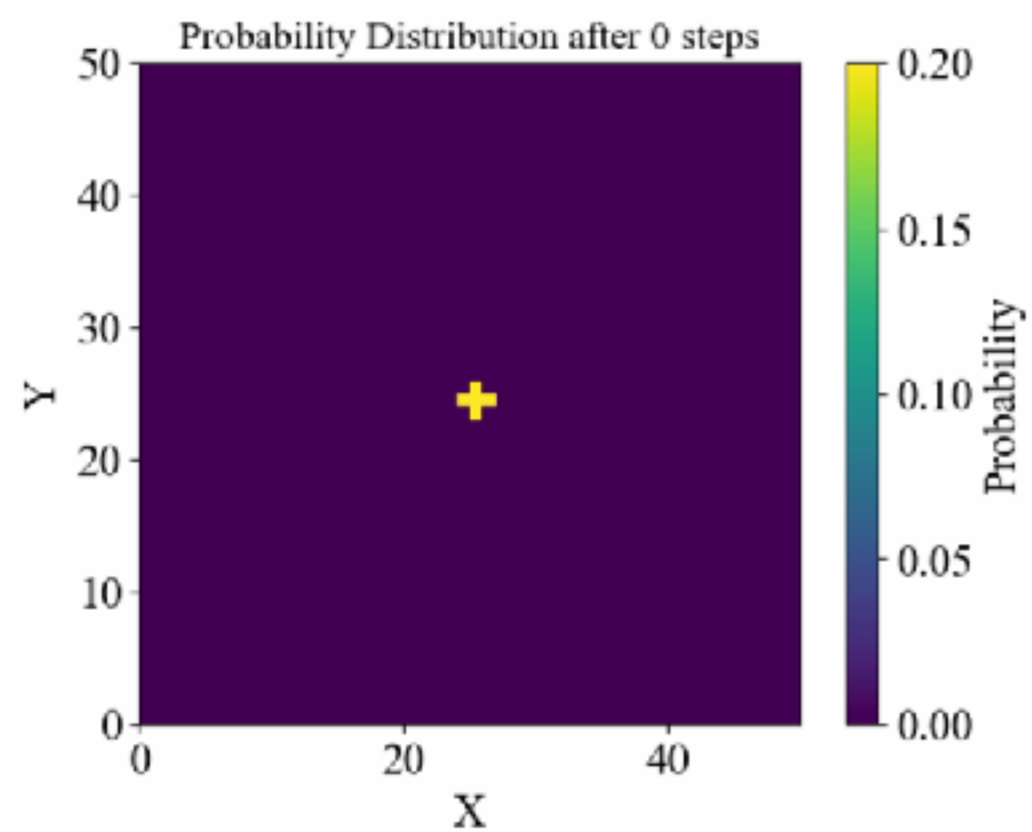
Master equation

A master equation is a phenomenological set of first-order differential equations describing the time evolution of (usually) the probability of a system to occupy each one of a discrete set of states with regard to a continuous time variable t.

$$\frac{\partial P(\mathbf{S},t)}{\partial t} = \sum_{\mathbf{S}'\neq\mathbf{S}} W(\mathbf{S}',\mathbf{S})P(\mathbf{S}',t) - W(\mathbf{S},\mathbf{S}')P(\mathbf{S},t)$$

Many physical problems in classical, quantum mechanics and problems in other sciences, can be reduced to the form of a master equation, thereby performing a great simplification of the problem



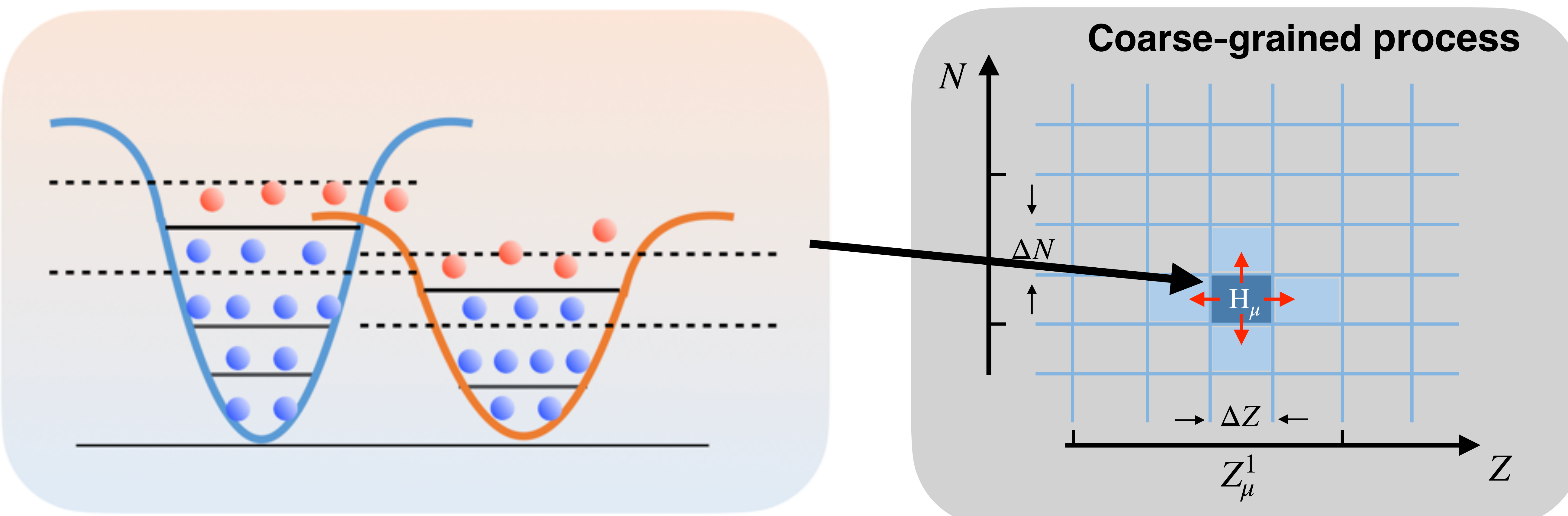


Master equation

The system moves in a multidimensional collective space $\mathbf{S}$, this motion is governed by the master equation:

