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Resonance self-shielding in the epi-thermal region

Workshop on Nuclear Data for Activation Analysis

Trieste, Italy, 7 – 18 March 2005

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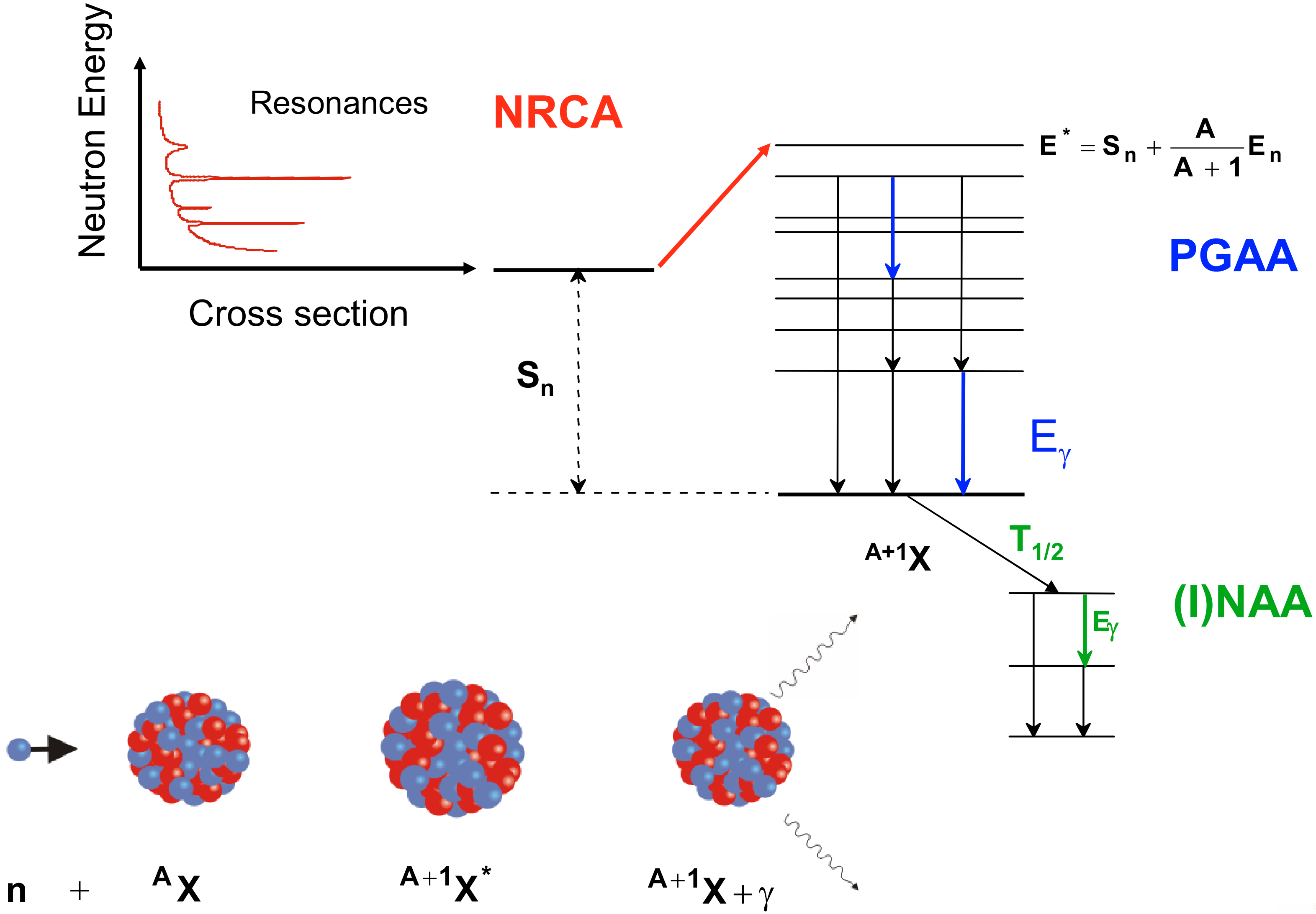
Neutron resonances and NAA

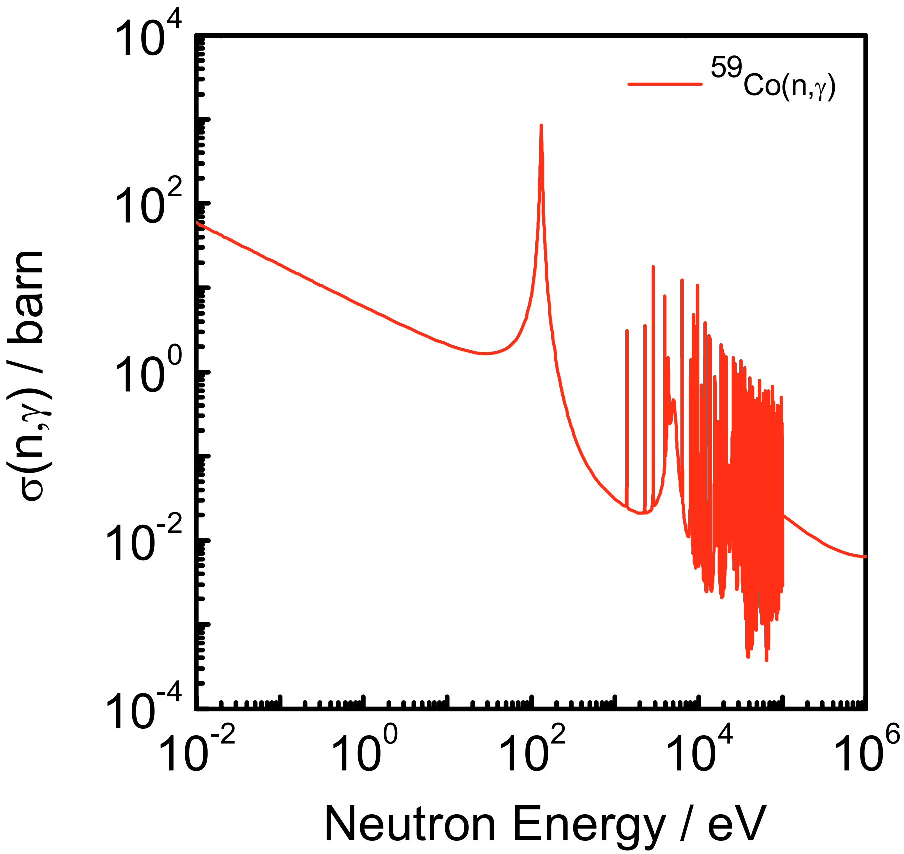
- Basic principles of NAA
- Nuclear reaction theory
- Neutron resonances and NAA
- Resonance self-shielding for a parallel beam
- Resonance self-shielding for an isotropic beam

1. Basic principles of NAA

Neutron capture process



Capture cross section



- **The probability that a neutron interacts with a nucleus is expressed as a cross section σ, which has the dimension of an area**
- **The unit of a cross section is taken as : 1 barn, 1 b = 10^{-24} cm^2**
- **To calculate reaction probabilities we express the target thickness in atoms per barn :**

$$n = \frac{0.6022}{m_A} \rho\, t$$

m_A : atomic mass
ρ : density in g/cm^3
t : thickness in cm
n : target thickness in at/b

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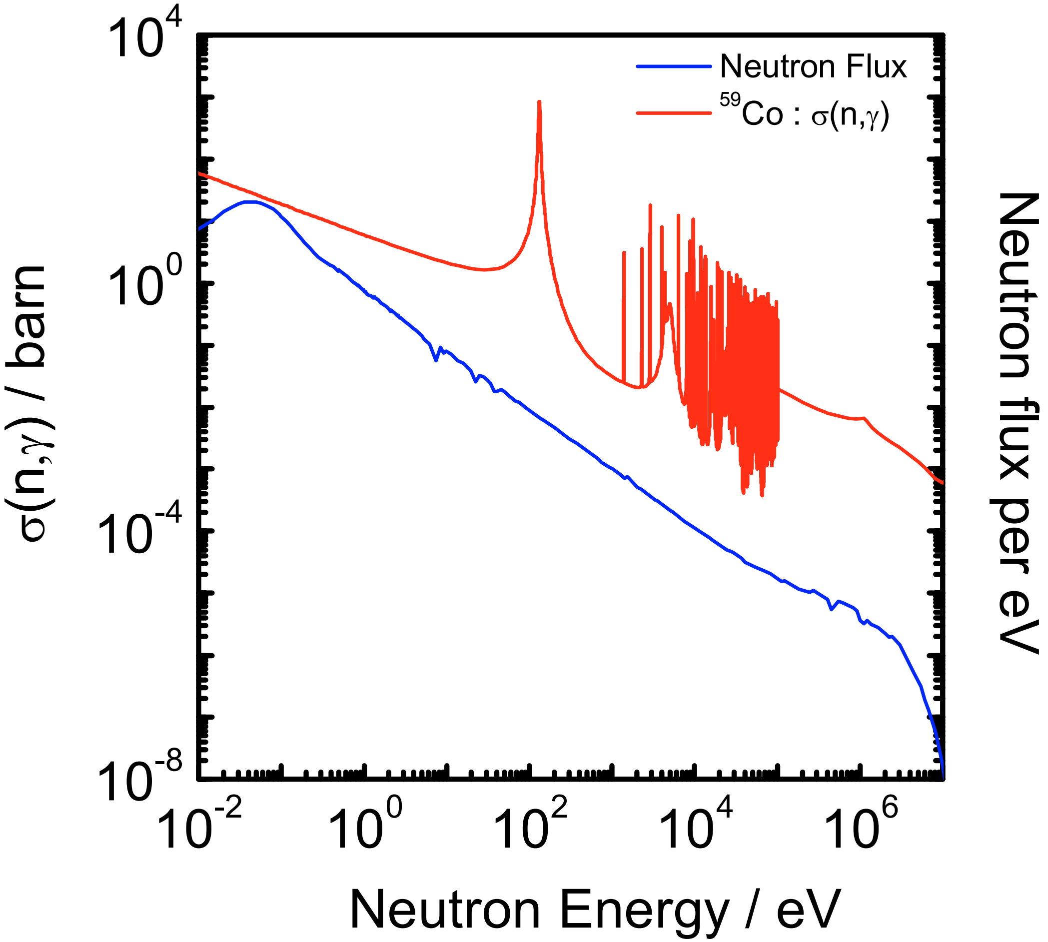
Total reaction rate



The total reaction rate per atom:

$$R = \int_0^\infty \varphi(E_n)\,\sigma_\gamma(E_n)\,dE_n$$

depends on:

$\varphi(E_n)$ the neutron flux and

$\sigma_\gamma(E_n)$ the capture cross section

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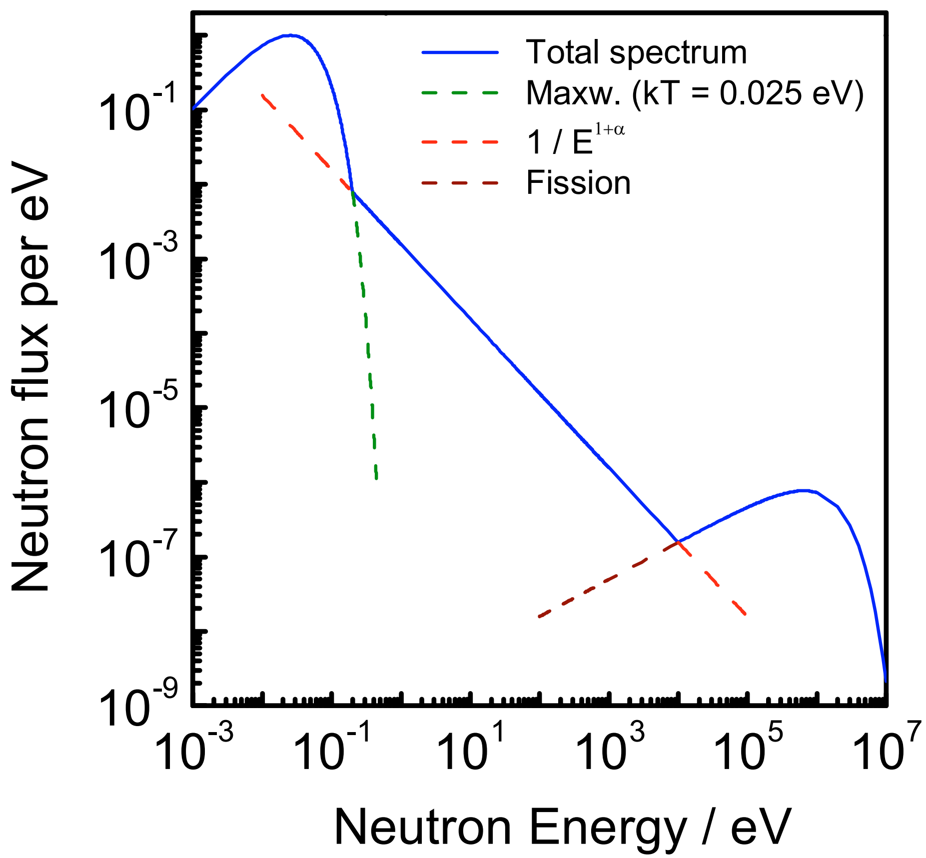
Neutron flux in a thermal reactor



The neutron flux in <u>a thermal reactor</u> is a sum of three components

- **Maxwellian distribution with maximum at $E_n = kT$**
- **$1/E^{1+\alpha}$ distribution due to moderation process of the fast neutrons (epi-thermal spectrum)**
- **"Watt spectrum" of fission neutrons**

At <u>a neutron guide</u>, the neutron flux can be described by a Maxwellian distribution

(cfr. PGAA at Budapest, i.e no resonance shielding !)

Reaction rate in a thermal reactor

- **The total reaction rate per atom is:**

$$R = \int_0^{\infty} \varphi(E_n)\,\sigma(E_n)\,dE_n$$

- **To solve the integral one separates between the thermal and the epi-thermal region:**

$$R = \int_0^{E_{Cd}} \varphi(E_n)\,\sigma(E_n)\,dE_n + \int_{E_{Cd}}^{E_3} \varphi(E_n)\,\sigma(E_n)\,dE_n + \text{fast}$$

with E_{Cd} = 0.55 eV

- **Conventions : Høgdahl & Westcott convention**

Importance of resonance region

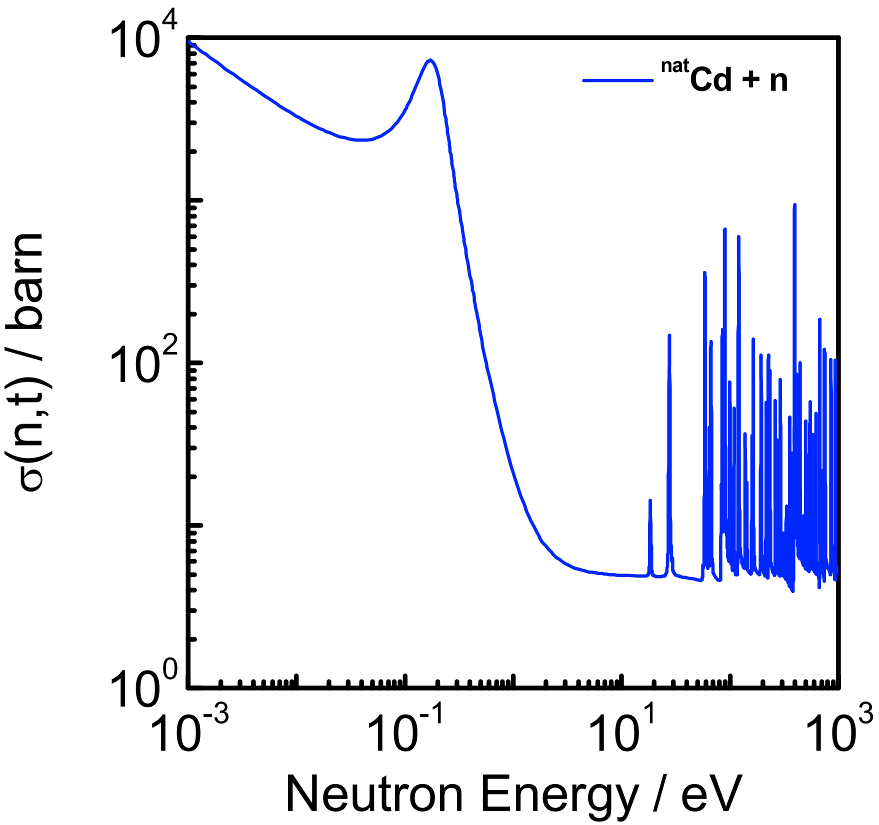
- **Reaction rate** $R \cong G_t \varphi_t \sigma_o g_w + G_R \varphi_e I_R$
  - σ_o : capture cross section at thermal
  - g_w : Westcott g-factor (expresses the deviation of σ_γ from 1/v)
  - φ_t : thermal neutron flux
  - G_t : thermal self-shielding

