



Istituto Nazionale di Fisica Nucleare



Nuclear Physics Applied to the Production of Innovative Radiopharmaceuticals

PART ONE

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Demand for radionuclides and cyclotrons (iaea)

It is rapidly growing!
Cyclotrons:
2008:
 $\rightarrow \simeq 700$
2015:
 $\rightarrow 1218$

TABLE 2.1. THE RADIOISOTOPES THAT HAVE BEEN USED AS TRACERS IN THE PHYSICAL AND BIOLOGICAL SCIENCES

| Isotope | Isotope | Isotope |
|--------------|--------------|----------------|
| Actinium-225 | Fluorine-18 | Oxygen-15 |
| Arsenic-73 | Gallium-67 | Palladium-103 |
| Arsenic-74 | Germanium-68 | Sodium-22 |
| Astatine-211 | Indium-110 | Strontium-82 |
| Beryllium-7 | Indium-111 | Technetium-94m |
| Bismuth-213 | Indium-114m | Thallium-201 |
| Bromine-75 | Iodine-120g | Tungsten-178 |
| Bromine-76 | Iodine-121 | Vanadium-48 |
| Bromine-77 | Iodine-123 | Xenon-122 |
| Cadmium-109 | Iodine-124 | Xenon-127 |
| Carbon-11 | Iron-52 | Yttrium-86 |
| Chlorine-34m | Iron-55 | Yttrium-88 |
| Cobalt-55 | Krypton-81m | Zinc-62 |
| Cobalt-57 | Lead-201 | Zinc-63 |
| Copper-61 | Lead-203 | Zirconium-89 |
| Copper-64 | Mercury-195m | |
| Copper-67 | Nitrogen-13 | |

Summary

Introduction to Scattering concepts

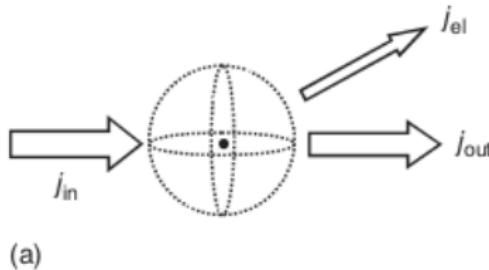
Coupled-Channel processes

Compound-nucleus reactions

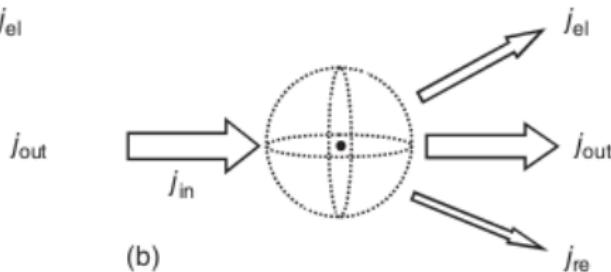
Pre-equilibrium reactions

INTRODUCTION

Importance of scattering and reaction methods for applications in the production of radionuclides!



(a)



(b)

scattering

scattering & reactions

J : incident flux (number of incident particles crossing unit surface in unit time).

\mathcal{N} : number of particles scattered per unit time into the solid angle $d\Omega$ in the direction $\Omega(\theta, \phi)$

N : number of nuclear scattering centers, of which the target is made up

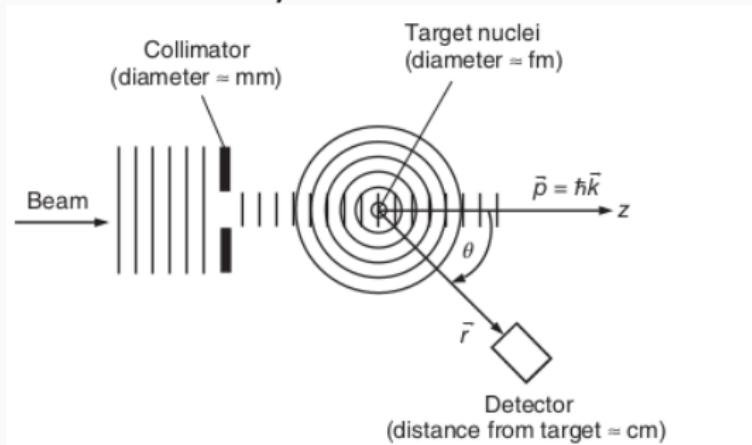
$$\mathcal{N} = JN\sigma(\Omega)d\Omega \quad (1)$$

The area $\sigma(\Omega)$ is the differential scattering cross section of the incident particle by the scattering center. Integrating over angles we get the total scattering cross section,

$$\sigma_{tot} = \int \sigma(\Omega)d\Omega \quad (2)$$

Nuclear physics (size $\simeq 10^{-13}$ to $\simeq 10^{-12}$ cm)
cross sections are in barns:

$$1 \text{ barn} = 10^{-24} \text{ cm}^2, 1 \text{ mb} = 10^{-27} \text{ cm}^2.$$



wf in the asymptotic limit $r \rightarrow \infty$

$$\psi(\mathbf{r}) \rightarrow e^{i\mathbf{k} \cdot \mathbf{r}} + f(\Omega) \frac{e^{ikr}}{r} \quad \hbar k = p$$

$$\sigma(\Omega) = |f(\Omega)|^2 \quad (3)$$

scattering amplitude :

$$f(\theta) = \frac{1}{2ik} \sum_0^{\infty} (2\ell + 1)(S_{\ell} - 1)P_{\ell}(\cos \theta) \quad (4)$$

The s-matrix S_{ℓ} can be determined by the asymptotic behaviour of the decomposed Schrödinger equation

$$\left(\frac{d^2}{dr^2} - \frac{\ell(\ell + 1)}{r^2} + 2mV(r) + k^2 \right) \psi_{\ell}(r) = 0$$

in the limit $r \rightarrow \infty$

$$\psi_{\ell}(r) \rightarrow \frac{i}{2} (h_{\ell}^{-}(kr) - S_{\ell} h_{\ell}^{+}(kr))$$