$$\frac{\partial P(\mathbf{S},t)}{\partial t} = \sum_{\mathbf{S}'\neq\mathbf{S}} W(\mathbf{S}',\mathbf{S})P(\mathbf{S}',t) - W(\mathbf{S},\mathbf{S}')P(\mathbf{S},t)$$

collective space $\mathbf{S}$



Transition Probability

The transition probability from the state to another state

Macroscopic transition probabilities

Microscopic transition probabilities

$$W^{\pm} = \lambda_0 \sqrt{\frac{\rho(S \pm 1)}{\rho(S)}} P_{tr}(R, \beta, S \rightarrow S \pm 1)$$

Diffusion constant

Probability of a nucleon transfer

Diffusion constant λ_0

$$W^{\pm} = \lambda_0 \sqrt{\frac{\rho(A \pm 1)}{\rho(A)}} P_{tr}(R, \beta, A \to A \pm 1)$$

Diffusion constant

It can be qualitatively assumed to be related to the size of the system and the temperature.

Therefore, we treat it as a free parameter.

	Value	Ref
Moretto et. al.	$2\pi k \frac{R_1 R_2}{R_1 + R_2}, \quad k = 10^{21}(\text{unit/fm/s})$	Phys. Lett. B (1975) 58 26
Zagrebaev et. al.	$0.1 * 10^{22}(\text{unit/s})$	Nucl. Phys. A (2015) 944, 257.
Karpov et. al.	$5 * \text{A}_{\text{tot}} * (\text{T/MeV}) * 10^{16}(\text{unit/s})$	Eur. Phys. J. A (2022) 58:41

This value is adopted in the model

Microscopic transition probabilities

Moretto L G and Sventek J S 1975 Phys. Lett. B 58 26

$$\dot{\phi}_z = \sum_{z'} (\Lambda_{zz'}\phi_{z'} - \Lambda_{z'z}\phi_z)$$

The macroscopic transition probabilities can be written in terms of the microscopic transition probabilities and of the level densities of the macroscopic states:

$$\Lambda_{zz'} = \lambda_{zz'}\rho_z; \quad \Lambda_{z'z} = \lambda_{z'z}\rho_{z'}; \quad \lambda_{zz'} = \lambda_{z'z}. \tag{2}$$

Since $\Lambda_{zz'}$ must be of the order of V_F/D (where D is a typical linear size of the system and V_F is the Fermi velocity of the nucleons), the $\lambda_{zz'}$ decrease as the level densities increase. For lack of better knowledge, we assume:

$$\lambda_{zz'} = \frac{\lambda_0}{[\rho_z \rho_{z'}]^{1/2}}. \tag{4}$$

Transition from a state S to another space S'

Macroscopic transition probabilities

Microscopic transition probabilities

$$W^{\pm} = \lambda_0 \sqrt{\frac{\rho(S \pm 1)}{\rho(S)}}$$

$$\rho_{\mathbf{S}} \propto \exp(-V(\mathbf{S})/T).$$

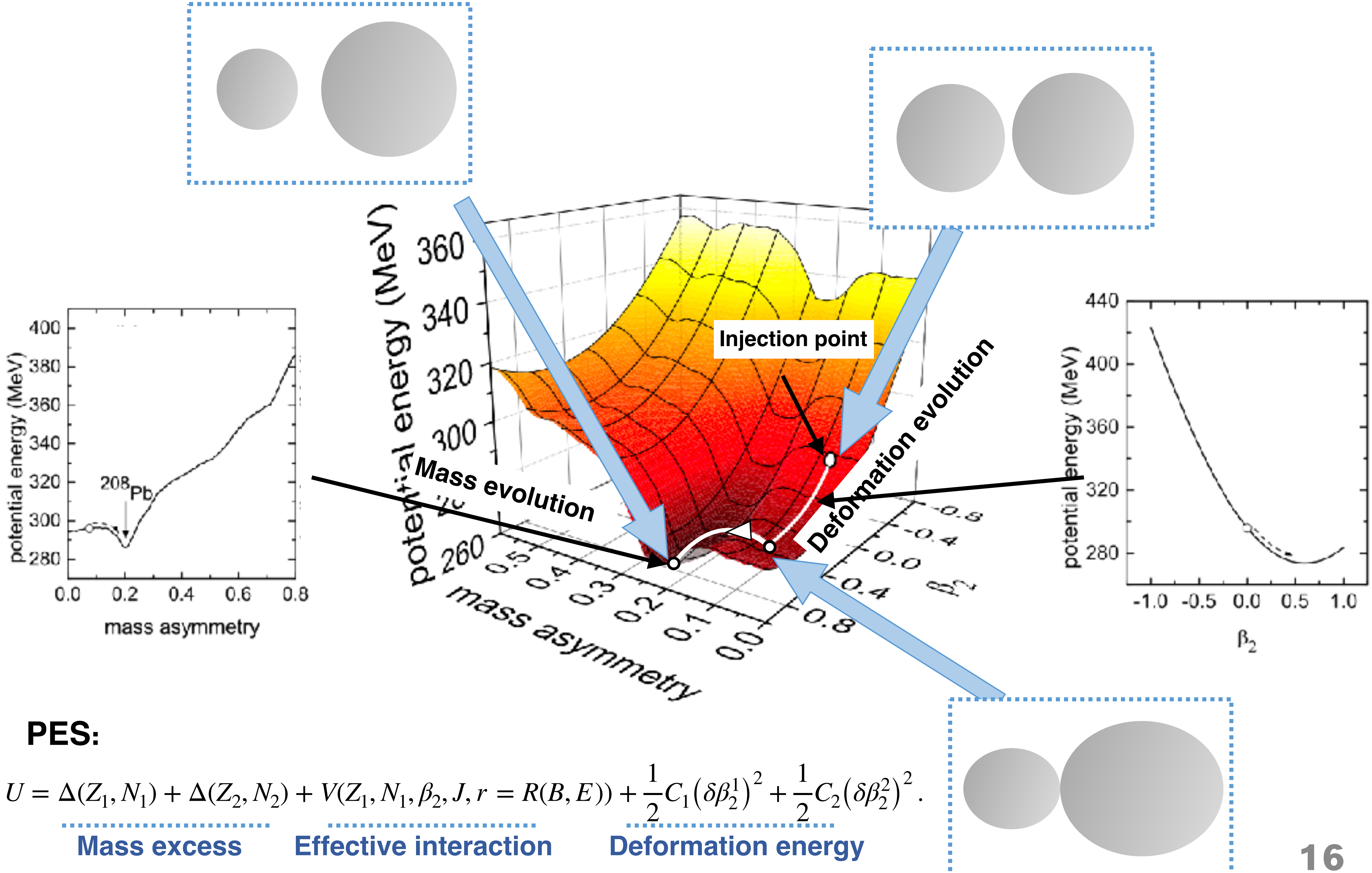
$$W(\mathbf{S}, \mathbf{S}') = \lambda_{\mathbf{S},\mathbf{S}'} \cdot \rho_{\mathbf{S}'} = \lambda_0 \frac{1}{\sqrt{\rho_{\mathbf{S}}\rho_{\mathbf{S}'}}}\rho_{\mathbf{S}'} = \lambda_0 \sqrt{\frac{\rho_{\mathbf{S}'}}{\rho_{\mathbf{S}}}}$$