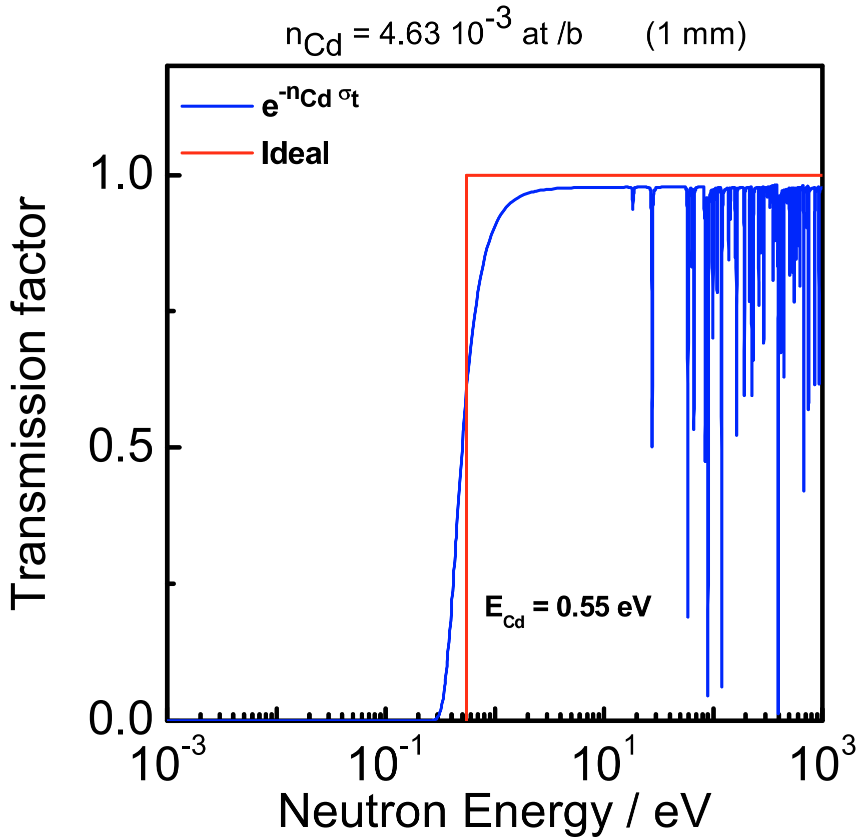
  - I_R : resonance integral
  - φ_e : epi-thermal neutron flux
  - G_R : resonance self-shielding

- **Cd-ratio measurements** $F_{Cd} = \dfrac{R_{Cd}}{G_R \varphi_e I_R}$

$\Rightarrow$ σ_o, g_w, I_R, G_R, F_{Cd} are influenced by the resonance structure

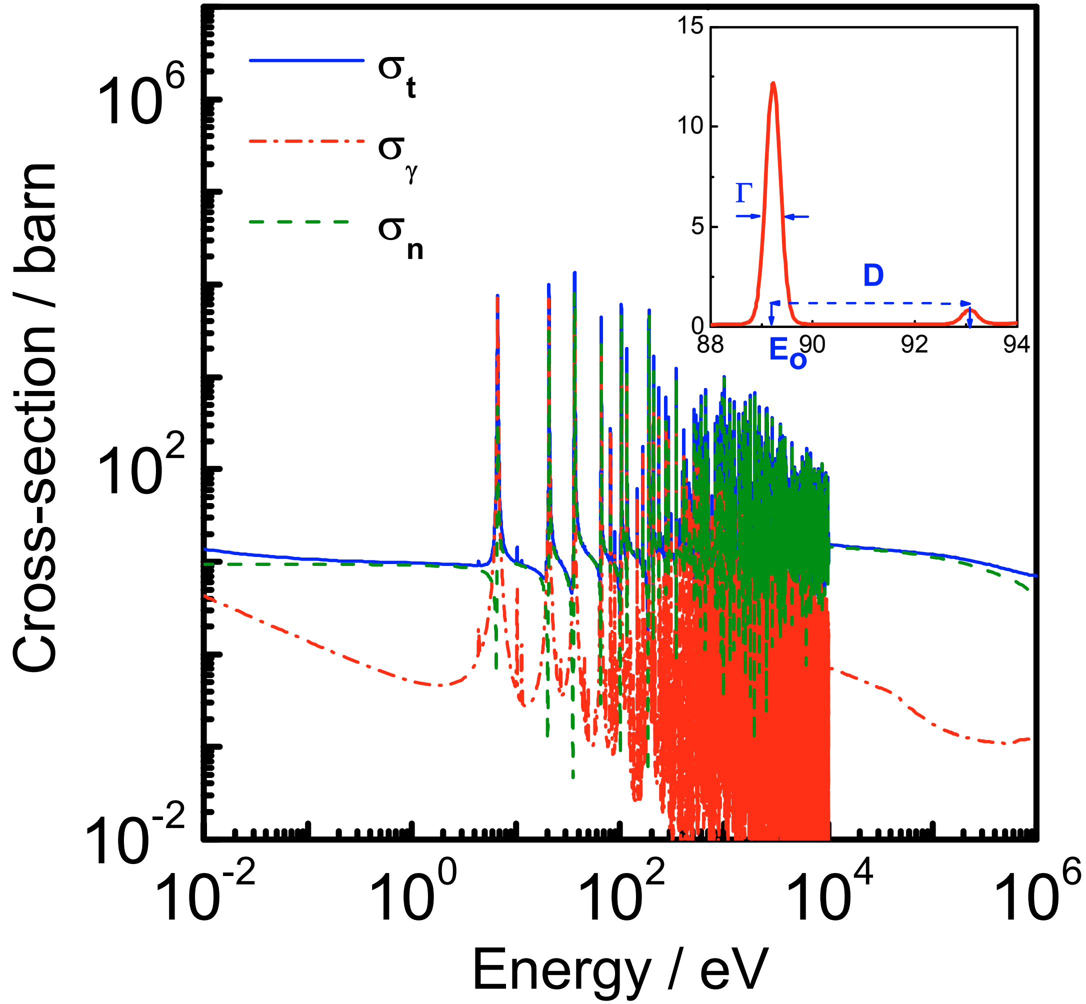
Transmission through cadmium





2. Nuclear reaction theory

Energy differential cross sections



^{238}U(n,tot) = ^{238}U(n,n) + ^{238}U(n,γ)

$\sigma_{tot} = \sigma_n + \sigma_\gamma$

- **Thermal**
- **Resonance Region : D > Γ**

  Resolved Resonance Region : Δ_R < D
  Unresolved Resonance Region : Δ_R > D

- **High Energy Region : D < Γ**

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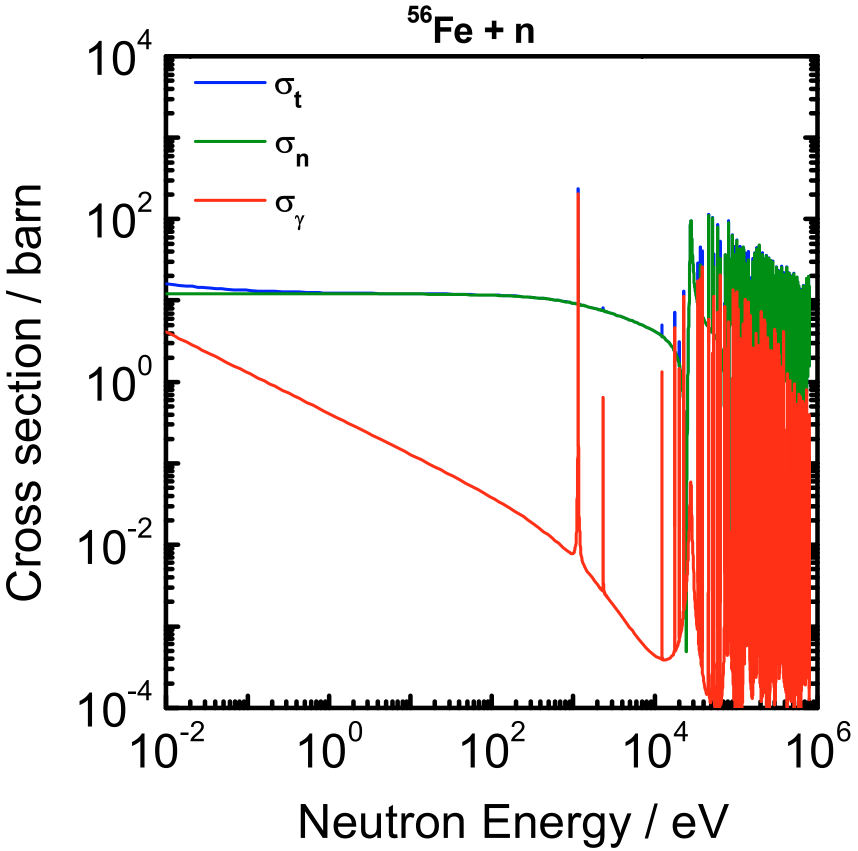
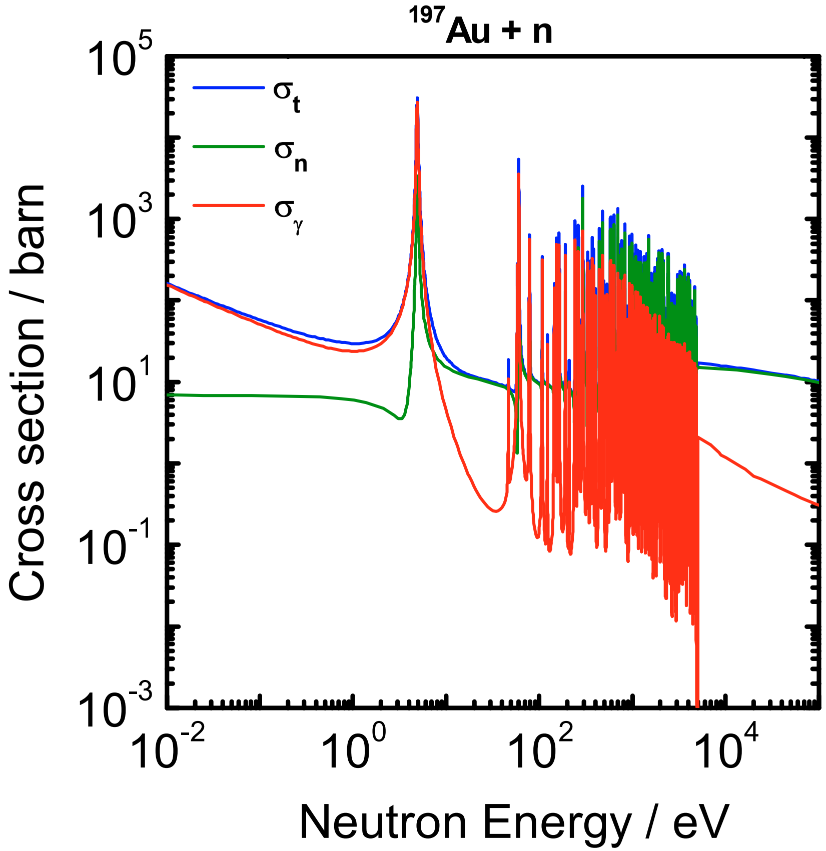
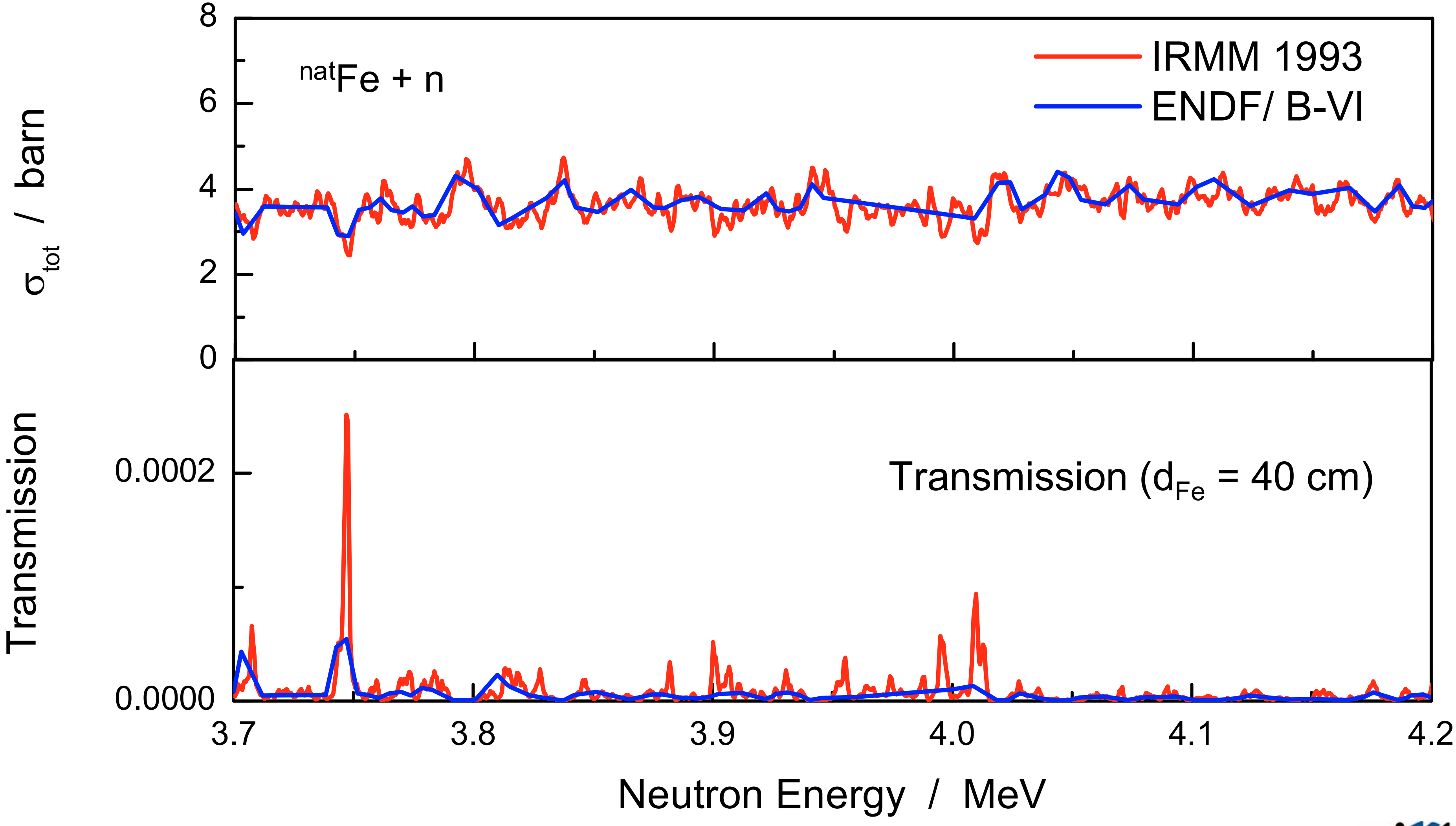
Energy Differential Cross Sections



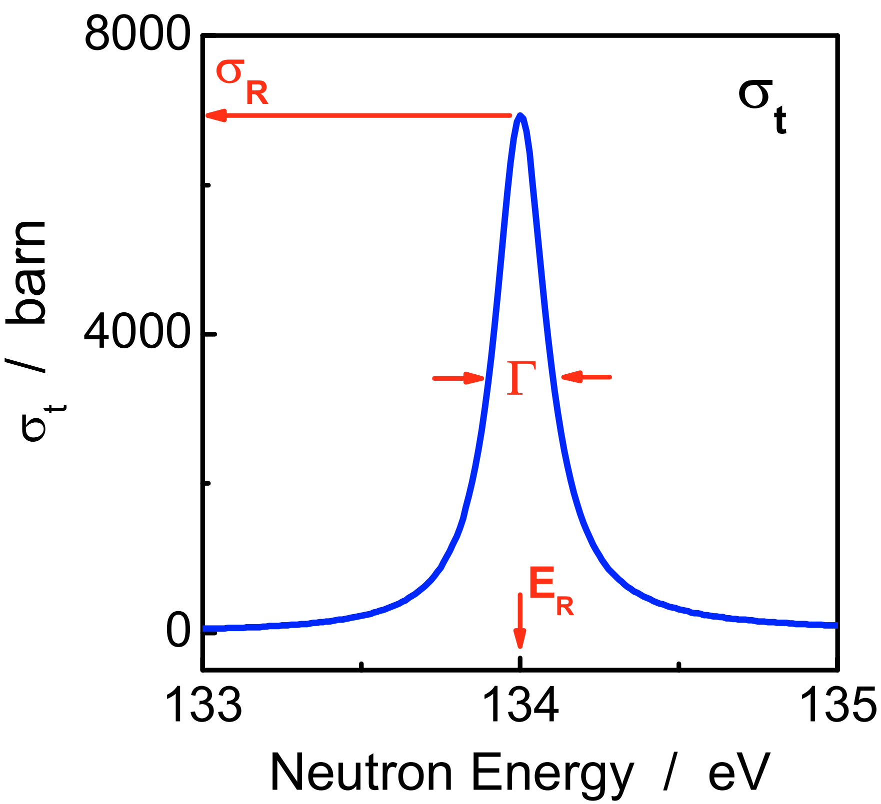


- No capture without scattering
- Relative contribution of σ_n and σ_γ to σ_t may be different
- Boundaries of the RRR differ