S-matrix: the response of the target

The S-matrix measures the response of the target: a real potential cannot create or destroy particles, it can change only the phase of the outgoing wave, not the modulus

$$S_\ell = e^{2i\delta_\ell}$$

The total elastic cross section becomes:

$$\sigma_{el} = \frac{\pi}{k^2} \sum_0^\infty (2\ell + 1) |S_\ell - 1|^2$$

No nuclear reactions here!

loss of elastic flux: $|S_\ell| < 1$

$$\sigma_{el} = \frac{\pi}{k^2} \sum_0^{\infty} (2\ell + 1) |S_\ell - 1|^2$$

$$\sigma_r = \frac{\pi}{k^2} \sum_0^{\infty} (2\ell + 1) (1 - |S_\ell|^2)$$

$$\sigma_{tot} = \sigma_{el} + \sigma_r = \frac{\pi}{k^2} \sum_0^{\infty} (2\ell + 1) (1 - \Re S_\ell)$$

The transmission coefficient T_ℓ is

$$T_\ell = 1 - |S_\ell|^2 \tag{5}$$

adding spin 1/2 projectile and spin-orbit complexity

$$S_\ell^\pm = S_\ell^{J=\ell\pm 1/2}$$

$$\sigma_{el} = \frac{\pi}{k^2} \sum_0^{\infty} [(\ell + 1) |S_\ell^+ - 1|^2 + \ell |S_\ell^- - 1|^2]$$

$$\sigma_r = \frac{\pi}{k^2} \sum_0^{\infty} [(\ell + 1)(1 - |S_\ell^+|^2) + \ell(1 - |S_\ell^-|^2)]$$

with transmission coefficients T_ℓ^\pm

$$T_\ell^\pm = 1 - |S_\ell^\pm|^2 \tag{6}$$

Potential must be complex for reactions.

Optical Potential $U(r)$

Coulomb part: $V_C(r) = Z_1 Z_2 e^2 / r$

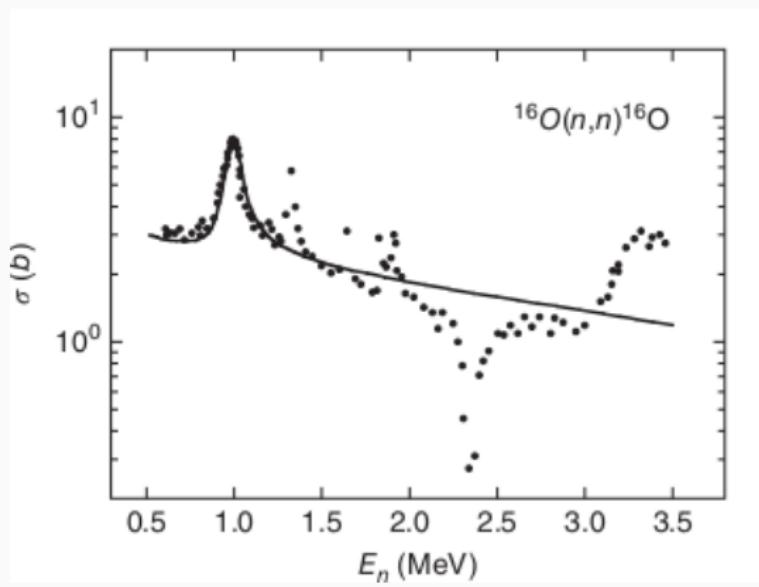
Real Nuclear part: $V(r)$ for nuclear attraction

Imaginary Part $W(r)$: for reaction to occur

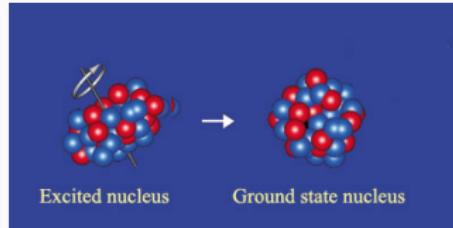
Spin-Orbit part: $V_{SO}(r)$ Important ingredient of nuclear force and for polarization data

$$U(r) = V_C(r) + V(r) + iW(r) + V_{SO}(r)$$

the optical potential averages



The optical potential averages over many narrow structures. It cannot cope with narrow resonances...



The coupled-channel **Schrödinger** equation:

$$(H_{0c} - E_c) \psi_c(r) = - \sum_{c'=1}^{\sigma} \int_0^{\infty} U_{cc'}(r, r') \psi_{c'}(r') dr' , \quad c = 1, 2, \dots, C .$$

the **Free** Hamiltonian (or Coulomb-distorted) :

$$H_{0c} = -\frac{1}{\mu} \left(\frac{d^2}{dr^2} - \frac{l(l+1)}{r^2} \right) + V_c(r) ,$$

the **Interacting** Hamiltonian:

$$U_{cc'}(r, r') = V_{cc'}(r) \delta(r - r') + K_{cc'}(r, r') .$$

Model of CC interaction with collective excitations (quadrupole, etc).

$$V_{cc'}(r) = \sum_{n=C,LS,LL,SI} V_n <(\ell s)jI|\mathcal{O}_n f_n(r, R, \theta_{\mathbf{r}, \mathbf{R}})|(\ell' s')j'I>$$

The deformation parameter ($R = R_0(1 + \beta_2 P_2(\theta))$) generates the CC dynamics. Pauli principle requires a non-local term:

$$\mathcal{V}_{cc'}(r, r') = V_{cc'}(r)\delta(r - r') + \delta_{cc'} \sum_i \lambda_{ci} A_{ci}(r)A_{ci}(r')$$

$A_{ci}(r)$ are the (Pauli-forbidden, for $\lambda \rightarrow +\infty$) deep bound states.