$$= \lambda_0 \sqrt{\frac{\exp(-V(\mathbf{S}')/T)}{\exp(-V(\mathbf{S})/T)}}$$

$$= \lambda_0 \sqrt{\exp\left(\frac{V(\mathbf{S}) - V(\mathbf{S}')}{T}\right)}$$

$$= \lambda_0 \exp\left(\frac{V(\mathbf{S}) - V(\mathbf{S}')}{2T}\right)$$

Potential energy Surface (PES)



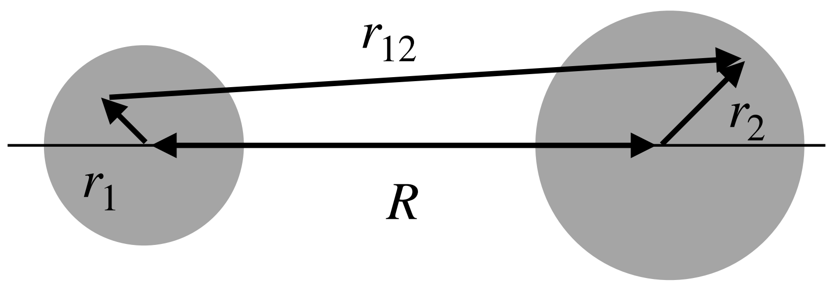
PES:

$$U = \Delta(Z_1, N_1) + \Delta(Z_2, N_2) + V(Z_1, N_1, \beta_2, J, r = R(B, E)) + \frac{1}{2}C_1\left(\delta\beta_2^1\right)^2 + \frac{1}{2}C_2\left(\delta\beta_2^2\right)^2 .$$