Importance of resonance structure



Resonance structure



A cross section as a function of E_n shows a resonant structure, which can be described by a Breit-Wigner shape :

$$\sigma_t \sim \frac{1}{\left(E_n - E_R\right)^2 + (\Gamma/2)^2}$$

with

Γ **natural line width (FWHM)**

E_R **resonance energy**

Bohr's hypothesis : Compound nucleus reaction

- **Two step process**

(1) Formation of compound nucleus σ_{C^*}

$$\sigma_{C^*}(E_n) = g\frac{\pi}{k_n^2}\frac{\Gamma_n\Gamma}{(E_n - E_R)^2 + (\Gamma/2)^2}$$

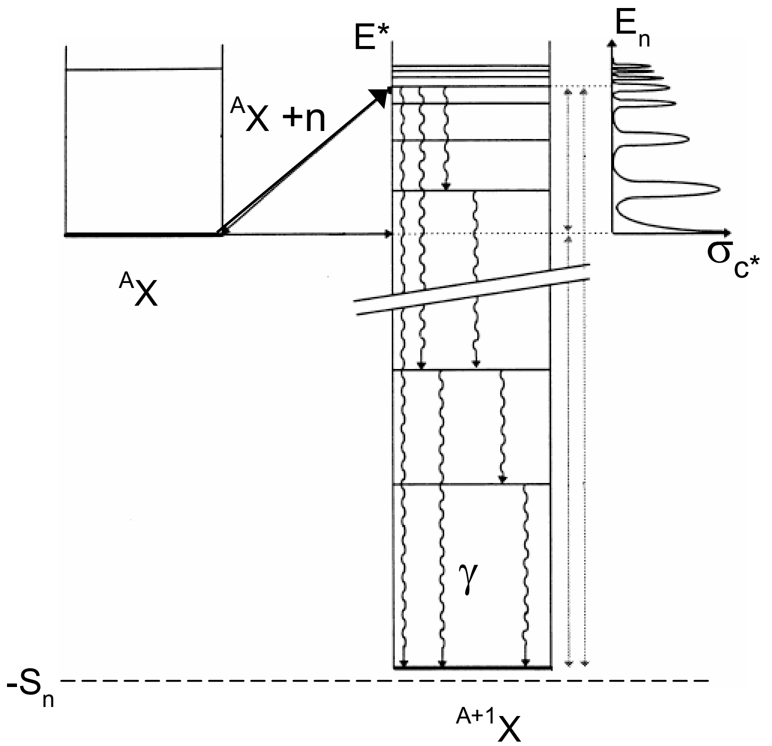
$$\Gamma = \sum_r \Gamma_r \quad (r = n, \gamma, f, ...)$$

(2) Decay of compound nucleus P_r

$$P_r = \frac{\Gamma_r}{\Gamma} \quad (r = n, \gamma, f, ...)$$

- **Partial cross section**

$$\sigma_r = \sigma_{C^*} P_r$$



$$E^* = S_n + \frac{A}{A+1}E_n$$

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Breit – Wigner formula
Resonance part of the cross section

- **Total Cross Section** **(n,tot)**

$$\sigma_t(E_n) = g\frac{\pi}{k_n^2}\frac{\Gamma_n\Gamma}{(E_n - E_R)^2 + (\Gamma/2)^2}$$

$$g = \frac{2J+1}{2(2I+1)};\ k_n = \text{wavenumber}$$

- **Elastic Cross Section** **(n,n)**

$$\sigma_n(E_n) = \sigma_t(E_n)\frac{\Gamma_n}{\Gamma}$$

- **Capture Cross Section** **(n,γ)**

$$\sigma_\gamma(E_n) = \sigma_t(E_n)\frac{\Gamma_\gamma}{\Gamma}$$

e.g. ^{109}Ag s-wave at E_o = 134 eV
(E_o, Γ_n, Γ_γ , J^π)

$$\sigma_R(\text{barn}) = \frac{2.608 \times 10^6}{E_R(\text{eV})}\left(\frac{m_A + 1}{m_A}\right)^2 \frac{g\Gamma_n}{\Gamma}$$

$$\Gamma = \Gamma_n + \Gamma_\gamma$$

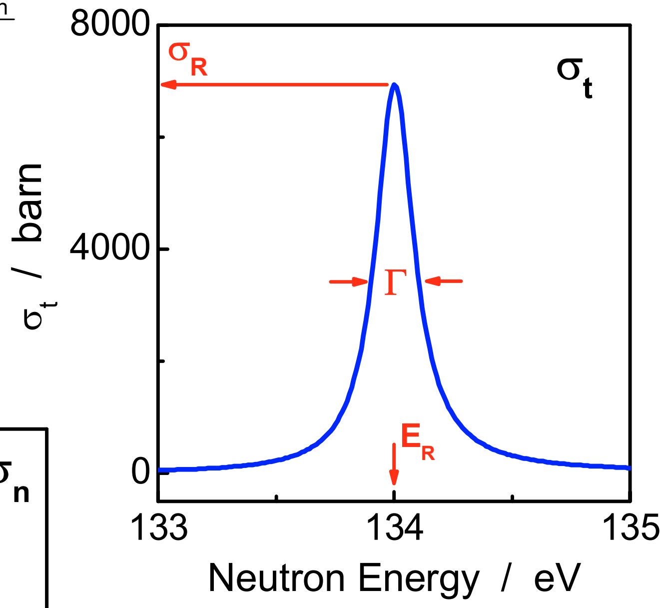
E_o = 134 eV

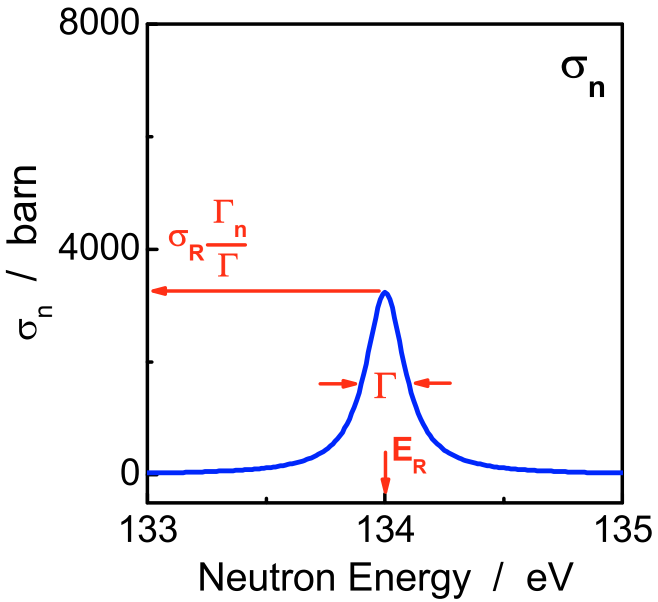
Γ_n = 0.093 eV

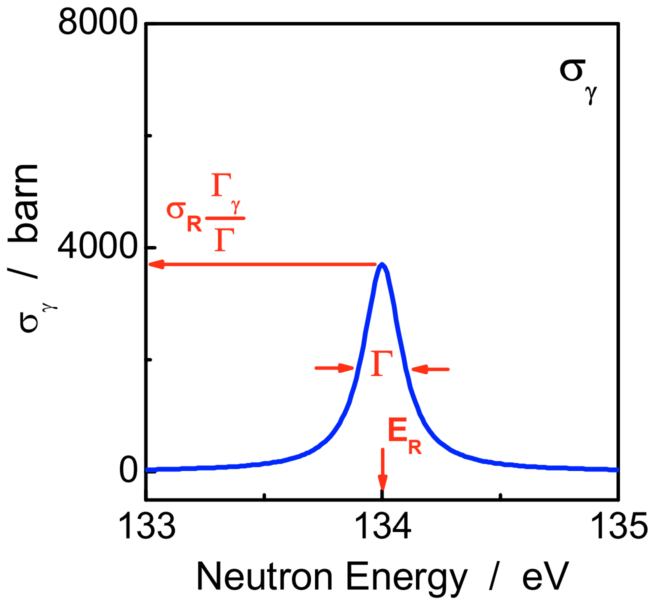
Γ_γ = 0.106 eV

J^π = 1^-

g = 3/4







Energy dependence of neutron width

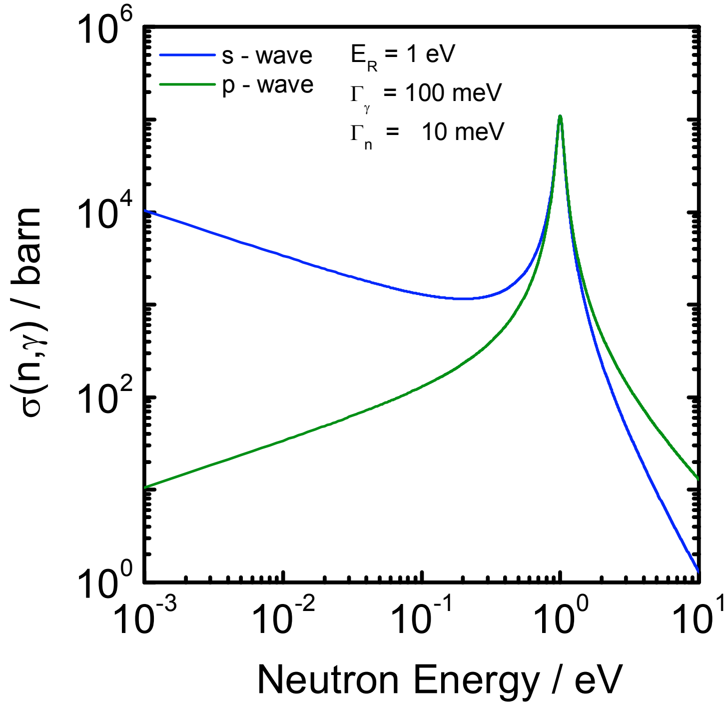
- The energy dependence of the neutron width is due to the centrifugal-barrier penetrability, which depends on the angular momentum of the incoming neutron ℓ and E_n

- The neutron width Γ_n depends on E_n

s-wave (ℓ = 0) $\Gamma_n(E_n) = \Gamma_n^0 \sqrt{\frac{E_n}{1\text{eV}}}$

p-wave (ℓ = 1) $\Gamma_n(E_n) = \Gamma_n^1 \sqrt{\frac{E_n}{1\text{eV}}} \frac{k_n^2 a^2}{1 + k_n^2 a^2}$

$$\sigma_\gamma(E_n) = g \frac{\pi}{k_n^2} \frac{\Gamma_n \Gamma_\gamma}{(E_n - E_R)^2 + (\Gamma/2)^2}$$

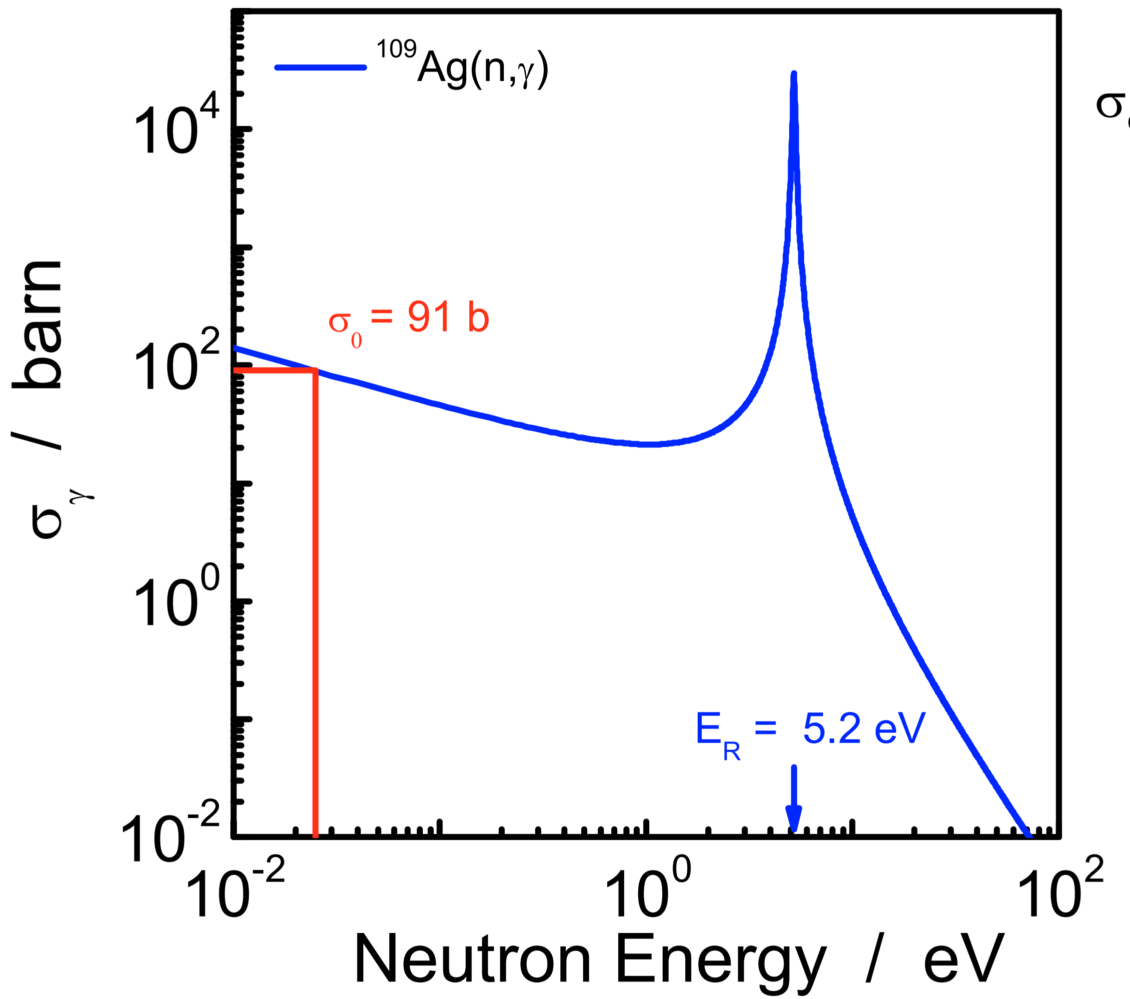


3. Neutron Resonances and NAA

Influence of resonance structure on:

- **Thermal capture cross section**
- **1/v behaviour of the capture cross section**
- **Westcott g_w - factor**

σ_o and contribution of s-wave resonances #1



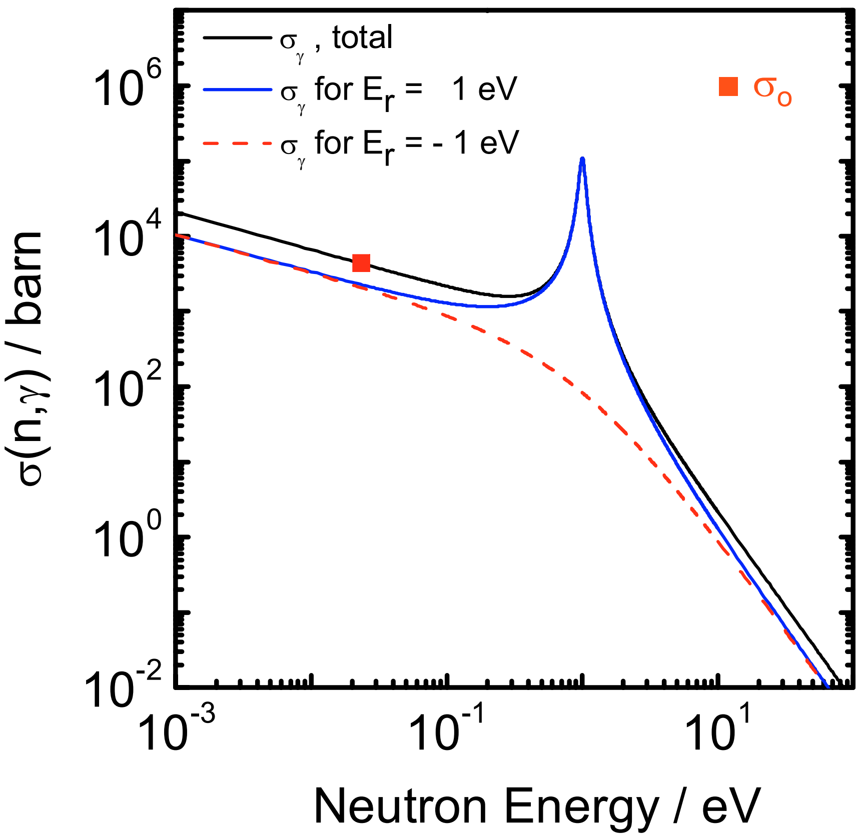
$$\sigma_{o,\ell=0} \cong 4.099\mathrm{x}10^6 \left(\frac{m_A + 1}{m_A} \right)^2 \sum_{j=1}^{N} \frac{g\Gamma_{nj}^o \Gamma_{\gamma j}}{E_{Rj}^2}$$

with

σ_o in barn

E_R, Γ_n^o, Γ_γ in eV

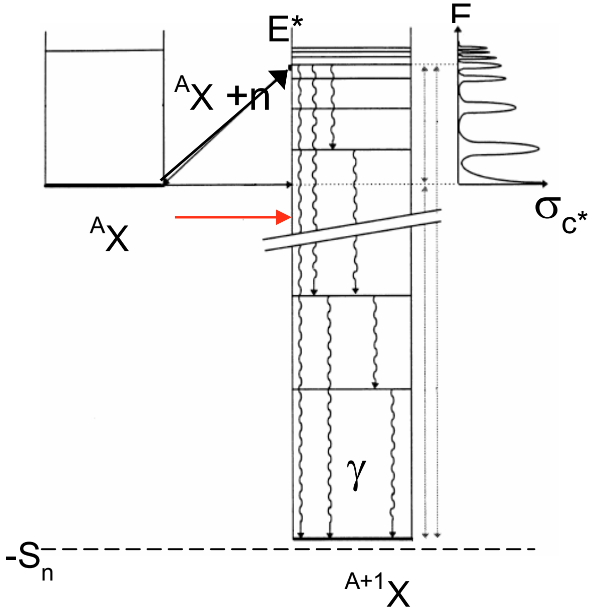
σ_o and contribution of s-wave resonances #2