■ Sturmians: a different QM

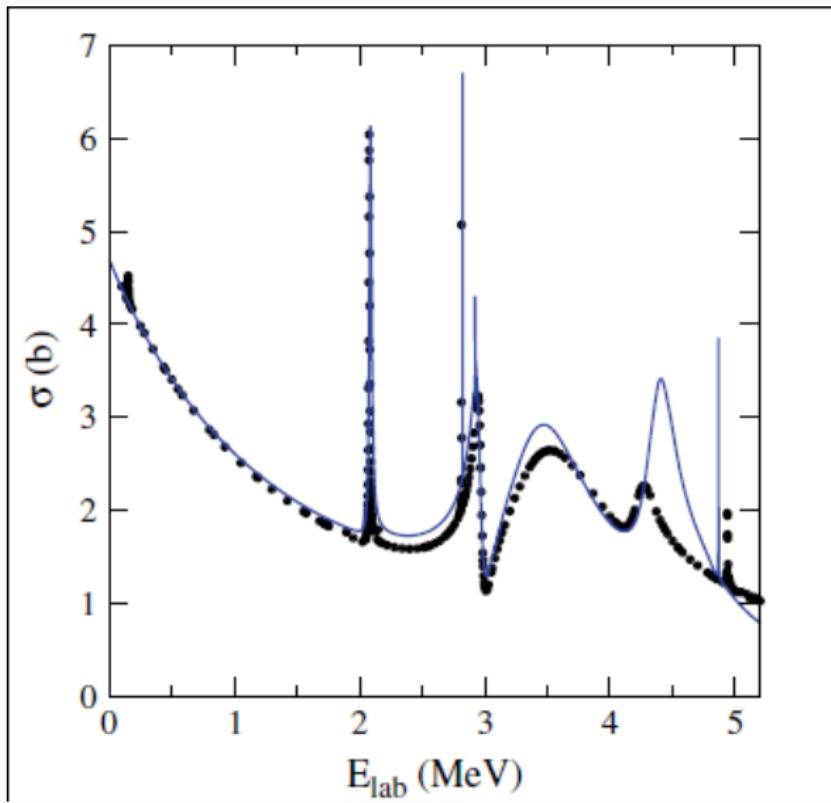
Sturmians are the eigensolutions of:

$$(E - H_o)\Phi_i(E) = \frac{V}{\eta_i(E)}\Phi_i(E),$$

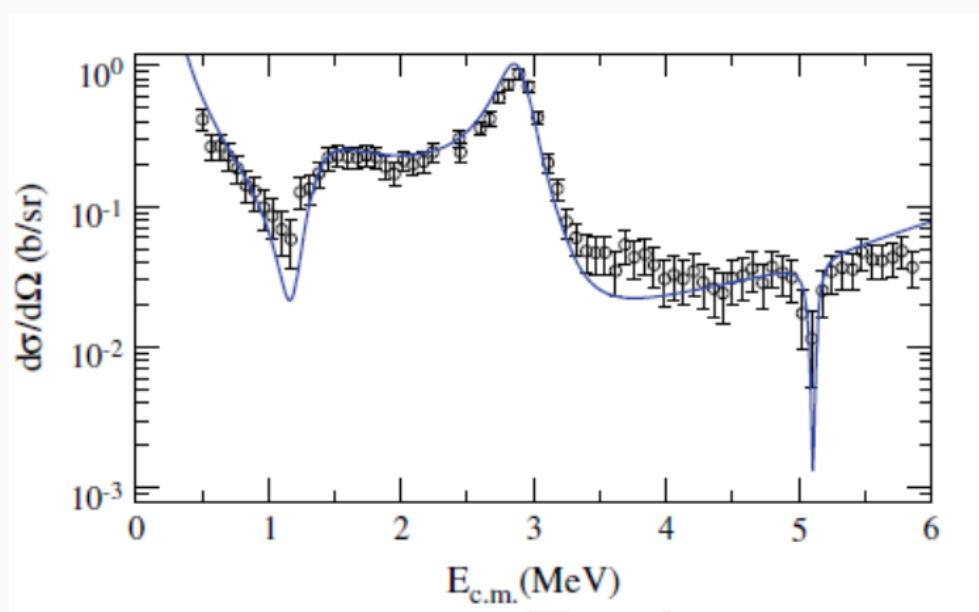
where E is a parameter. The eigenvalue η_i is the potential scale. With the form factor in momentum space $\hat{\chi}_{ci}(E^{(+)}; k) = \langle k, c | V | \Phi_i(E) \rangle$, the CC S-matrix is

$$S_{cc'}(E) = \delta_{cc'} - i\pi \sqrt{k_c k'_c} \sum_i \hat{\chi}_{ci}(E^{(+)}; k_c) \frac{1}{1 - \eta_i(E^{(+)})} \hat{\chi}_{c'i}(E^{(+)}; k_{c'})$$

n - ^{12}C Total elastic cross section



^{14}O -*p* differential cross-section @ 180°



Compound Nucleus

CN requires statistical equilibrium of all the nucleons, slow process, the last to occur.