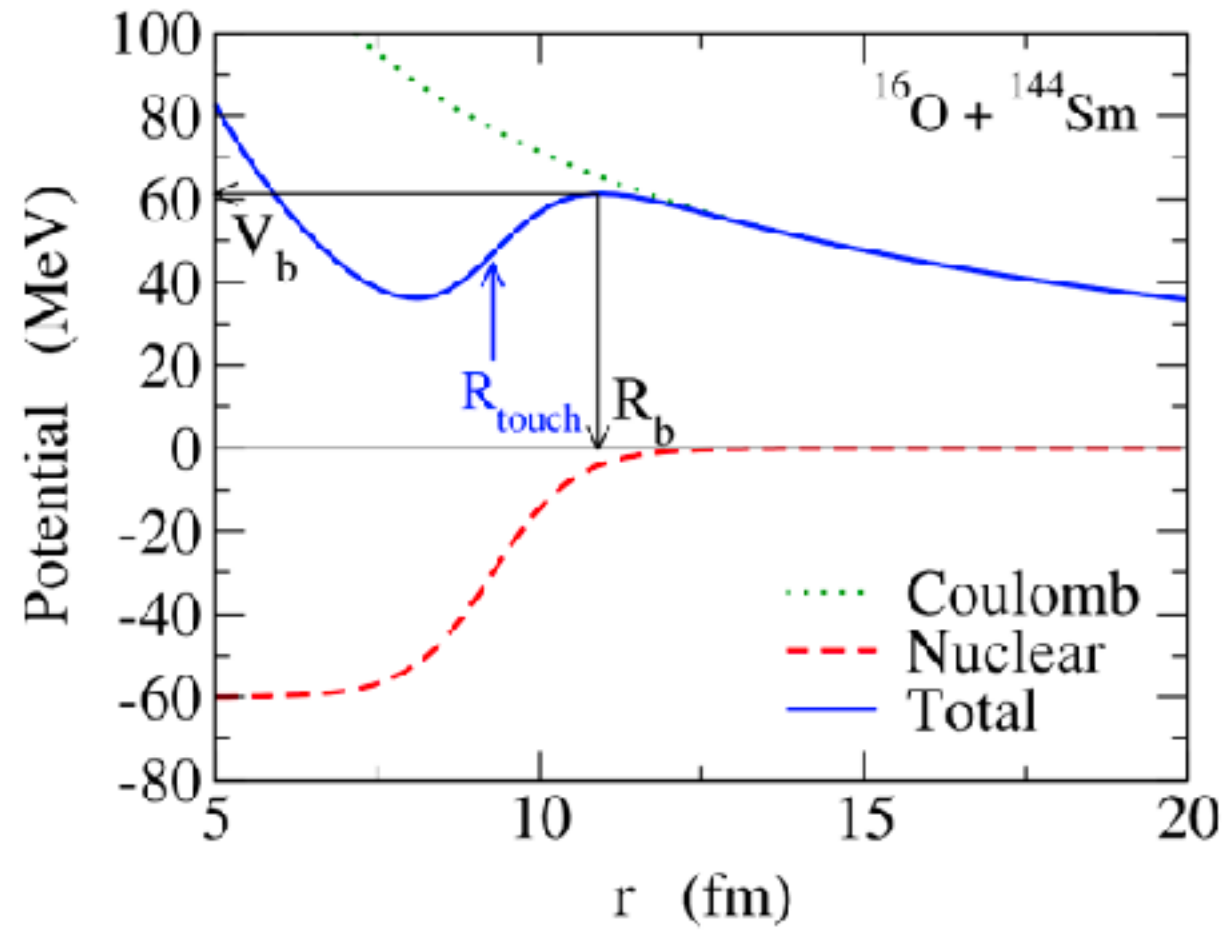
Mass excess Effective interaction Deformation energy

Nuclei-Nuclei Interaction

$$V(Z_1, A_1, Z_2, A_2, \beta(\beta_1, \beta_2), R(E, b))$$



$$V(R) = V_{\mathrm{C}}(R) + V_{\mathrm{N}}(R) + V_{\mathrm{cent}}(R)$$



- **Coulomb potential:**

$$V_{\mathrm{C}}(R, \theta_i) = \frac{Z_1 Z_2 e^2}{R} + \left(\frac{9}{20\pi}\right)^{1/2} \left(\frac{Z_1 Z_2 e^2}{R^3}\right) \times \sum_{i=1}^{2} R_i^2 \beta_2^{(i)} P_2(\cos\theta_i) + \left(\frac{3}{7\pi}\right) \left(\frac{Z_1 Z_2 e^2}{R^3}\right) \sum_{i=1}^{2} R_i^2 \left[\beta_2^{(i)} P_2(\cos\theta_i)\right]^2 .$$

C. Y. Wong, Phys. Rev. Lett. 31, 766 (1973).

- **Nuclear potential:**

$$U(\mathbf{r}_1 - \mathbf{r}_2) = \int \rho_1(\mathbf{r}_1)\rho_2(\mathbf{r}_2) v(\mathbf{r}_1 - \mathbf{r}_2 - \mathbf{R}) d\mathbf{r}_1 d\mathbf{r}_2$$

M3Y type interactions

Qingfeng. Li et al Eur. Phys. J. A 24, 223–229 (2005)

Probability of a nucleon transfer

$$W^{\pm} = \lambda_0 \sqrt{\frac{\rho(A \pm 1)}{\rho(A)}} P_{tr}(R, \beta, A \to A \pm 1)$$

Probability of a nucleon transfer

$$P_{tr}(R, \beta, A \to A \pm 1) = \exp(-2k[r_c - r_0])$$

中子 $k = \sqrt{M(-\epsilon_F)/2\hbar^2} + \sqrt{M(-\epsilon_F')/2\hbar^2}$ **[1/fm]**

质子 $k = \sqrt{M(-\epsilon_F + Z_T e^2/R_T)/2\hbar^2} + \sqrt{M(-\epsilon_F' + Z_P e^2/R_P)/2\hbar^2}$ **[1/fm]**

Z. Phys.A - AtomicNuclei 326,463-481 (1987)

Brink [25]. We will consider the barrier to be static and we assume that the nucleon tunnels between two levels with the same energy (binding energy E_B). In a WKB and first order approximation the probability for the transmission through the barrier between two points x_1 and x_2 is given by (see Fig. 16)