- **If**

$$\sigma_o > \sigma_{o,\ell=0} \cong 4.099.10^6 \left(\frac{m_A + 1}{m_A}\right)^2 \sum_{j=1}^{N} \frac{g\Gamma^o_{nj}\Gamma_{\gamma j}}{E_{Rj}^{\ 2}}$$

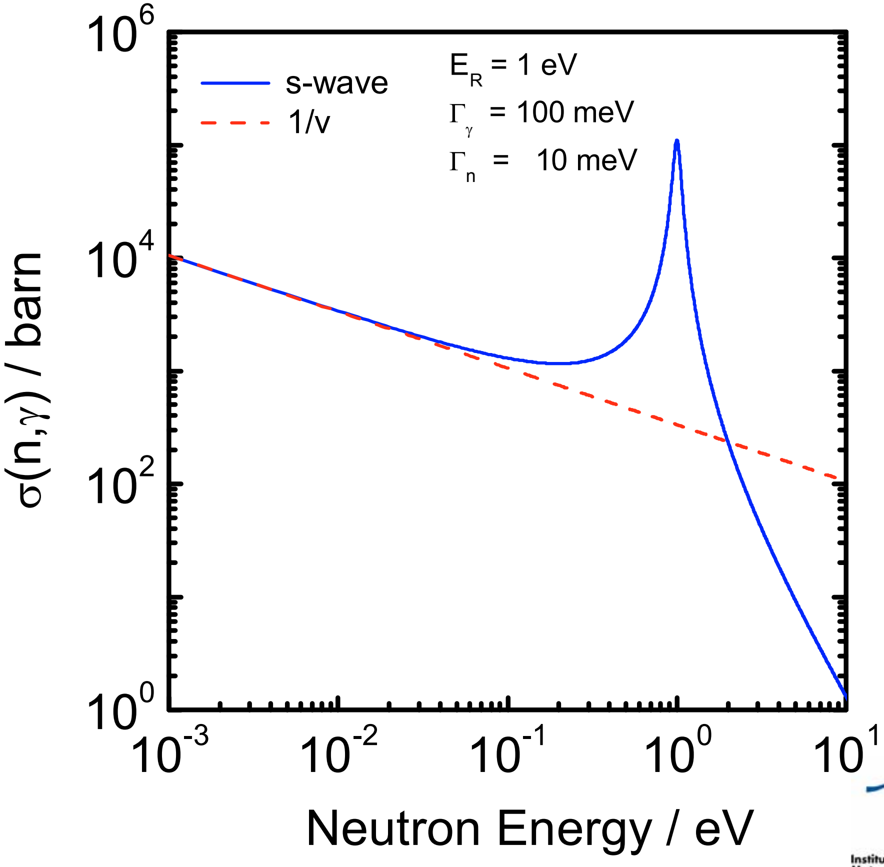
- **Additional contribution from bound states (negative resonances)**



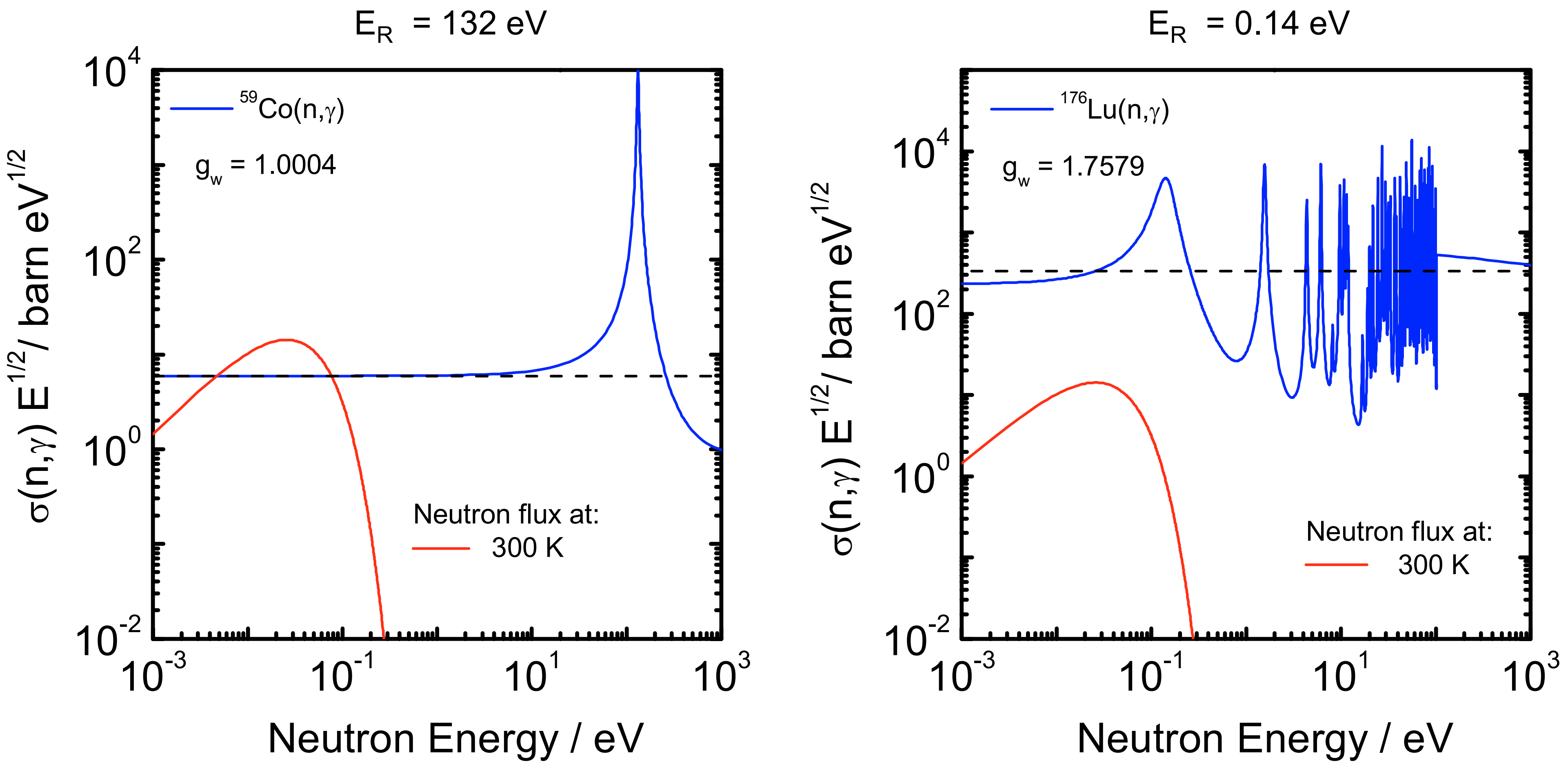
1/v behaviour of capture cross section

- **Capture cross section** $\sigma_\gamma(E_n) = g\frac{\pi}{k_n^2}\frac{\Gamma_n\Gamma_\gamma}{(E_n - E_R)^2 + (\Gamma/2)^2}$
- **Neutron width for a s-wave neutron** $\Gamma_n(E_n) = \Gamma_n^0\sqrt{\frac{E_n}{1\,\text{eV}}}$
- **For $E_n << E_R$ witk $k_n^2 \propto E_n$**

$$\sigma_\gamma(E_n) \propto \frac{1}{\sqrt{E_n}} = \frac{1}{v_n}$$



Westcott g_w - factor: deviation of 1/v behaviour #1



Resonance self-shielding in the epithermal region , P. Schillebeeckx 15_03_05

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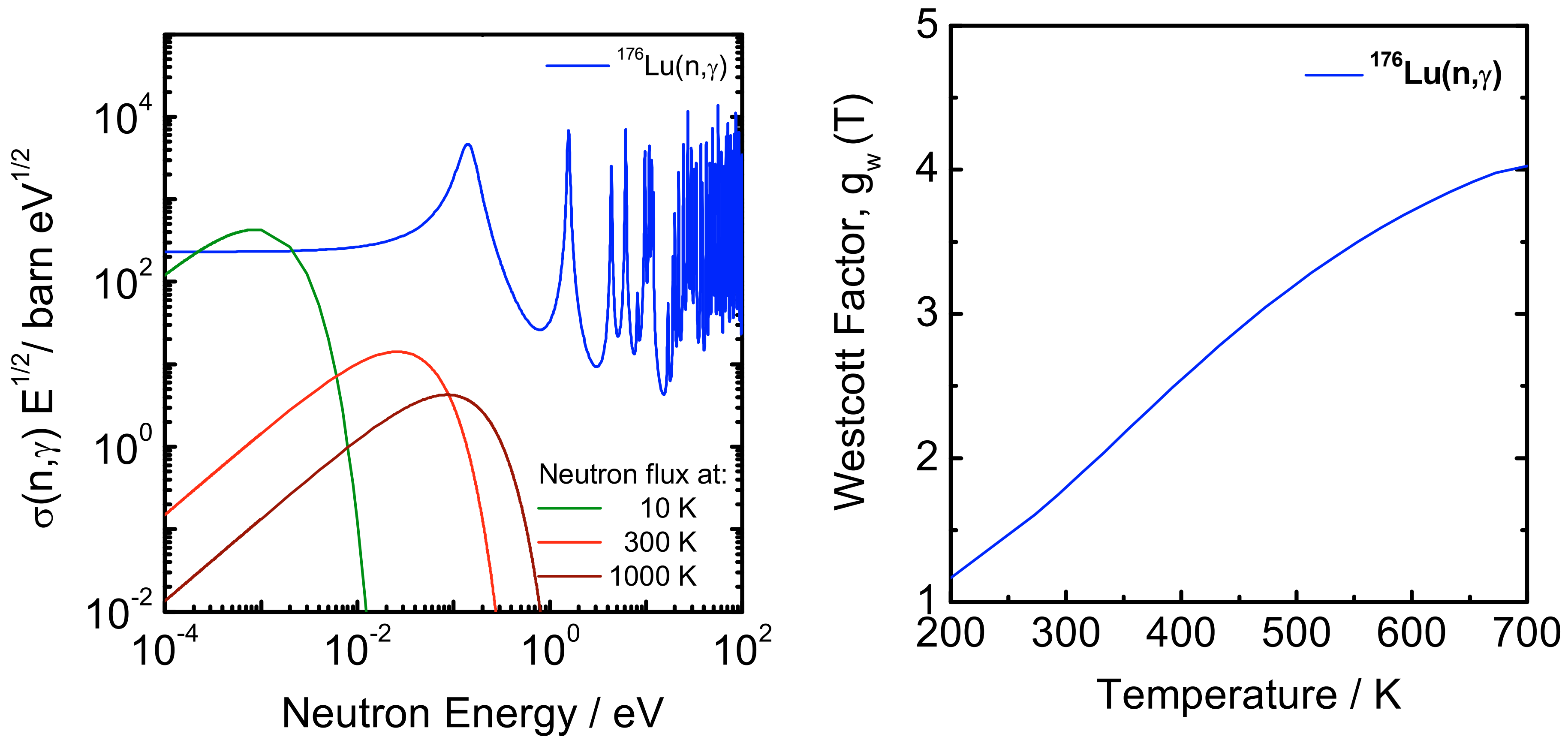
Westcott g_w - factor: deviation of 1/v behaviour #2

The Westcott g_w – factor is temperature dependent



⇒ ^{176}Lu(n,γ) temperature monitor

Data base of resonance parameters

- **Evaluated data libraries (see IAEA INDC website)**
  - JEF
  - ENDF-B
  - JENDL
  - CENDL
- **Compilation by S.F.Mughabghab**

“Neutron Resonance Parameters and Thermal Cross Sections”

Part A & B

NNDC, BNL, 1984

Data base of σ_o, g_w-factor and I_R

Compilation by S.F.Mughabghab, BNL, USA

"Thermal neutron capture cross sections, resonance integrals and g_w-factors"

INDC(NDS) – 440

February 2003

4. Resonance self-shielding for a parallel beam

Self-shielding and multiple scattering

Parallel neutron beam on a foil or a disc

- **Study the basic effects**

  (parallel beam is not directly applicable to NAA, but experimental verification of procedures is possible)

- **Influence of resonance structure on the self-shielding factor**

- **Doppler effect**

- **Total correction factors (due to self-shielding & scattering)**

- **Interference effects**

Parallel beam on thin sample

- **For a relatively thin sample, no scattering only self-shielding**

$$R(E_n) \propto \int_0^t \rho\, \sigma_\gamma(E_n)\, \varphi(x)\, dx$$

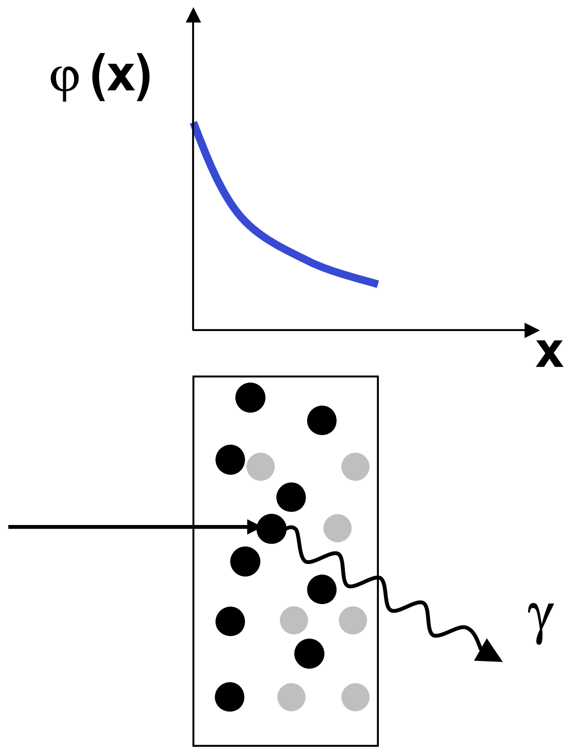
$$R(E_n) \propto \varphi_{x=0}\; n\sigma_\gamma(E_n) \boxed{\frac{\left(1-e^{-n\,\sigma_t(E_n)}\right)}{n\,\sigma_t(E_n)}}$$

self-shielding factor

- **Infinitely thin sample**

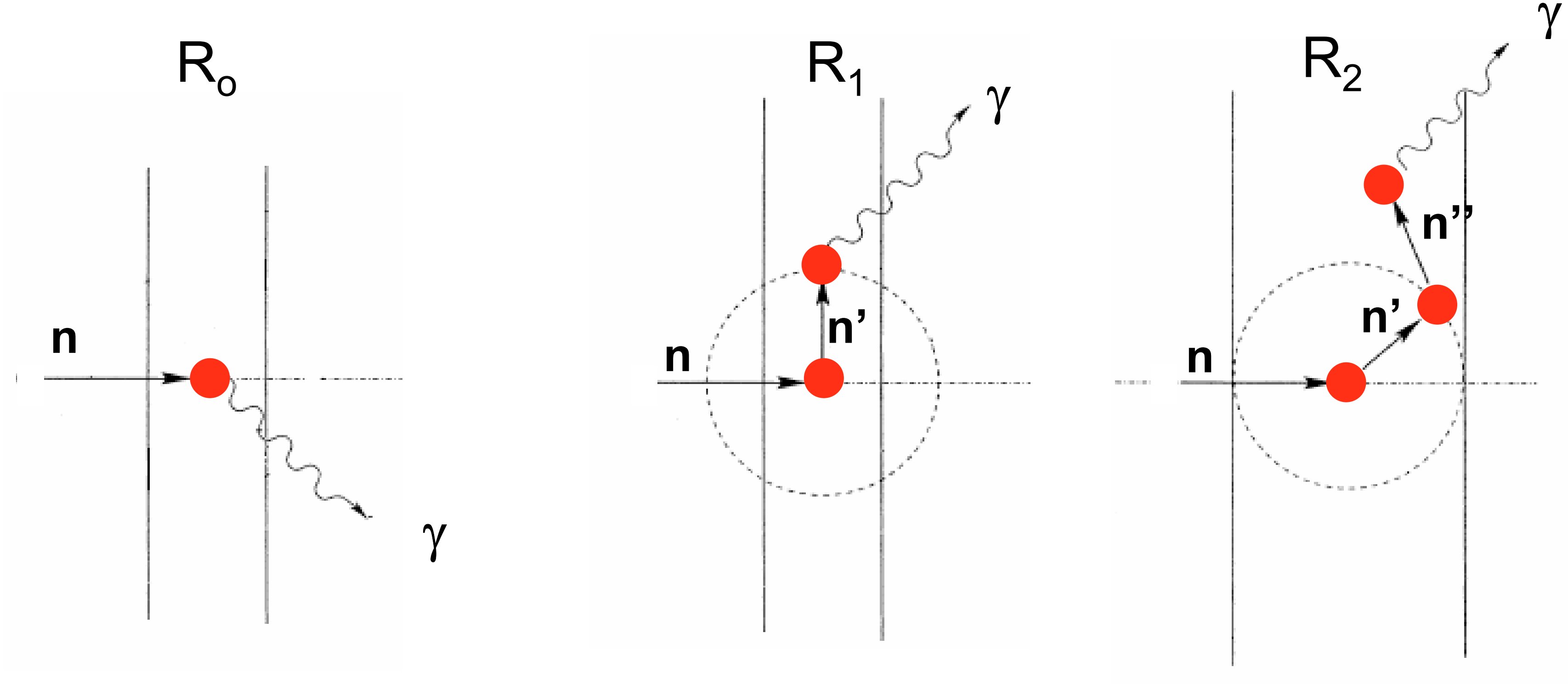
$$R_{thin} \propto n\sigma_\gamma \frac{\left(1-e^{-n_r\sigma_t}\right)}{n\,\sigma_t} = n\sigma_\gamma \quad \text{for } n\sigma_t << 1$$

$$\varphi(x)\, dx = \varphi_{x=0}\; e^{-\rho\sigma_t x}\; dx$$



Self-shielding and multiple scattering

$$R = \sum_j R_j$$



$$R \propto n\sigma_\gamma \frac{(1 - e^{-n\sigma_t})}{n\sigma_t}$$

$$E_n^{'} = E_n \left(\frac{m_n}{m_A + m_n}\right)\left(\cos\theta + \sqrt{\left(\frac{m_A}{m_n}\right)^2 - \sin^2\theta}\right)$$