$$\sigma_{react} = \sigma_{dir} + \sigma_{pre-eq} + \sigma_{\text{CN}}$$

$$N_0 \rightarrow N_0 - dN_D \rightarrow N_0 - dN_D - dN_{PE} = N$$

$$Z_0 \rightarrow Z_0 - dZ_D \rightarrow Z_0 - dZ_D - dZ_{PE} = Z$$

$$E_0^* \rightarrow E_0^* - dE_D^* \rightarrow E_0^* - dE_D^* - dE_{PE}^* = E^*$$

$$J_0 \rightarrow J_0 - dJ_D \rightarrow J_0 - dJ_D - dJ_{PE} = J$$

CN basics

1. Continuum overlapping levels
2. Independence **initial**/exit channel

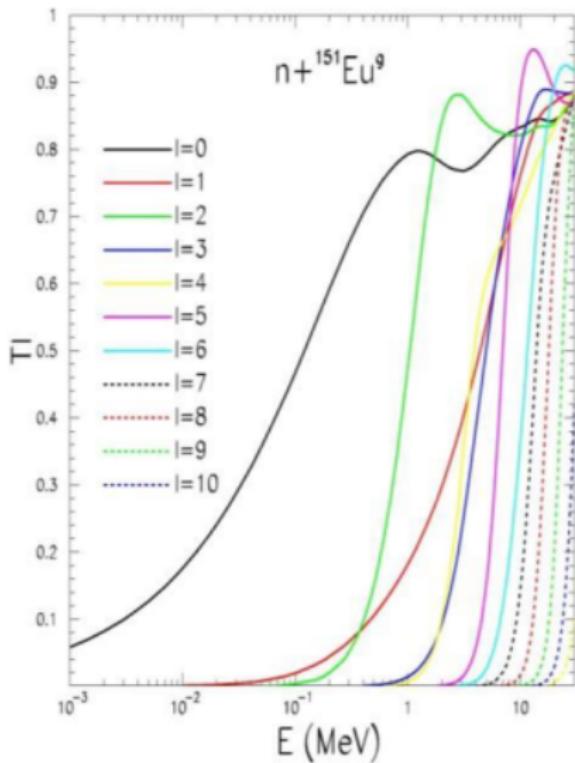
$$\sigma_{ab} = \sigma_a^{CN} P_b$$

Hauser-Feshbach formula:

$$\sigma_{ab} = \frac{\pi T_a}{k_a^2} \frac{T_b}{\sum_c T_c}$$

Need transmission coefficients (from optical potentials)

Transmission coefficients



CN Basic 2, complexities

Loop over angular momentum and parity

$$\sigma_{ab} = \frac{\pi}{k_a^2} \sum_{J,\pi} \sum_{\alpha,\beta} \frac{2J+1}{(2s+1)(2I+1)} \frac{T_a^{J,\pi}(\alpha) T_b^{J,\pi}(\beta)}{\sum_{\delta} T_d^{J,\pi}(\delta)}$$

Adding the WIDTH-FLUCTUATION CORRECTION

$$\sigma_{ab} = \frac{\pi}{k_a^2} \sum_{J,\pi} \sum_{\alpha,\beta} \frac{2J+1}{(2s+1)(2I+1)} \frac{T_a^{J,\pi}(\alpha) T_b^{J,\pi}(\beta)}{\sum_{\delta} T_d^{J,\pi}(\delta)} \mathbf{W}_{\alpha,\beta}$$

Adding the DENSITY OF RESIDUAL NUCLEUS LEVELS

$$\sigma_{ab} = \frac{\pi}{k_a^2} \sum_{J,\pi} \sum_{\alpha,\beta} \frac{2J+1}{(2s+1)(2I+1)} \frac{T_a^{J,\pi}(\alpha) < \mathbf{T}_b^{J,\pi}(\beta) >}{\sum_{\delta} < \mathbf{T}_d^{J,\pi}(\delta) >} W_{\alpha,\beta}$$

Width fluctuation correction

resonance cross sections integrated and averaged over an energy spread

$$\langle \sigma_{ab} \rangle = \frac{\pi}{k_a^2} \frac{2\pi}{D} \left\langle \frac{\Gamma_a \Gamma_b}{\Gamma_{tot}} \right\rangle .$$

Since

$$T_a \simeq \frac{2\pi \langle \Gamma_a \rangle}{D} .$$

This leads to

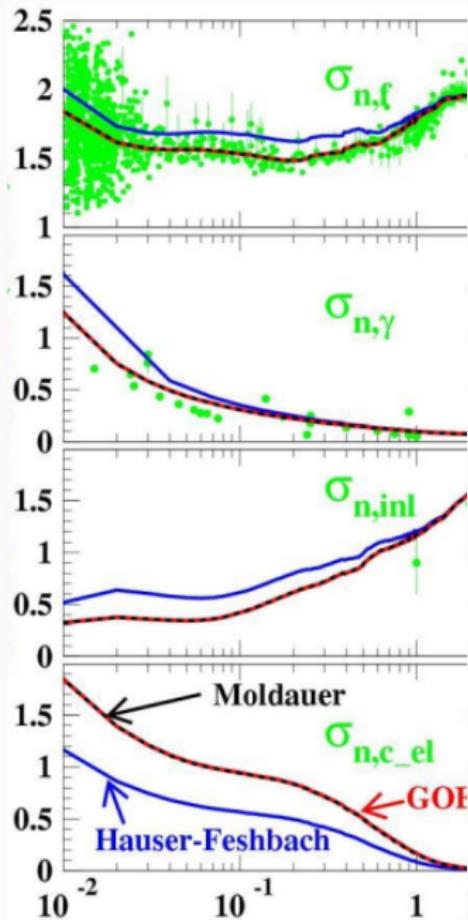
$$\langle \sigma_{ab} \rangle = \frac{\pi}{k_a^2} \frac{T_a T_b}{\sum_c T_c} \mathbf{W}_{ab}$$

$$\mathbf{W}_{ab} = \left\langle \frac{\Gamma_a \Gamma_b}{\Gamma_{tot}} \right\rangle \frac{\langle \Gamma_{tot} \rangle}{\langle \Gamma_a \rangle \langle \Gamma_b \rangle}$$