$$P \approx \exp(-2Q) \tag{14}$$

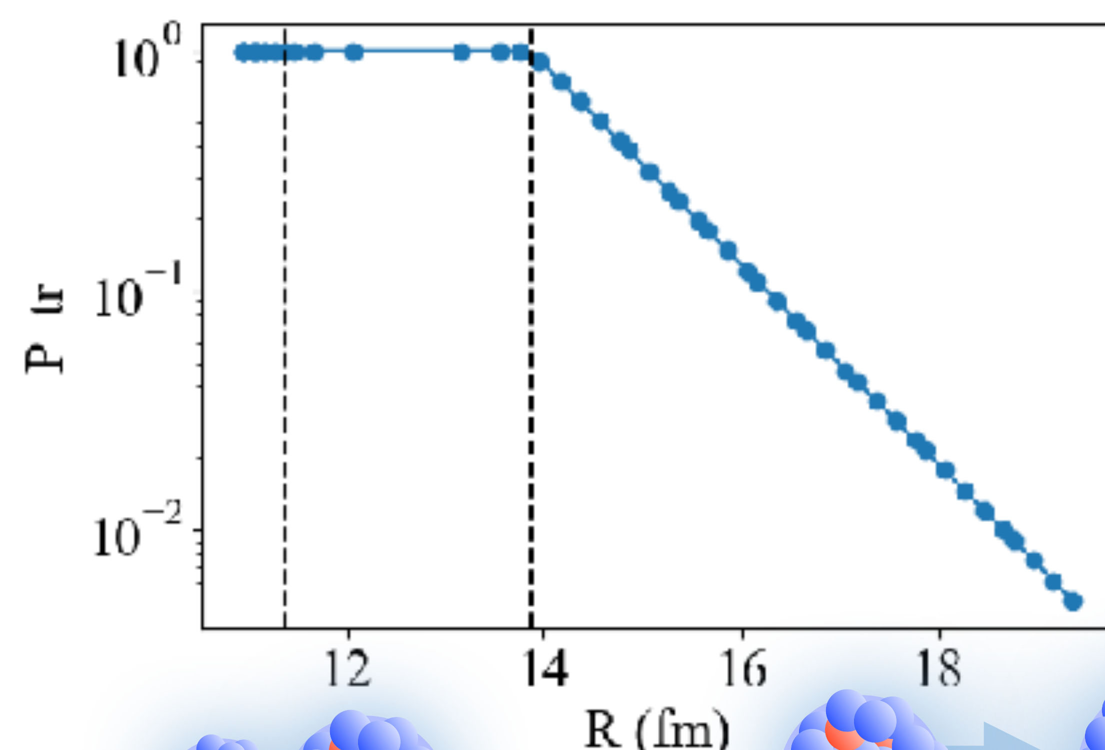
with

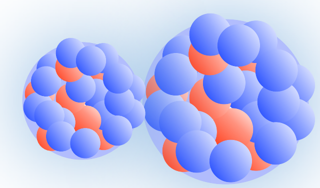
$$Q = \int_{x_1}^{x_2} \left(\frac{2m}{h^2}(V(x) - E_B)\right)^{1/2} \mathrm{d}x; \quad x_d = x_1 - x_2. \tag{15}$$

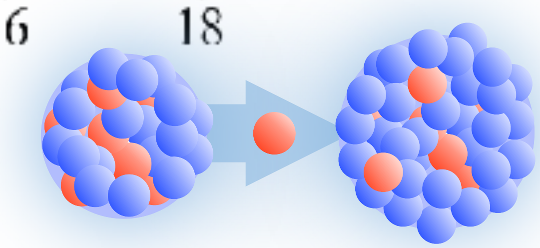
$V(x)$ is the potential barrier and x_d is the distance between the two potential edges x_1 and x_2.

If we assume $V(x)$ to be constant ($\equiv 0$), we immediately obtain with $\alpha = (2m/h^2 |E_B|)^{1/2}$, the exponential dependence used as an approximation to our data