Self-shielding and multiple scattering factor

- **Calculation**

  – Analytical expressions
  REFIT (parallel beam+ disc)
  SAMMY (parallel beam+ disc)

$$G_R = \frac{\int_{E_1}^{E_2} dE \int d\Omega \int_0^t dx\, \Sigma_\gamma(E)\, \Phi(x,\Omega,E)}{\int_{E_1}^{E_2} dE \int d\Omega \int_0^t dx\, \Sigma_\gamma(E)\, \Phi(x=0,\Omega,E)}$$

  – Monte Carlo simulations
  SAMSMC (parallel beam + disc)
  MCNP (no geometry limitations, use probability tables !!)

- **Definitions**

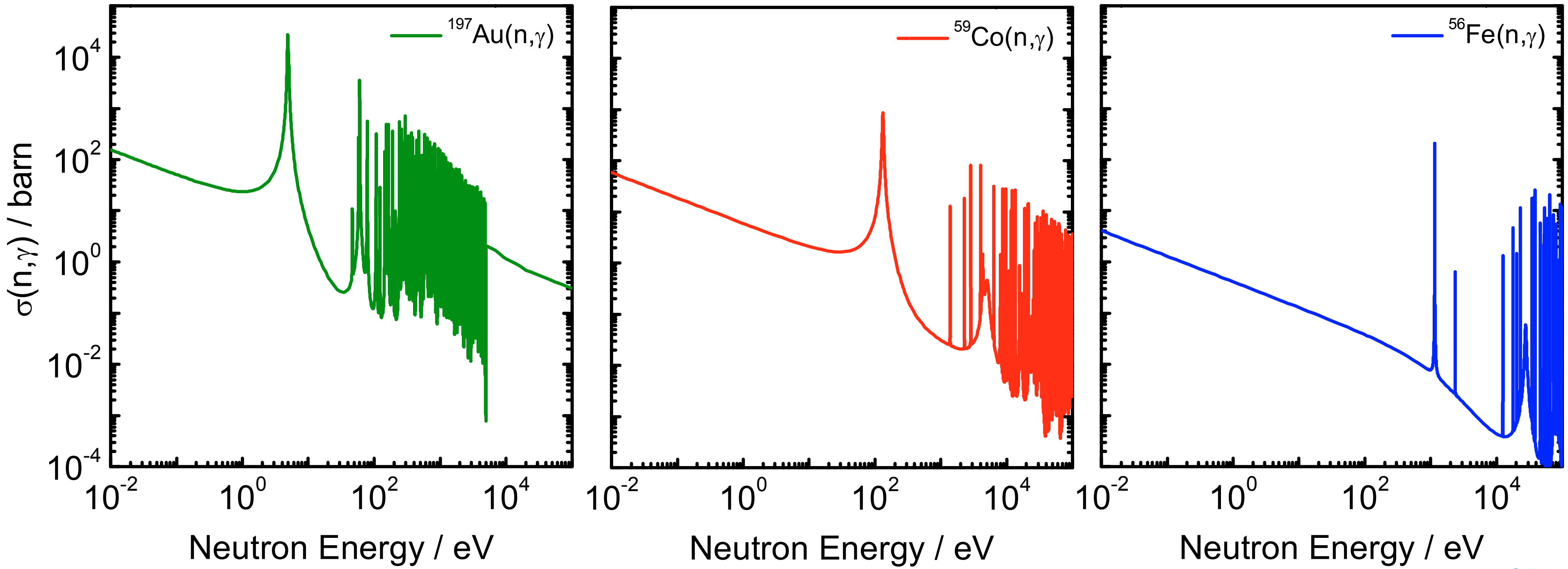
  – Self-shielding without scattering : $G_{R,0}$

  – Self-shielding + 1 scattering : $G_{R,1}$

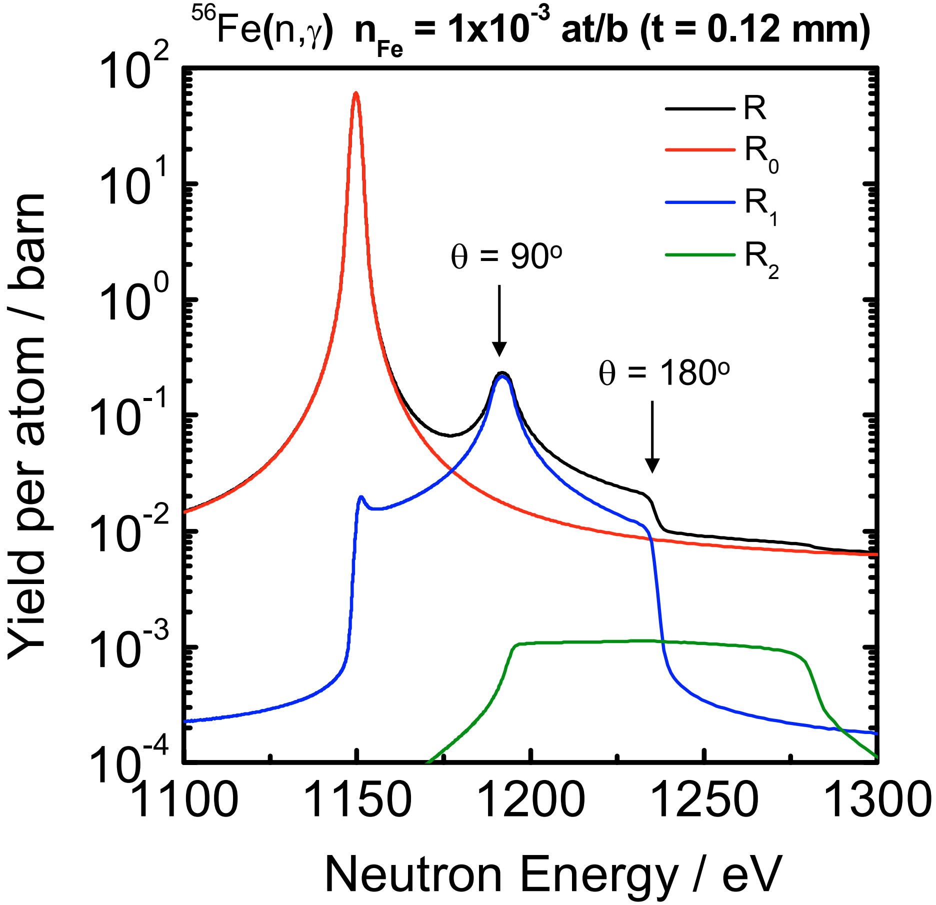
  – Self-shielding + 1,2 ,... scattering : $G_{R,2}$

Examples : $^{56}Fe(n,\gamma)$, $^{59}Co(n,\gamma)$ and $^{197}Au(n,\gamma)$

Reaction	E_R / eV	Γ_n / eV	Γ_γ / eV	Γ / eV	Δ_D / eV
$^{56}Fe + n$	1147.4	0.056	0.680	0.736	1.425
$^{59}Co + n$	132.0	5.150	0.470	5.620	0.470
$^{197}Au + n$	4.9	0.015	0.124	0.139	0.050



Self-shielding + multiple scattering for $^{56}Fe(n,\gamma)$ #1



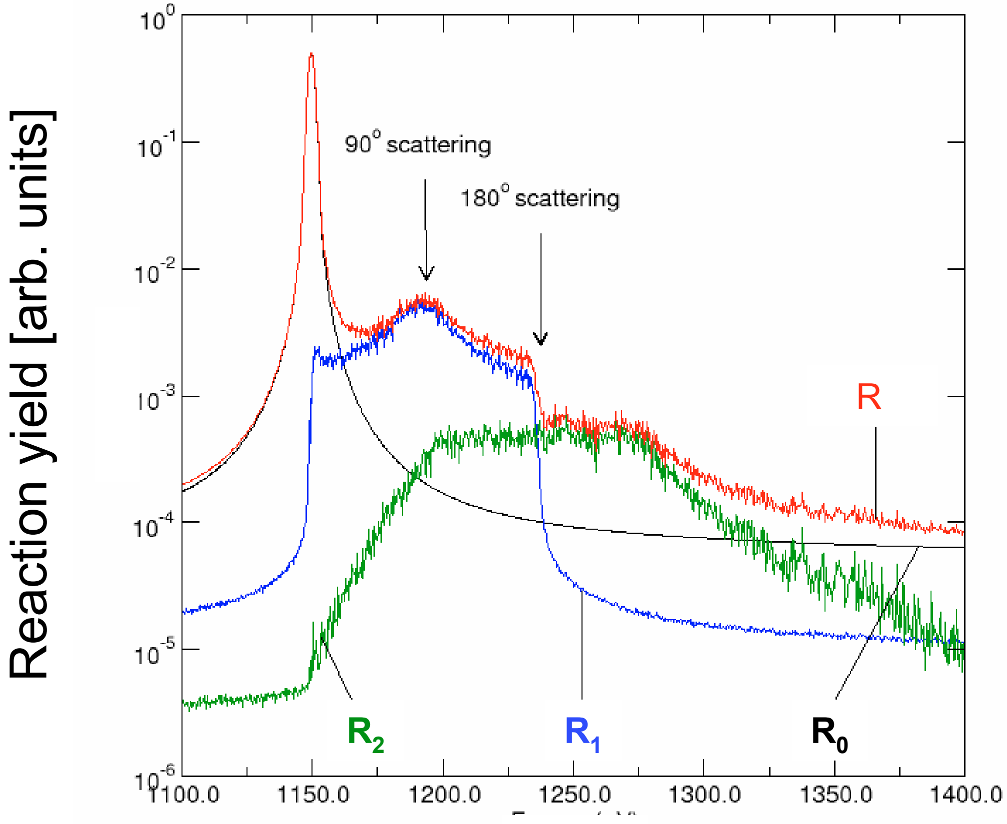
E_R = 1.15 keV in ^{56}Fe		
θ	E_n / keV	E_n' / keV
90°	1.192 keV	1.15 keV
180°	1.235 keV	1.15 keV
90° & 180°	1.280 keV	1.15 keV

$$\theta = 90^\circ \quad \Rightarrow \quad E_n' = E_n \frac{m_A - 1}{m_A + 1}$$

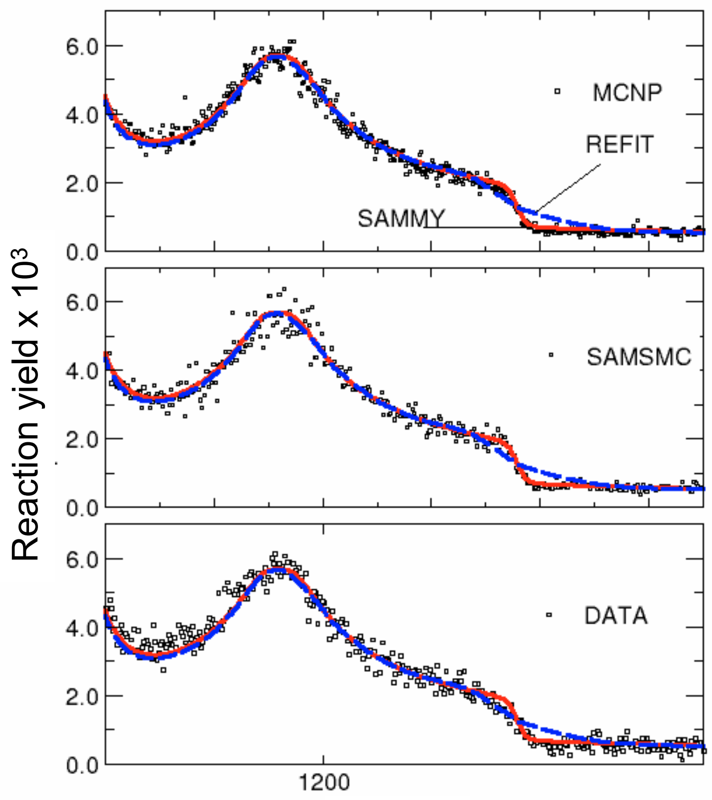
$$\theta = 180^\circ \quad \Rightarrow \quad E_n' = E_n \left(\frac{m_A - 1}{m_A + 1}\right)^2$$