width-fluctuation
correction

approximate:
Moldauer 1975

"exact": GOE 1985
Gauss Orth. Ensemble



Averaging over Residual Nucleus Density Levels

Emission → discrete level with energy E_b

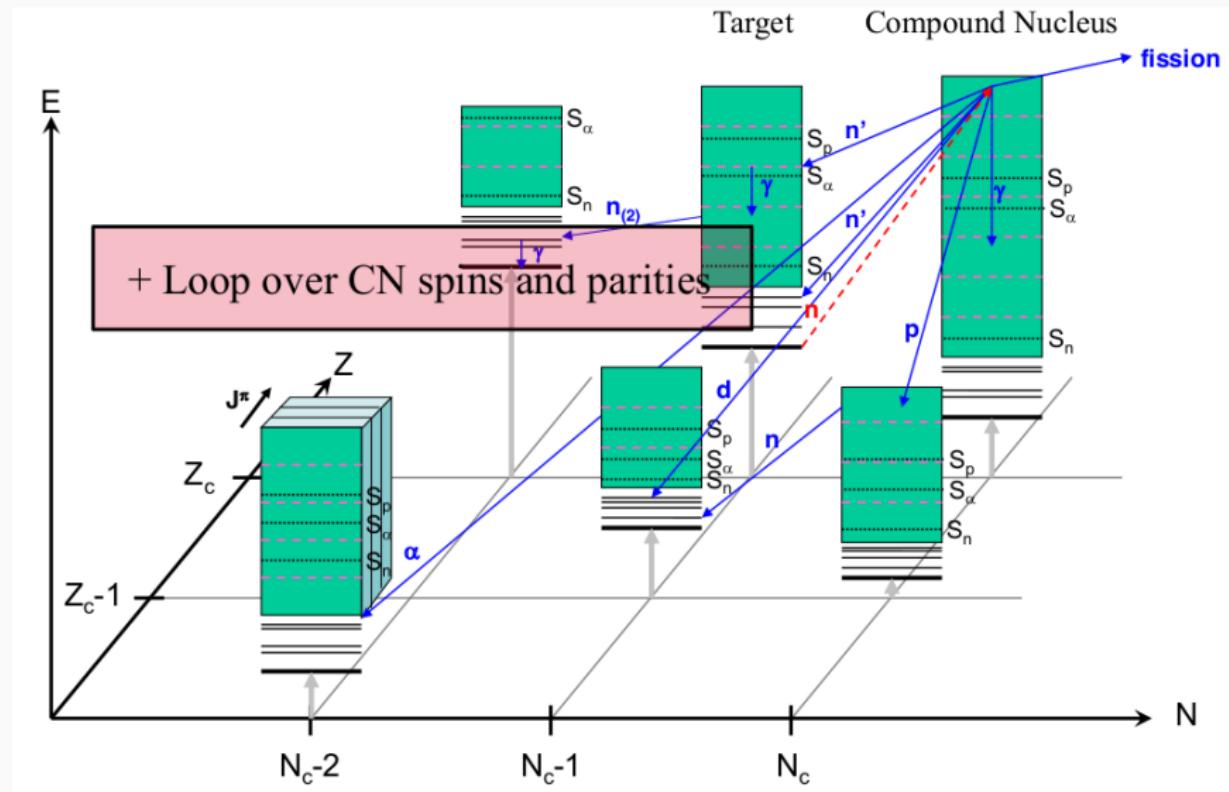
$$\langle T_b(\beta) \rangle = T_b^\pi(\beta) \text{ (from O.M.Potential)}$$

Emission in the continuum level

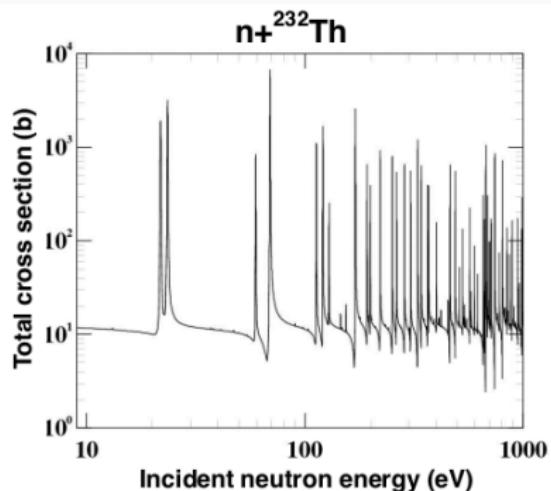
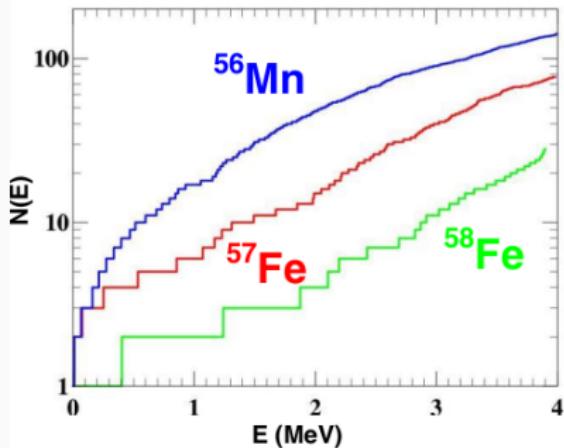
$$\langle T_b(\beta) \rangle = \int_E^{E+\Delta E} T_b^\pi(\beta) \rho(E, J, \pi) dE$$

$\rho(E, J, \pi)$ DENSITY of residual nuclear levels (J, π) with excitation energy E