$$P \simeq \exp(-2\alpha x_d). \tag{16}$$



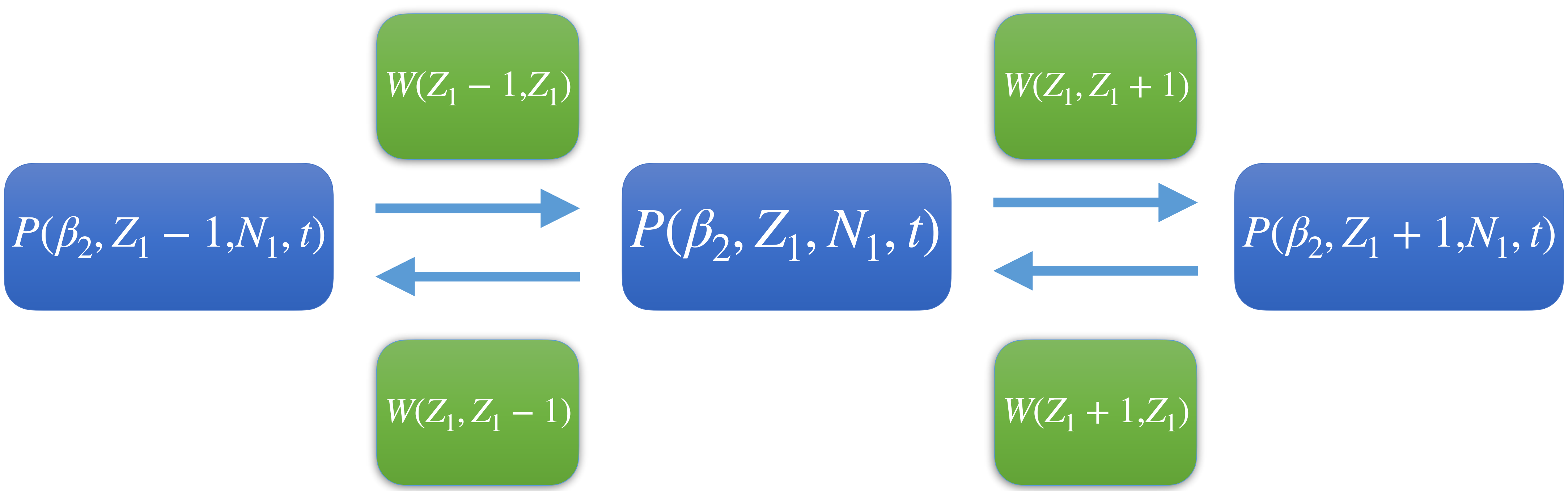




Master Equation

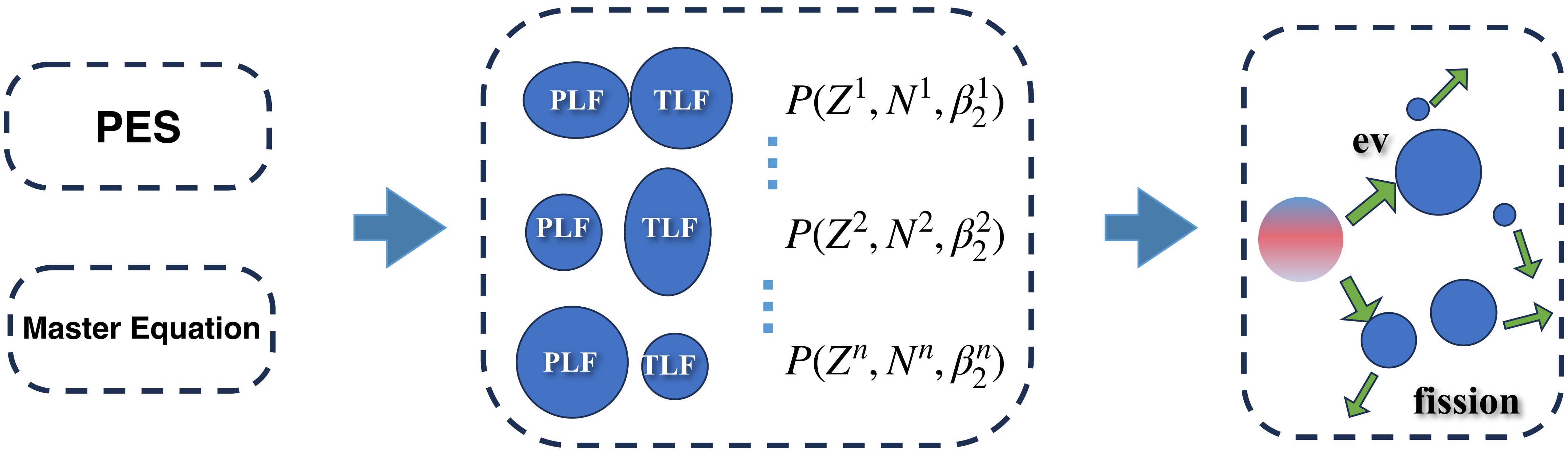
If this space contains the proton Z, neutron N, and the deformation β of the system,

$$\begin{aligned}\frac{\partial P(\beta_2, Z_1, N_1, t)}{\partial t} &= W(Z_1-1, Z_1)P(\beta_2, Z_1-1, N_1, t) + W(Z_1+1, Z_1)P(\beta_2, Z_1+1, N_1, t)\\ &+ W(\beta_2-1, \beta_2)P(\beta_2-1, Z_1, N_1, t) + W(\beta_2+1, \beta_2)P(\beta_2+1, Z_1, N_1, t)\\ &+ W(N_1-1, N_1)P(\beta_2, Z_1, N_1-1, t) + W(N_1+1, N_1)P(\beta_2, Z_1, N_1+1, t)\\ &-[W(\beta_2, \beta_2+1) + W(\beta_2, \beta_2-1)]P(\beta_2, Z_1, N_1, t)\\ &-[W(Z_1, Z_1+1) + W(Z_1, Z_1-1)]P(\beta_2, Z_1, N_1, t)\\ &-[W(N_1, N_1+1) + W(N_1, N_1-1)]P(\beta_2, Z_1, N_1, t)\,.\end{aligned}$$



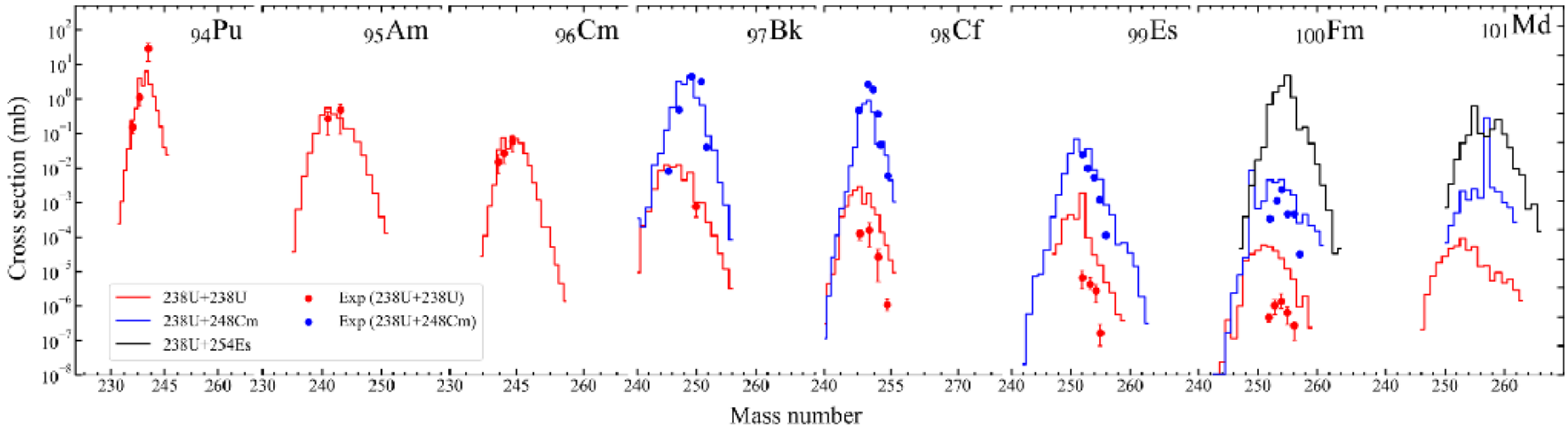
If this space contains the proton Z, neutron N, and the deformation β of the system,