Self-shielding + multiple scattering for $^{56}Fe(n,\gamma)$ #2

$n_{Fe} = 1.075\ 10^{-2}$ at/b, t = 1.3 mm



Neutron Energy / eV



Neutron Energy / eV

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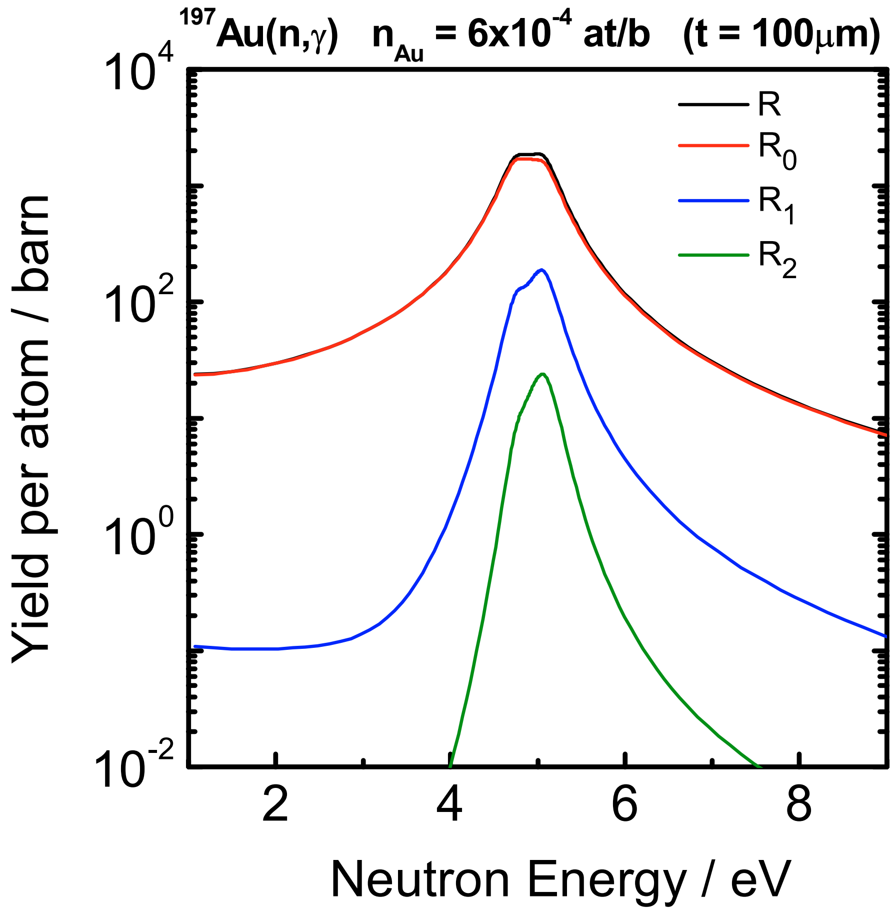
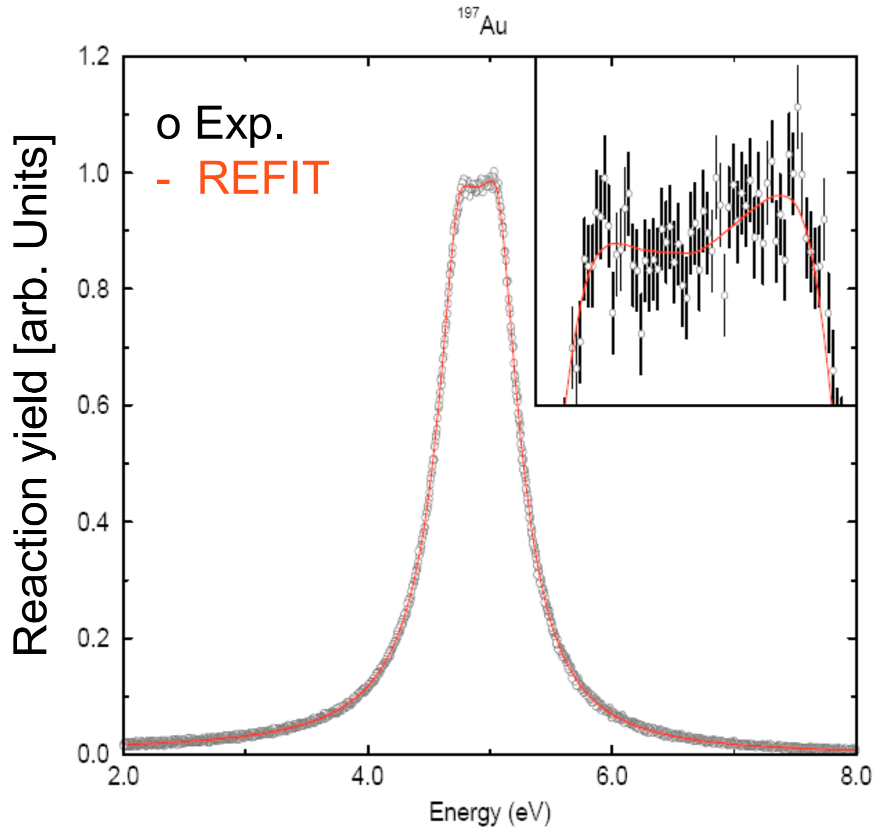
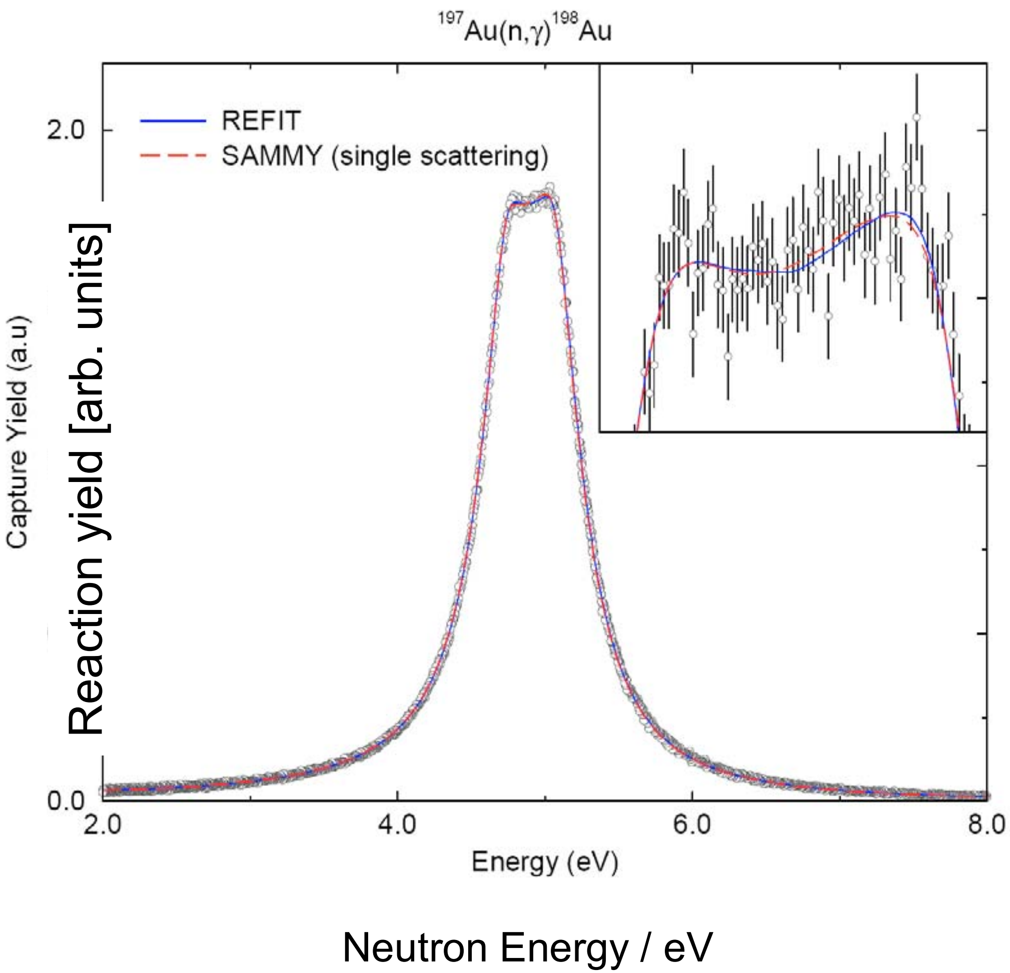
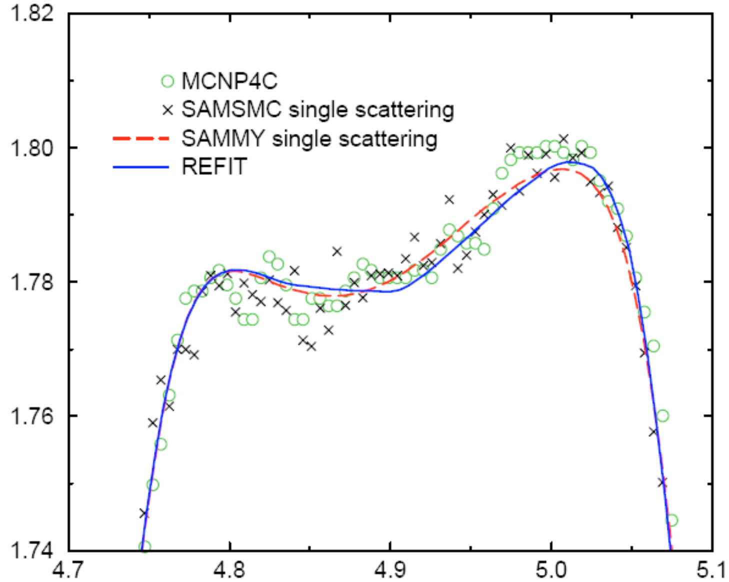
Self-shielding + multiple scattering for $^{197}Au(n,\gamma)$ #1





Self-shielding + multiple scattering for $^{197}Au(n,\gamma)$ #2





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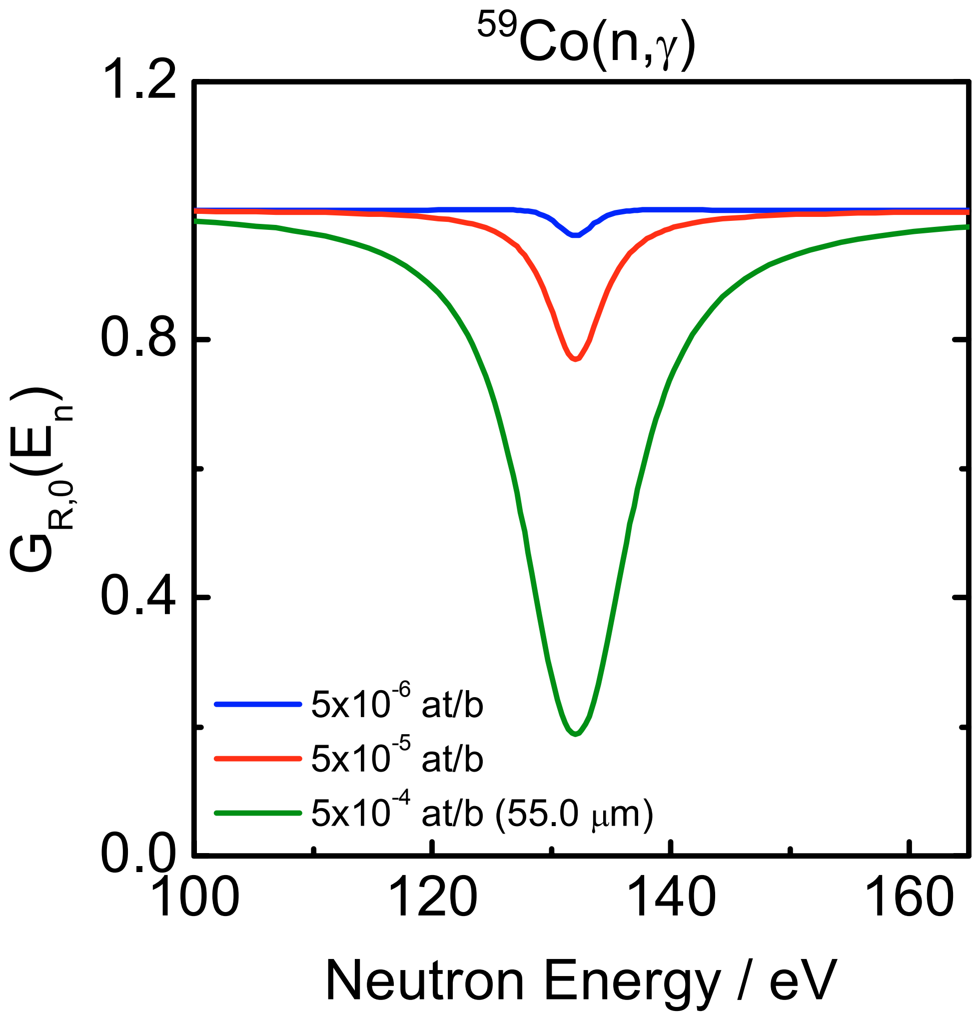
Influence of the resonance structure on G_R #1

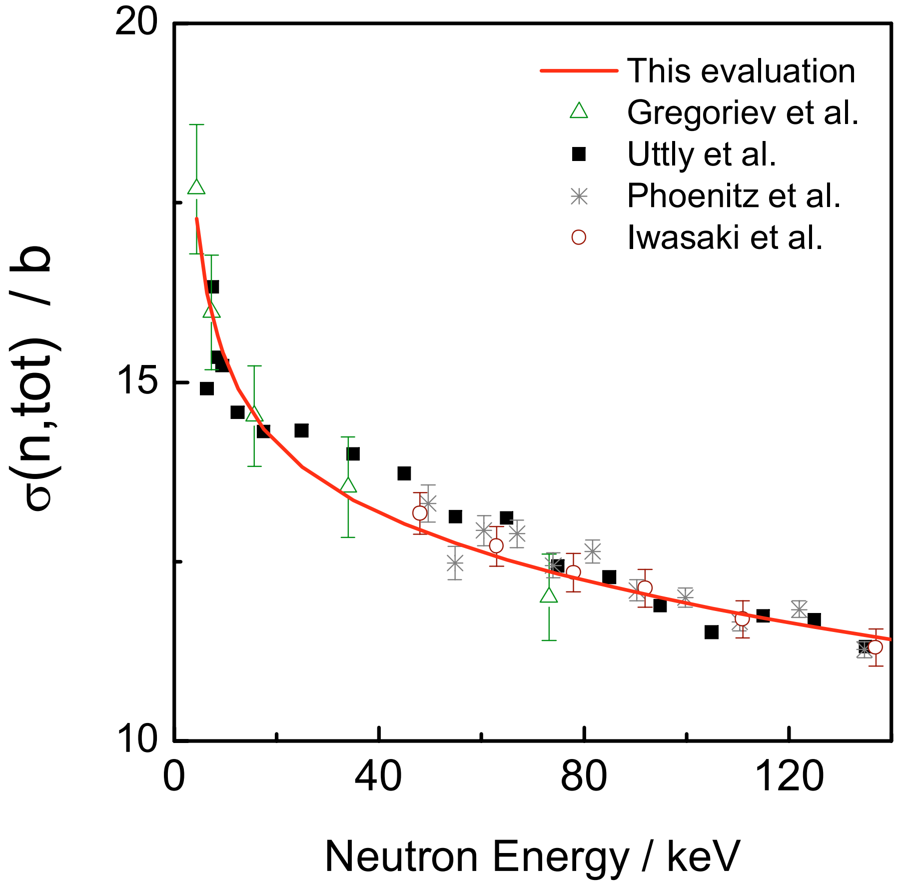


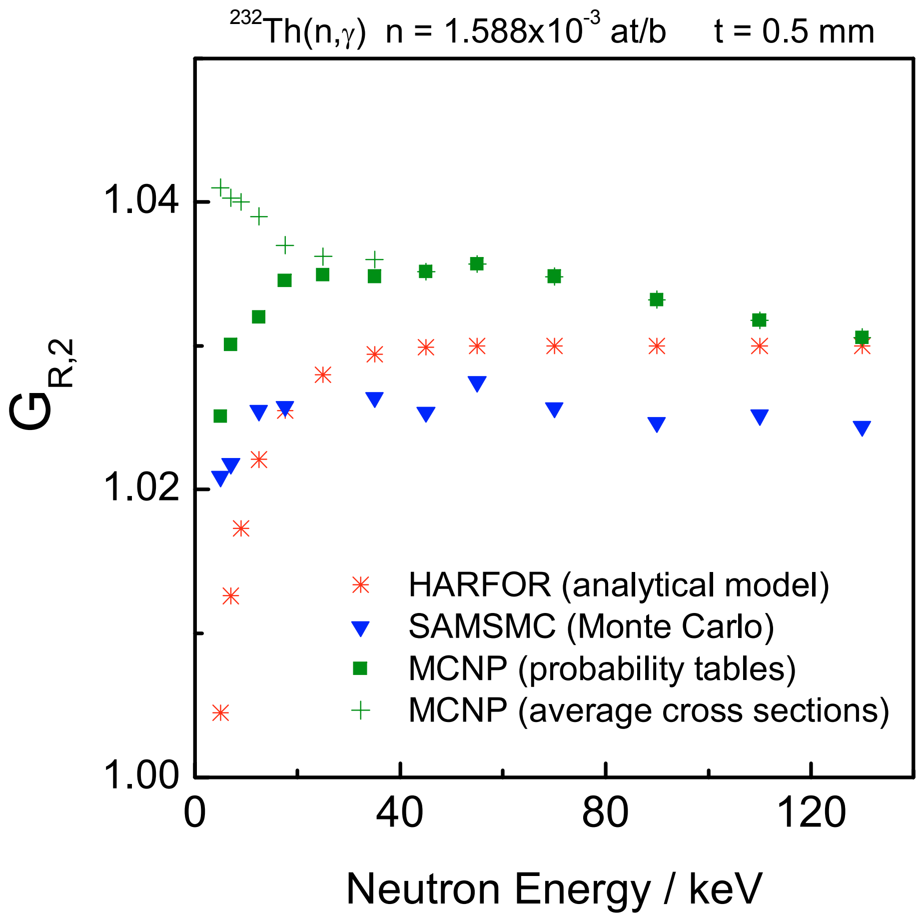
^{59}Co foil thickness		$(1-e^{-n\sigma_t}) / n\sigma_t$		$G_{R,0}$
		$<\sigma_t>$	$\sigma_{t,max}$	
at /b	μm	472 b	10539 b	
5×10^{-6}	0.55	1.00	0.97	0.99
5×10^{-5}	5.50	0.99	0.78	0.88
5×10^{-5}	55.0	0.89	0.19	0.46

$$< e^{-n\sigma_t} > \neq e^{-<n\sigma_t>}$$

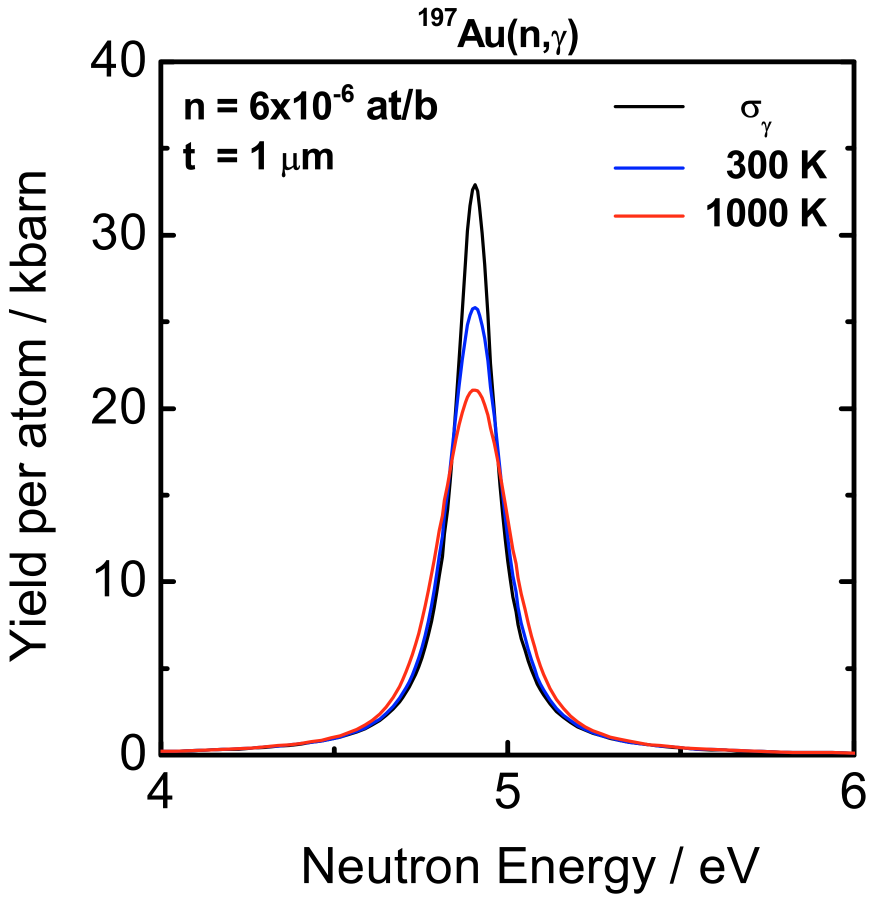
$$< e^{-n\sigma_t} > \approx e^{-<n\sigma_t>}\left(1 + \frac{n^2}{2}\,\mathrm{var}\,\sigma_t - \ldots\right)$$