CN multiple emissions



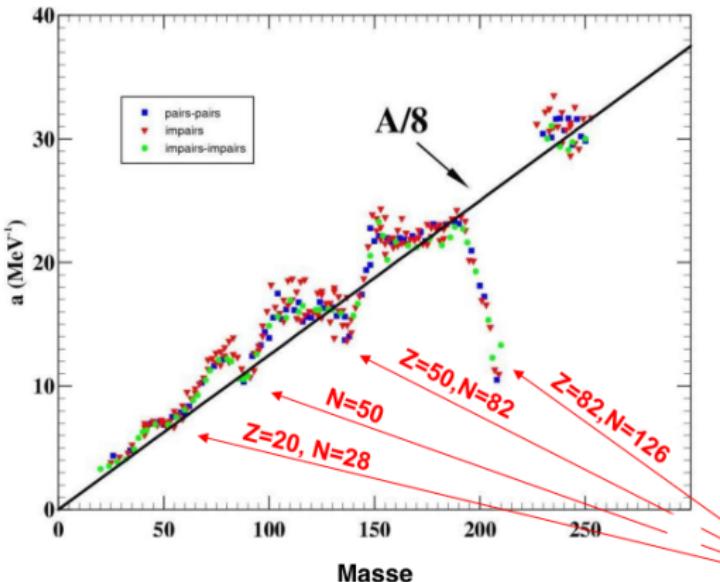
Level Density : overview



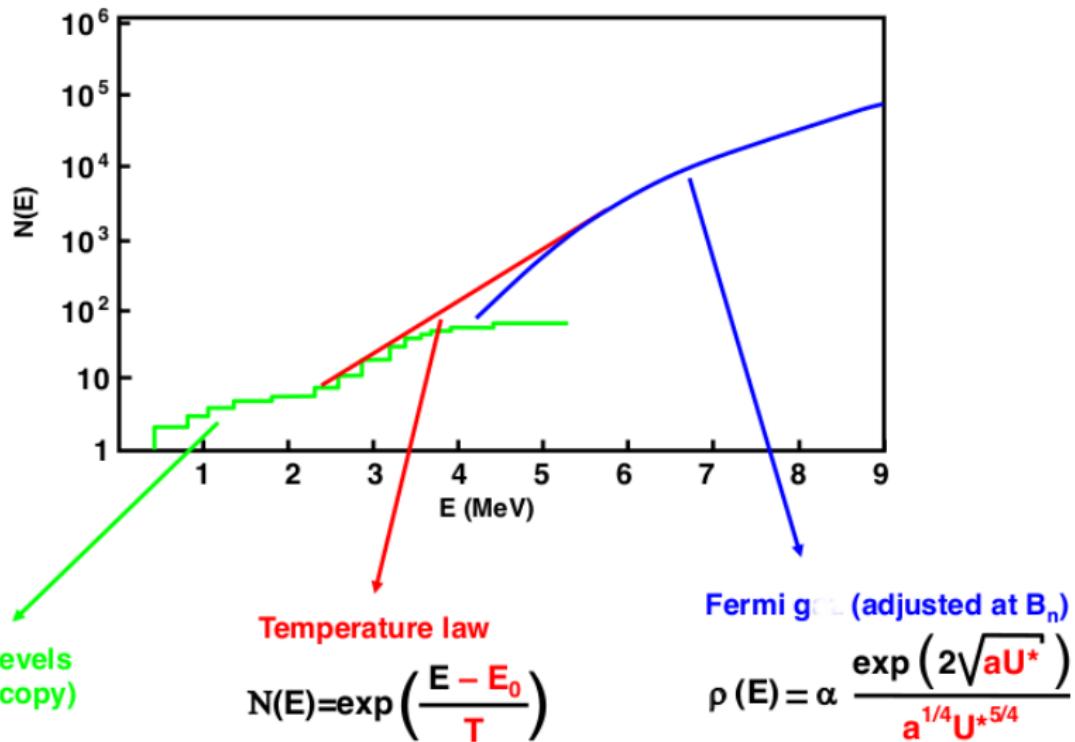
- Exponential increase of the cumulated number of discrete levels $N(E)$ with energy
- $\Rightarrow \rho(E) = \frac{dN(E)}{dE}$ increases exponentially
- \Rightarrow odd-even effects

Level Density: quantitative aspects

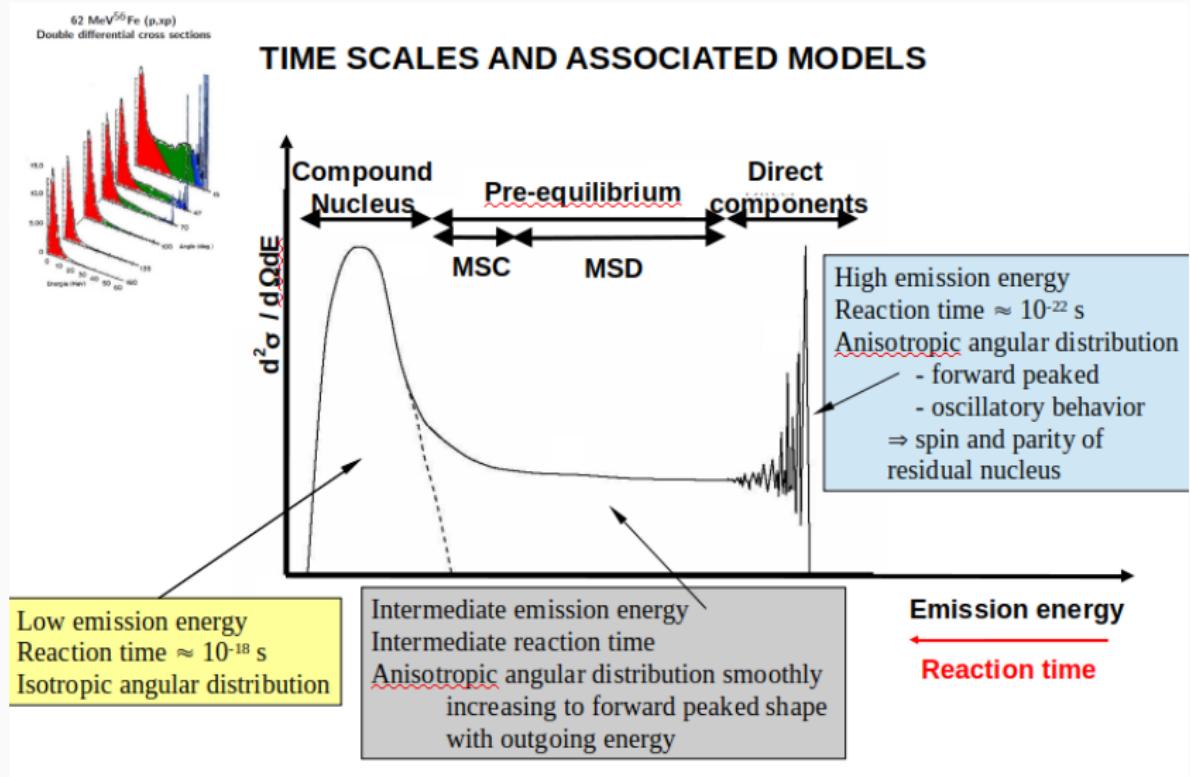
$$\rho(U, J, \pi) = \frac{1}{2} \frac{\sqrt{\pi}}{12} \frac{\exp(2\sqrt{aU})}{a^{1/4} U^{5/4}} \frac{2J+1}{2\sqrt{2\pi}\sigma^3} \exp - \left[\frac{(J+\frac{1}{2})^2}{2\sigma^2} \right]$$