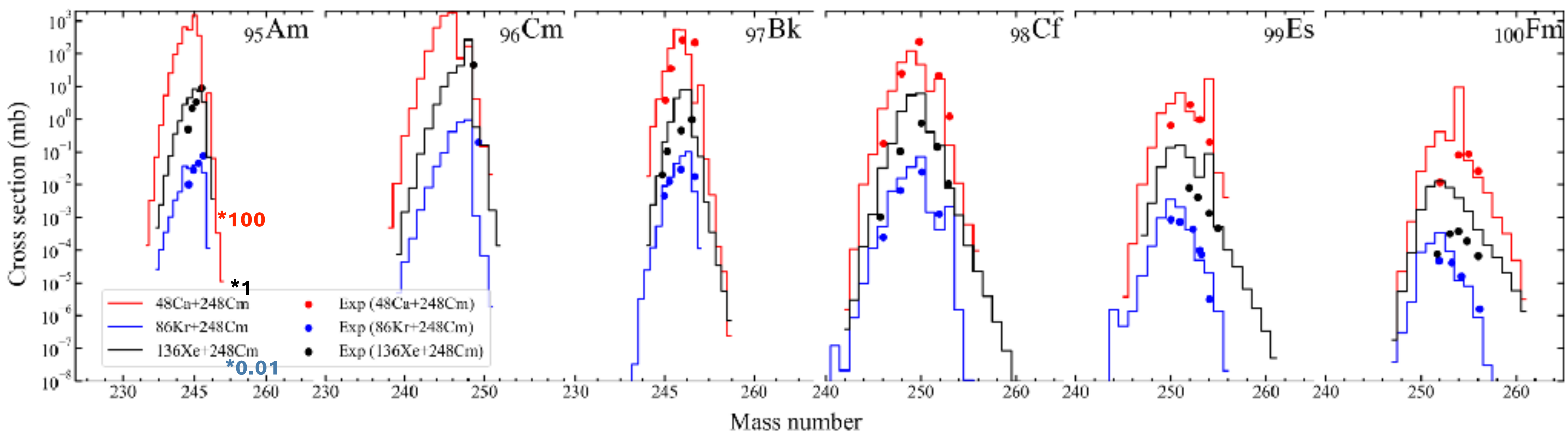
$$
\begin{aligned}
\frac{\partial P(\beta_2, Z_1, N_1, t)}{\partial t} = {} & W(\beta_2 - 1, \beta_2) P(\beta_2 - 1, Z_1, N_1, t) + W(\beta_2 + 1, \beta_2) P(\beta_2 + 1, Z_1, N_1, t) \\
& + W(Z_1 - 1, Z_1) P(\beta_2, Z_1 - 1, N_1, t) + W(Z_1 + 1, Z_1) P(\beta_2, Z_1 + 1, N_1, t) \\
& + W(N_1 - 1, N_1) P(\beta_2, Z_1, N_1 - 1, t) + W(N_1 + 1, N_1) P(\beta_2, Z_1, N_1 + 1, t) \\
& - [W(\beta_2, \beta_2 + 1) + W(\beta_2, \beta_2 - 1)] P(\beta_2, Z_1, N_1, t) \\
& - [W(Z_1, Z_1 + 1) + W(Z_1, Z_1 - 1)] P(\beta_2, Z_1, N_1, t) \\
& - [W(N_1, N_1 + 1) + W(N_1, N_1 - 1)] P(\beta_2, Z_1, N_1, t) \,.
\end{aligned}
$$



Linked to Statistical model

$$
\sigma_{\mathrm{pr}}(Z_1, N_1, E_{\mathrm{c.m.}}) = \frac{\pi \hbar^2}{2\mu E_{\mathrm{c.m.}}} \sum_{J=0}^{J_{\max}} (2J+1) T_{\mathrm{cap}}(J, E_{\mathrm{c.m.}}) \times \sum_{\beta_2} P(Z_1, N_1, \beta_2, J, E_{\mathrm{c.m.}}, \tau_{\mathrm{int}}) \times W_{\mathrm{sur}}(Z_1, N_1, J, E^*),
$$