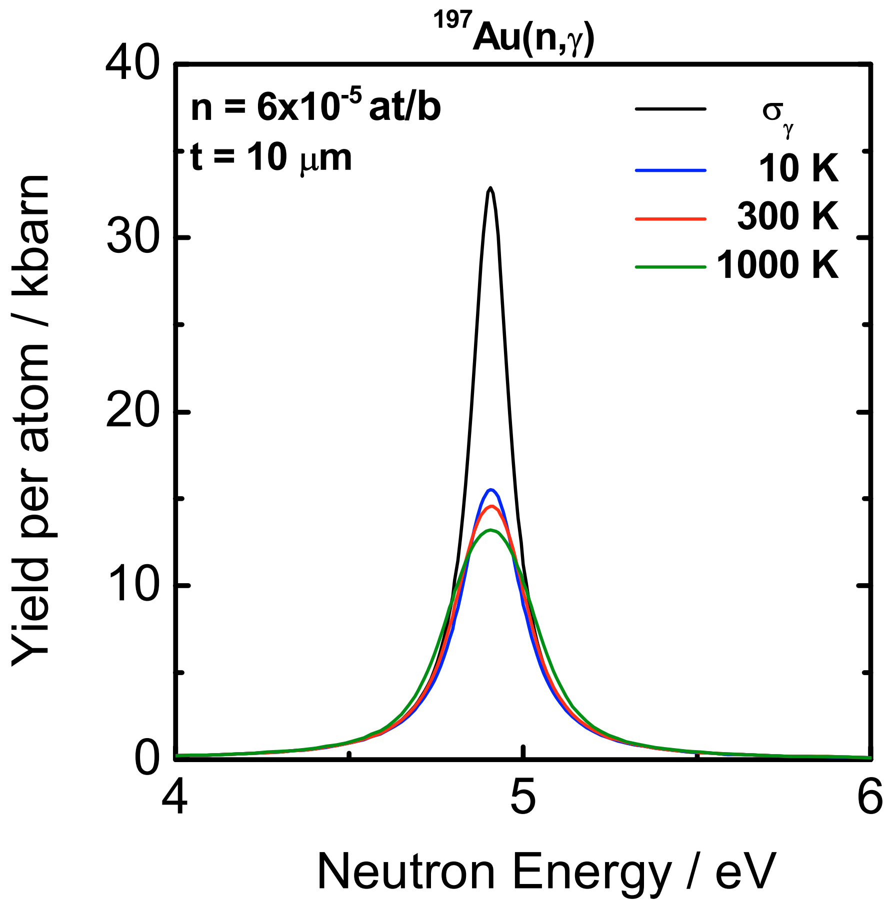
Influence of the resonance structure on G_R #2



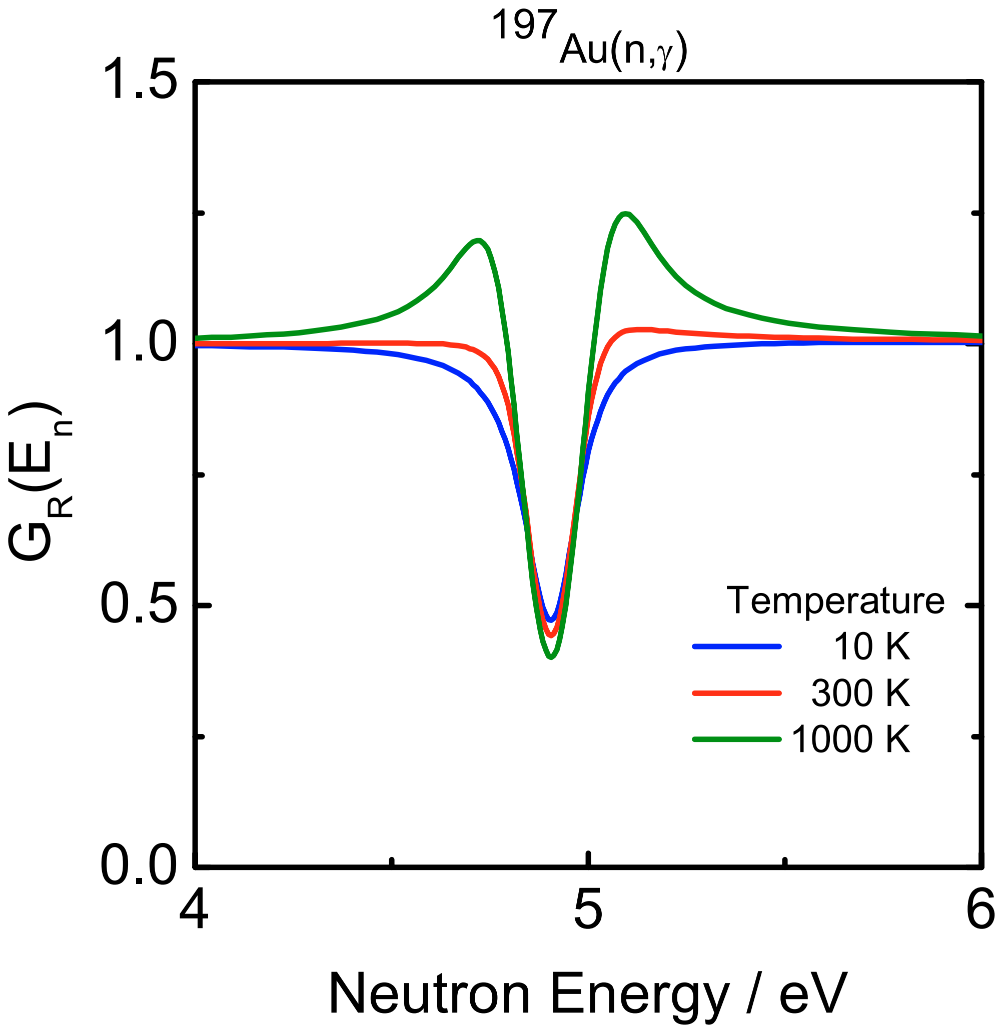


Influence of the Doppler effect   #1



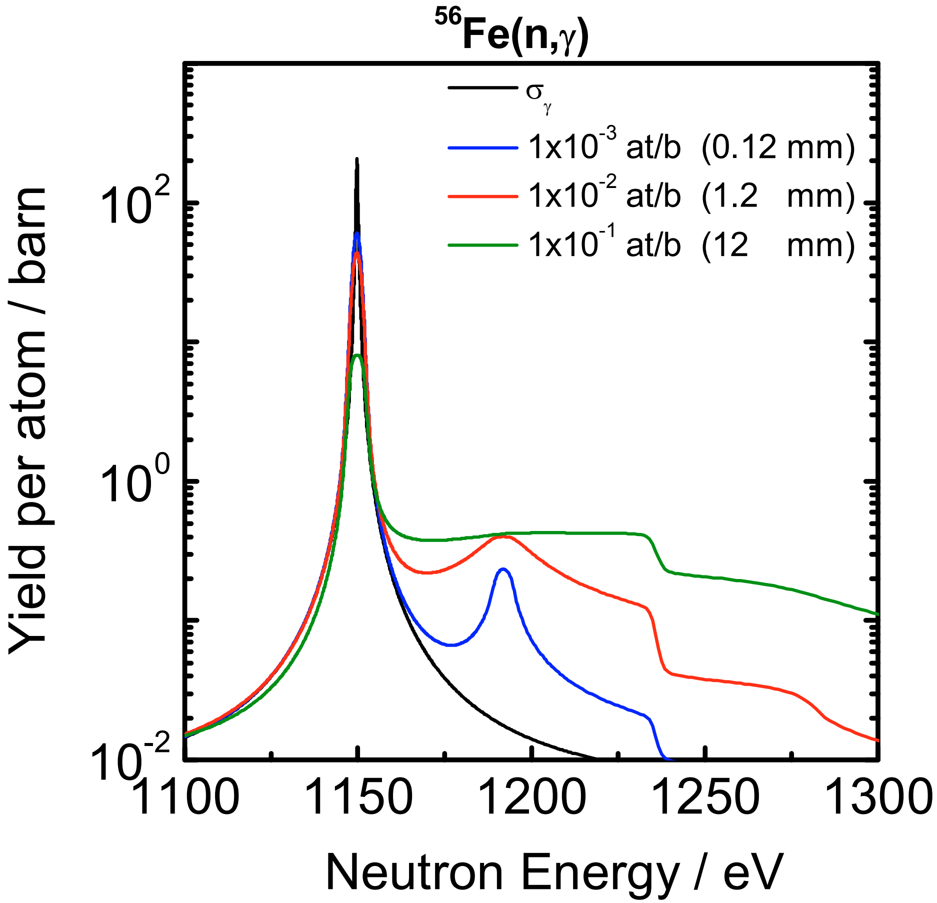


Influence of the Doppler effect #2



Temperature	Correction factor $G_{R,2}$	
	t = 1 μm	t = 10 μm
10 K		0.71
300 K	0.97	0.73
1000 K	0.98	0.76

Reaction yields and correction factors for $^{56}Fe(n,\gamma)$



Foil thickness		Correction factor		
at / b	mm	$G_{R,0}$	$G_{R,1}$	$G_{R,2}$
1×10^{-3}	0.12	0.97	0.99	0.99
1×10^{-2}	1.2	0.78	0.86	0.87
1×10^{-1}	12.0	0.23	0.35	0.47

E_R	=	1147.4	eV
Γ_n	=	0.056	eV
Γ_γ	=	0.680	eV
Γ	=	0.736	eV
Δ_D	=	1.425	eV

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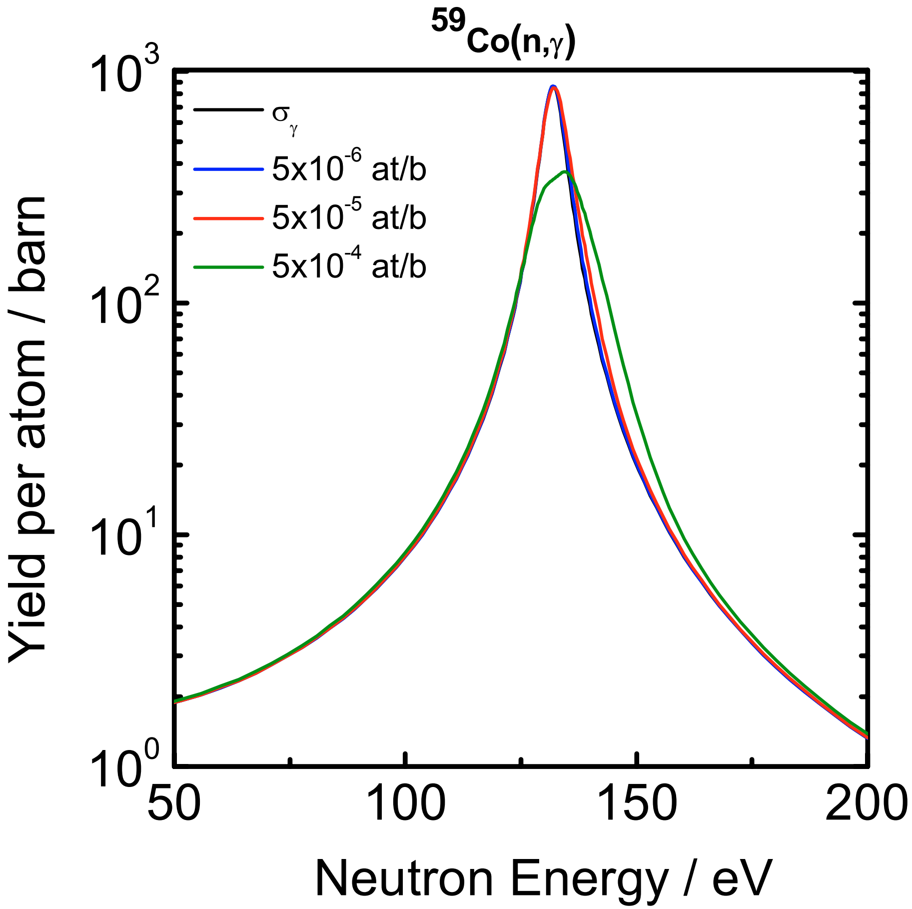
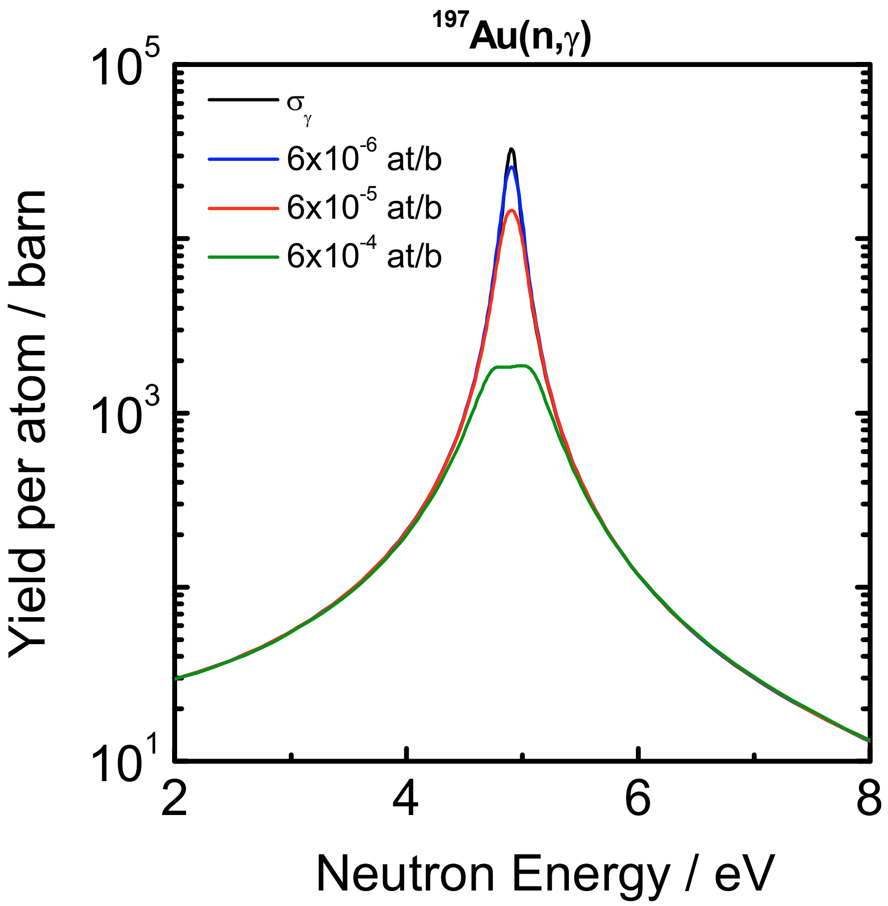
Reaction yields and correction factors for $^{59}Co(n,\gamma)$



Foil thickness		Correction factor		
at / b	μm	$G_{R,0}$	$G_{R,1}$	$G_{R,2}$
$5x10^{-6}$	0.55	0.99	1.02	1.03
$5x10^{-5}$	5.5	0.88	1.05	1.08
$5x10^{-4}$	55.0	0.46	0.67	0.86

E_R	=	132.0	eV
Γ_n	=	5.15	eV
Γ_γ	=	0.47	eV
Γ	=	5.62	eV
Δ_D	=	0.47	eV

Reaction yields and correction factors for $^{197}Au(n,\gamma)$



Foil thickness		Correction factor		
at / b	µm	$G_{R,0}$	$G_{R,1}$	$G_{R,2}$
6x10^{-6}	1.0	0.96	0.97	0.97
6x10^{-5}	10.0	0.69	0.73	0.73
6x10^{-4}	100.0	0.25	0.26	0.27