Level Density : general description

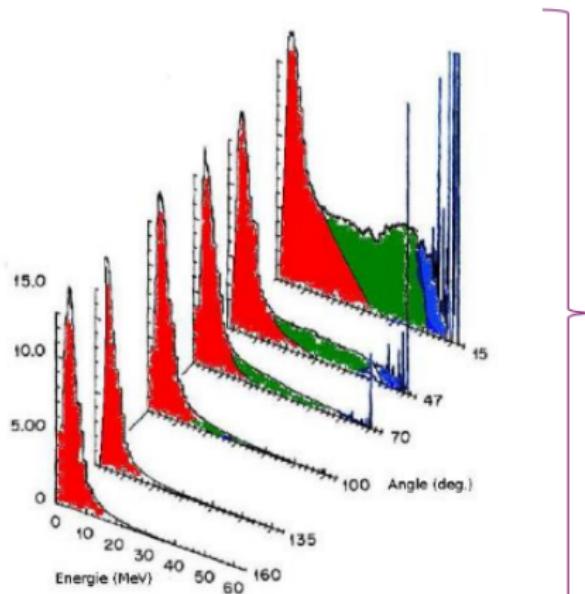


Phenomenology of nuclear reactions



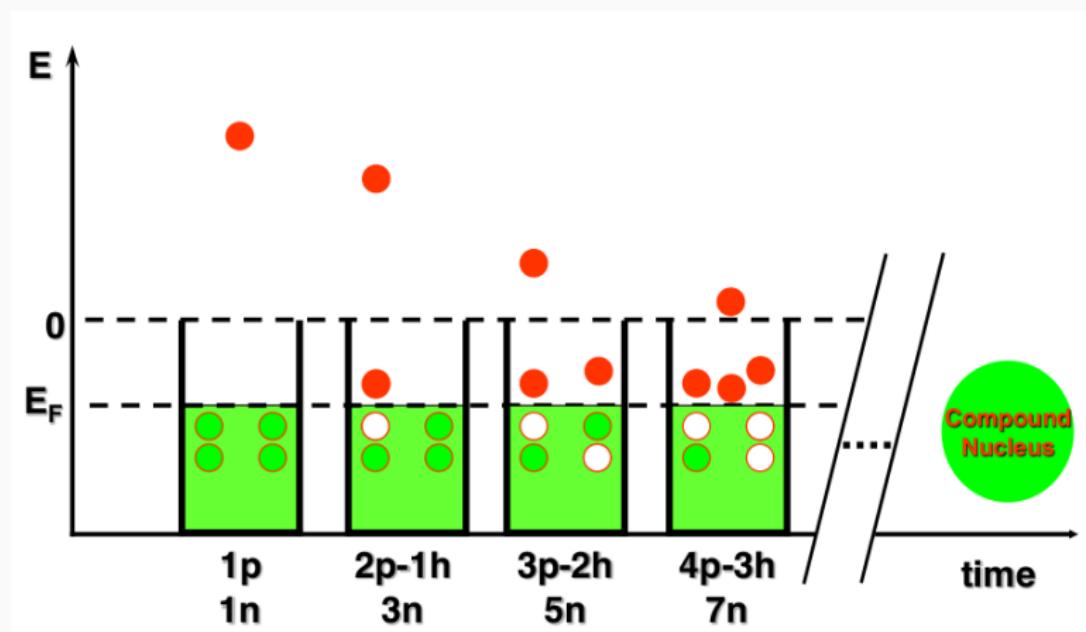
Angular/energy distributions

62 MeV ^{56}Fe (p,xp)
Double differential cross sections



- Always evaporation peak
- Discrete peaks at forward angles
- **Flat intermediate region**

Sketch of the excitation model



Sketch of the master equation

- $q(n, E, t)$ Probability of finding the composite system in exciton n and energy E
- $\lambda^\pm(n, E)$ Transition rate $n \rightarrow n \pm 2$
- $w(n, E)$ Total emission rate from n excitons

$$\begin{aligned}\dot{q}(n, t) = & \lambda^+(n-2)q(n-2, t) + \lambda^-(n+2)q(n+2, t) \\ & - (\lambda^+(n) + \lambda^-(n) + w(n))q(n, t)\end{aligned}$$

$$\frac{d\sigma^{P.E.}}{dE}(a, b) = \sigma_a \sum_{n, \Delta n=2} w_b(n, E) \int_0^\infty q(n, E, t) dt$$