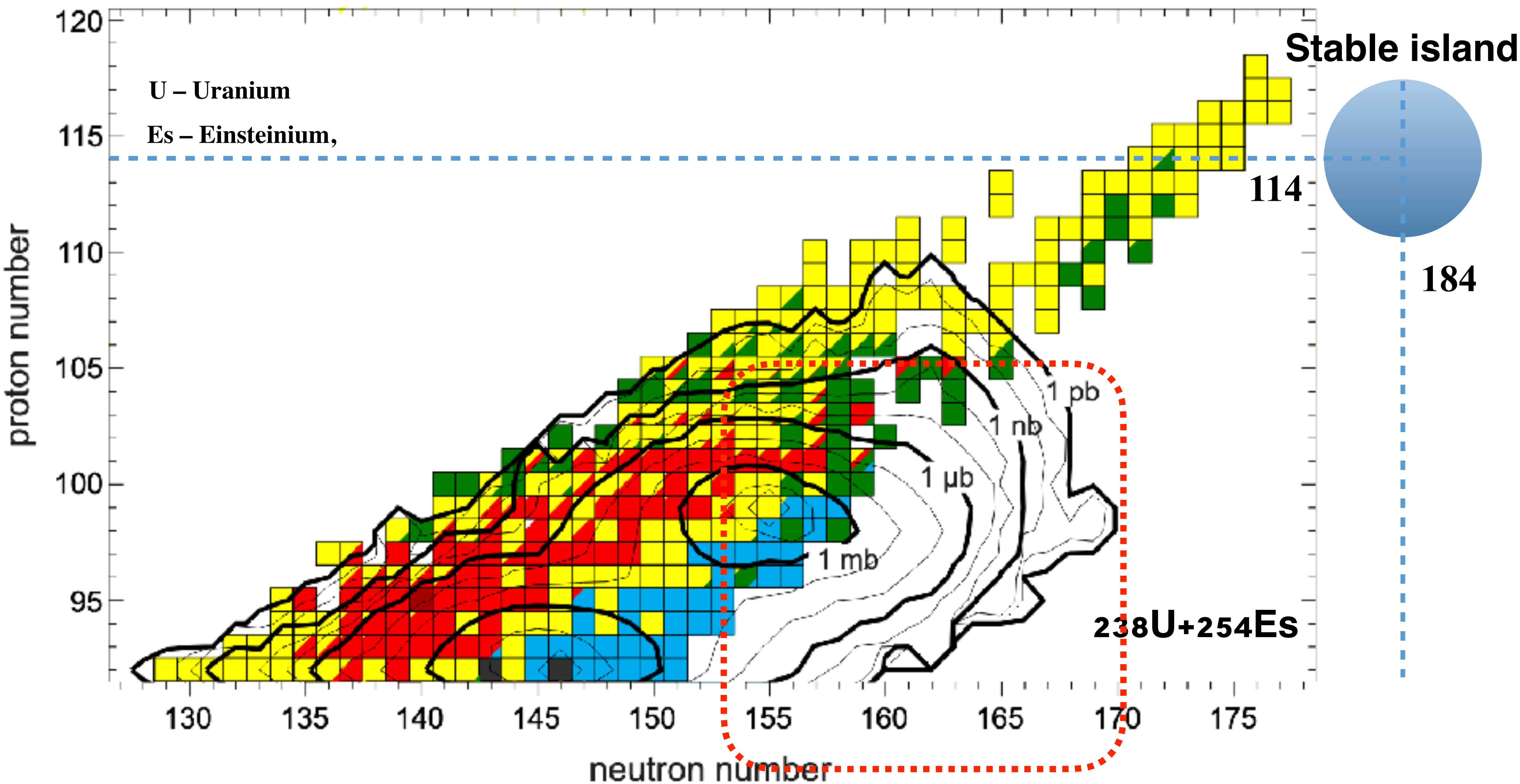


Pu – Plutonium, Am – Americium, Cm – Curium, Bk – Berkelium, Cf – Californium, Es – Einsteinium, Fm – Fermium, Md – Mendelevium

Ref: Phys. Rev. Lett. 41, 469 (1978).
Phys. Rev. Lett. 48, 852 (1982).
Phys. Rev. C 33, 1315 (1986).
Phys. Rev. C 88, 054615 (2013).
Phys. Rev. C 31, 1763 (1985).

$$\sigma_{\rm pr}(Z_1, N_1, E_{\rm c.m.}) = \frac{\pi \hbar^2}{2\mu E_{\rm c.m.}} \sum_{J=0}^{J_{\rm max}} (2J+1) T_{\rm cap}(J, E_{\rm c.m.}) \times \sum_{\beta_2} P(Z_1, N_1, \beta_2, J, E_{\rm c.m.}, \tau_{\rm int}) \times W_{\rm sur}(Z_1, N_1, J, E^*),$$

Prediction



unknown superheavy nuclides in MNT collisions of actinides is rather limited.

unknown neutron-enriched isotopes of elements from U to Md can be produced

U – Uranium **Md – Mendelevium**

Summary

The model remains a semi-classical approach, striking a balance between computational resource and physical realism.

This makes it especially suited for large-scale surveys of nuclear reactions. That helps to search for the optimal experimental condition (systems, energy, angular distribution).

There remain open development.

One possible direction is the integration of microscopic inputs,

such as single-particle energy levels and nucleon density distributions from constrained density functional theory, to provide more accurate potential energy surfaces and transition rates.

2. 为何“势能”项出现在 Boltzmann 因子里

1. **势能决定微观态的能量高低**
    - 每一个具体的核构型 $\mathbf{S}$（包括形状、单粒子排布等）对应一个势能 $V(\mathbf{S})$。
    - 如果我们关心“在特定形状 $\mathbf{S}$ 出现的相对权重”，就需要看所有微观态中有多少状态落在这一形状附近，以及这些态的能量（主要由势能决定）有多高。
    - 根据 Boltzmann 分布，**能量越高的构型被占据的概率越小**，比例正比于 $\exp[-V(\mathbf{S})/T]$。
2. **分离动能与势能的近似**
    - 在许多模型（如广义蒙特卡洛、形状自由度统计）中，我们只关心形状变量 $\mathbf{S}$ 的统计权重，把所有动能自由度积分掉，剩下一个势能 dependent 的权重：

$$\int dK \ \exp\big(-(K+V(\mathbf{S}))/T\big) = \Big[\int dK \, e^{-K/T}\Big] \ e^{-V(\mathbf{S})/T} \ \propto \ e^{-V(\mathbf{S})/T}.$$

    - 这里的 $\int dK$ 就是把所有动能态密度（本身与动能有关的部分）累加（或积分）掉，留下一个仅依赖于势能的指数因子。
3. **有效态密度的定义**

    当我们讲 “$\rho_{\mathbf{S}} \propto \exp[-V(\mathbf{S})/T]$” 时，严格来说指的是 **在固定宏观构型 $\mathbf{S}$ 下，所有可用微观态（包含动能）的总数加权后，和一个势能项有关。**

$$\rho_{\mathbf{S}} = \int dK \ g_{\mathbf{S}}(K) \ \exp\big(-K/T\big) \ \propto \ \exp\big(-V(\mathbf{S})/T\big),$$