Details of the master equation

Transition rates

$$\lambda(n \rightarrow n', E) = \frac{2\pi}{\hbar} \langle M^2 \rangle \omega(p', h', E) \quad \text{with } p' + h' = n'$$

$$w_b(n, E) = \frac{2s_b + 1}{2\pi^2 \hbar^3} \mu_b \epsilon_b \sigma_{inv,b}(\epsilon_b) \frac{\omega(p - p_b, h, E - \epsilon_b - B_b)}{\omega(p, h, E)}$$

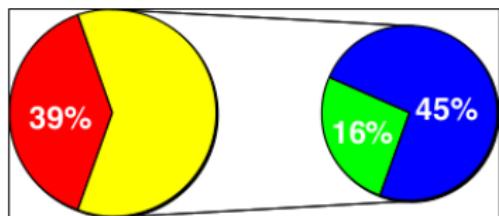
where the particle-hole state density is

$$\omega(p, h, E) = \frac{g^n}{p! h! (n-1)!} (E - B(p, h, g))^{n-1}$$

(plus many complications and improvements...)

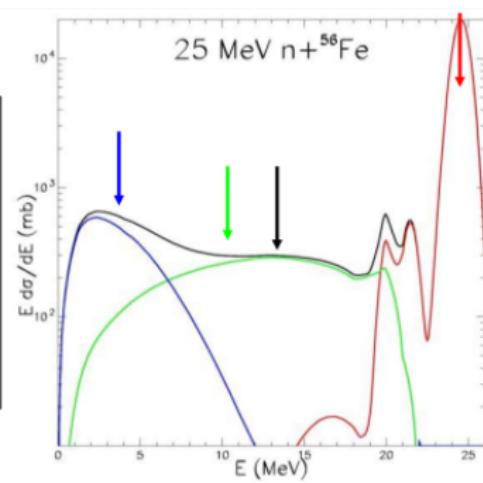
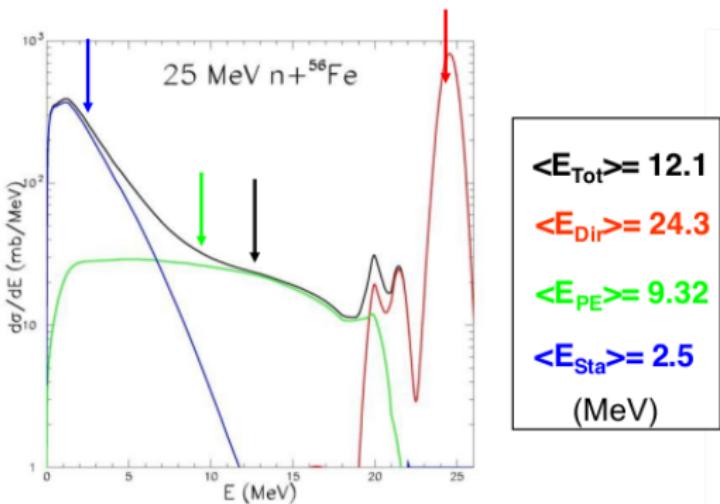
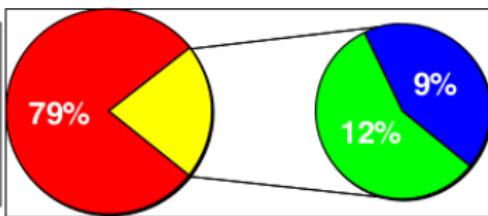
Overview of reaction components

Cross section



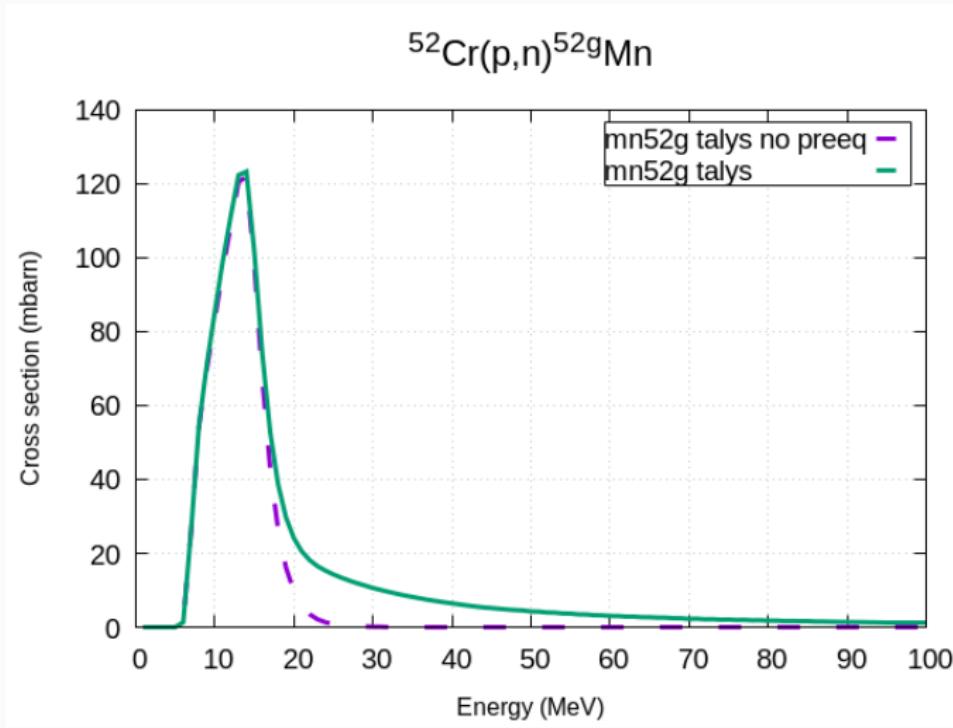
Total
Direct
Pre-equilibrium
Statistical

Outgoing energy



Production of radioactive ^{52}Mn

→ multimodal imaging (PET/NMR)



References

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