E_R	=	4.9	eV
Γ_n	=	0.015	eV
Γ_γ	=	0.124	eV
Γ	=	0.139	eV
Δ_D	=	0.050	eV

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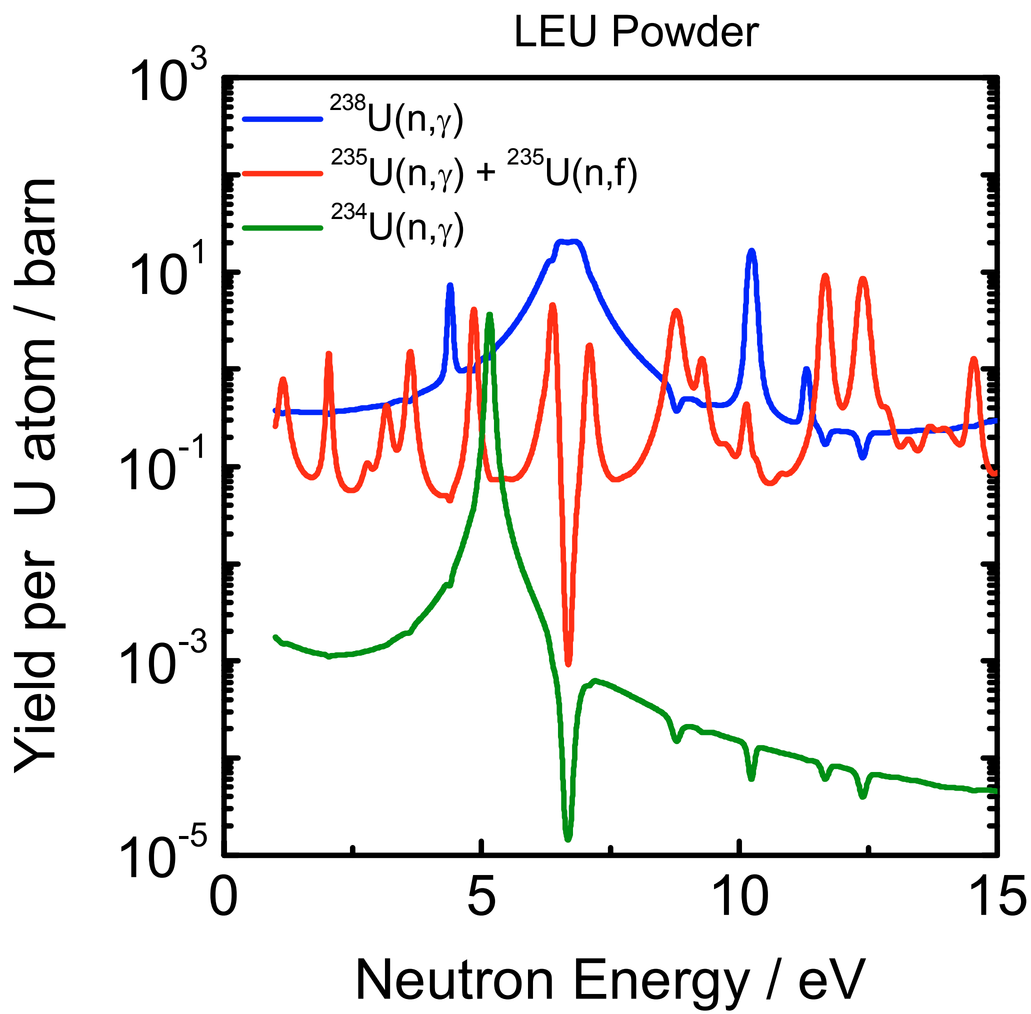
Interference effects, Uranium #1



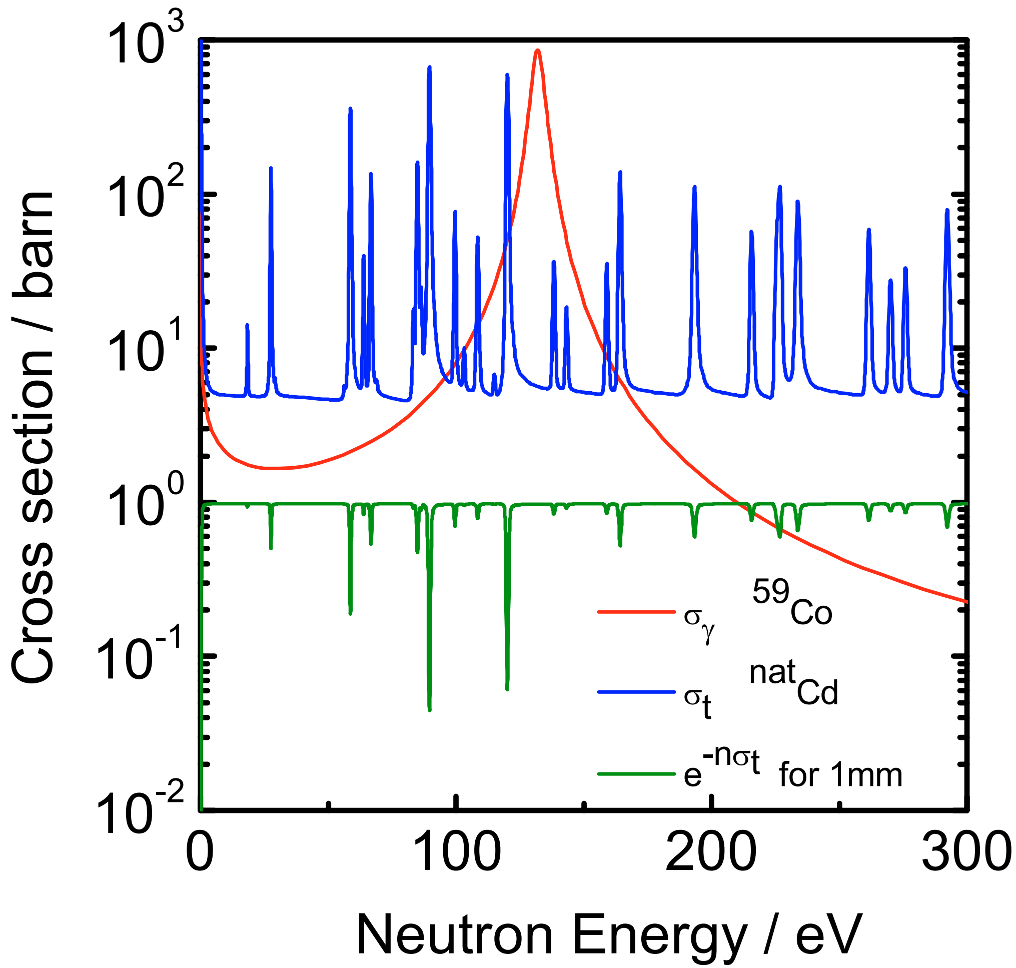
LEU Powder with 0.048 at/b U

^{238}U 96.97 wt%

^{235}U 3.00 wt%

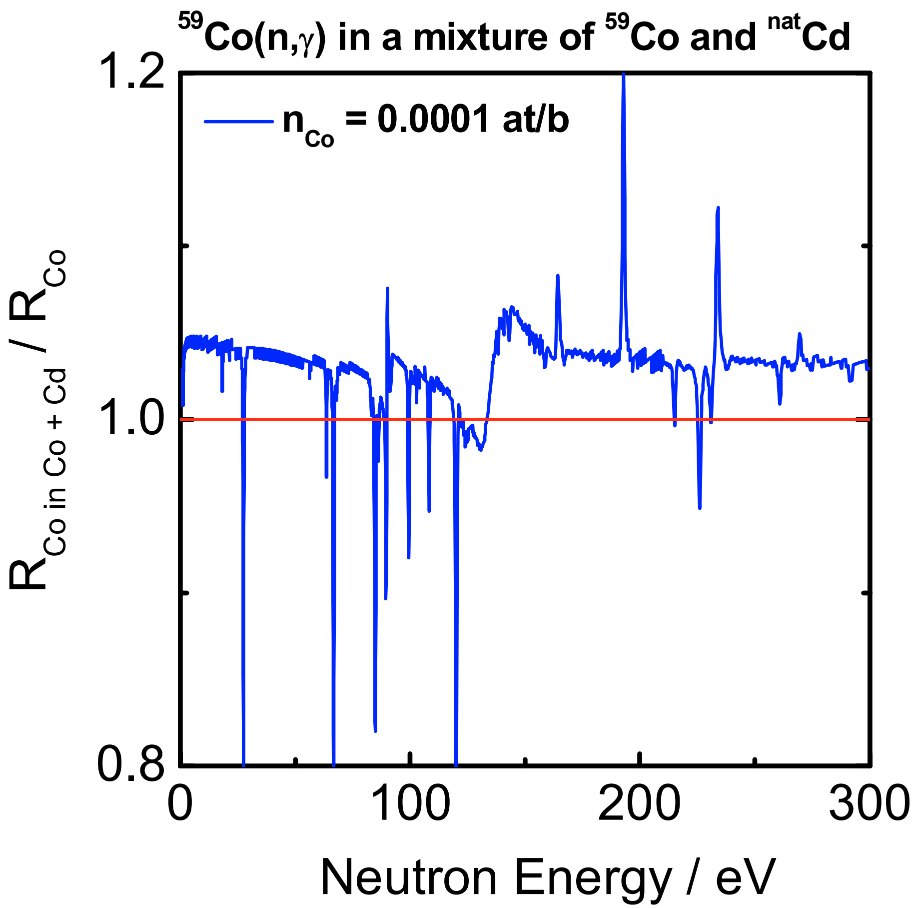
^{234}U 0.03 wt%

Interference effects, Cd-ratio measurements #2



$$F_{Cd} = \frac{R_{Cd}}{G_R \varphi_e I_R}$$

Interference effects, Cd-ratio measurements #3



$$F_{Cd} = \frac{R_{Cd}}{G_R \varphi_e I_R}$$

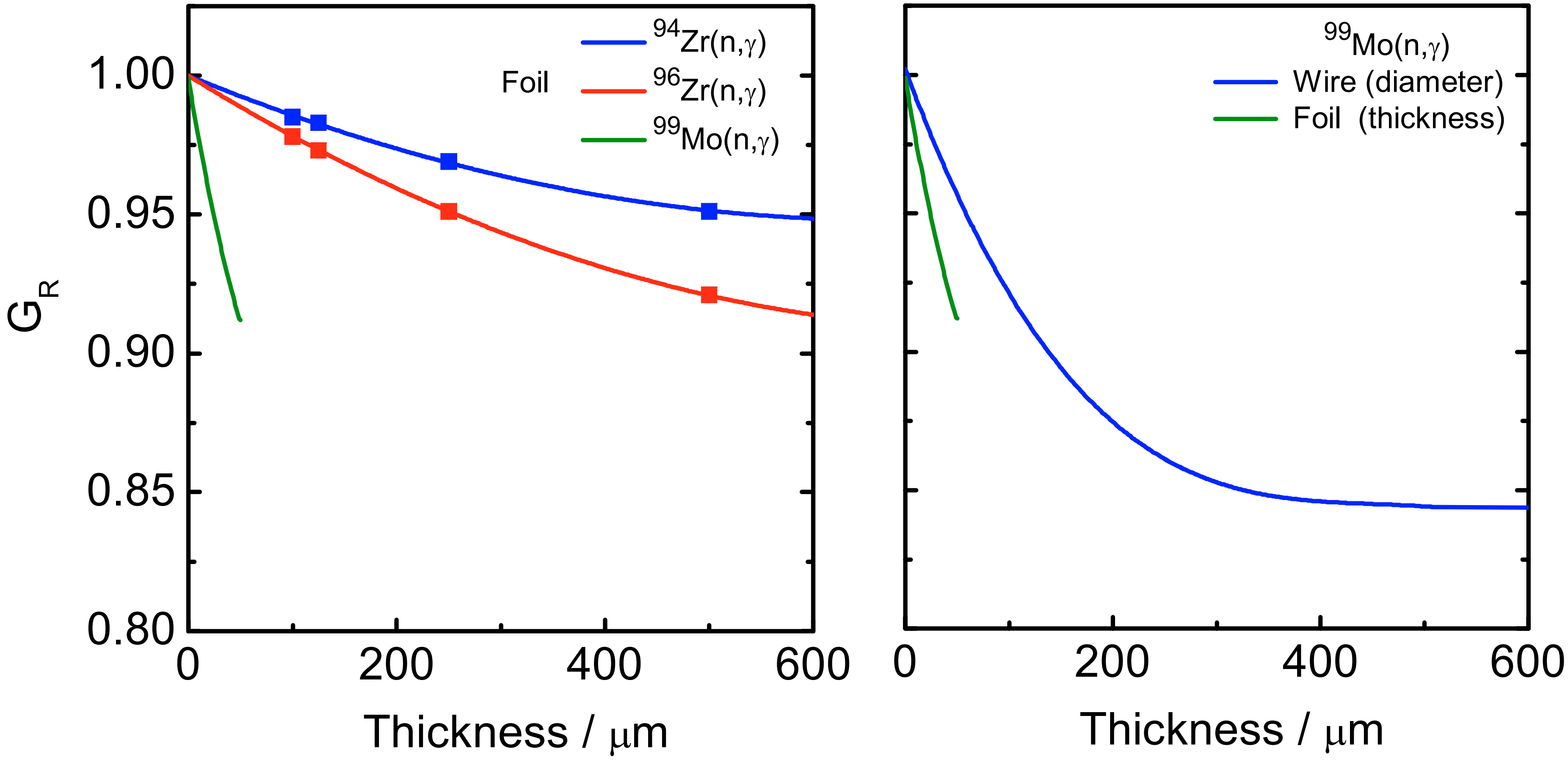
F_{Cd} is influenced by the resonance structure of the cross sections

5. Resonance self-shielding for an isotropic beam

Isotropic beam (NAA applications)

- **Experimental data**
- **Only self-shielding, no scattering & Doppler effect**
- **Self-shielding + Doppler**
- **Self-shielding + scattering**
- **Universal method**

Experimental G_R factors, F. De Corte et al.



Theoretical calculation, discussion #1

Reaction	E_R / eV	Γ_n / eV	Γ_γ / eV	Γ / eV	Δ_D / eV
$^{56}Fe + n$	1147.4	0.056	0.680	0.736	1.425
$^{59}Co + n$	132.0	5.150	0.470	5.620	0.470
$^{94}Zr + n$	2243.0	1.230	0.097	1.327	1.545
$^{96}Zr + n$	301.0	0.215	0.258	0.473	0.560
$^{197}Au + n$	4.9	0.015	0.124	0.139	0.050

$$\Delta_D = \sqrt{\frac{4\,kT\,E_R}{m_A}}$$

with k the Boltzman constant

Theoretical calculation, discussion $^{96}Zr(n,\gamma)$ #2

Foil	G_R for $^{96}Zr(n,\gamma)$		
	$G_{R,0}$ (theory, no scattering)		Experiment
Thickness (μm)	No Doppler Trubey*	With Doppler Roe*	De Corte
100	0.969	0.979	0.978
125	0.963	0.975	0.973
250	0.928	0.956	0.951
500	0.888	0.925	0.921

*Taken from P. De Neve, Thesis, Mol, 1992

De Corte 87, Aggregraatsproefschrift Hoger Onderwijs, Universiteit Gent, 1987

Γ_n = 0.21 eV
Γ_γ = 0.26 eV
Γ = 0.47 eV
Δ_D = 0.56 eV

$\Rightarrow$ Importance of Doppler effect increases with thickness especially for $\Delta_D \geq \Gamma$

Theoretical calculation, discussion $^{94}Zr(n,\gamma)$ #3

Foil thickness (μm)	G_R for $^{94}Zr(n,\gamma)$	
	$G_{R,0}$ with Doppler Roe*	Experiment De Corte
100	0.996	0.985
125	0.996	0.983
250	0.992	0.969
500	0.986	0.951

*Taken from P. De Neve, Thesis, Mol, 1992

De Corte 87, Aggregraatsproefschrift Hoger Onderwijs, Universiteit Gent, 1987

Γ_n = 1.23 eV
Γ_γ = 0.10 eV
Γ = 1.33 eV
Δ_D = 1.55 eV

$\Rightarrow$ Importance of multiple scattering for $\Gamma_n > \Gamma_\gamma$

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Theoretical calculation, discussion $^{59}Co(n,\gamma)$ #4

Foil Thickness (μm)	G_R for $^{59}Co(n,\gamma)$	
	Theory ($R_o + R_1$) No Doppler Lopes and Avila	Experiment Eastwood and Werner
10.2	0.810	0.840
22.9	0.690	0.700
25.4	0.675	0.630
50.8	0.554	0.590
91.4	0.455	0.460
101.6	0.438	0.450

M.C. Lopes and J.M. Avila, Nucl. Sci. & Eng. , 104 (1990) 40

T.E. Eastwood and R.D. Werner, Nucl. Sci. & Eng., 13 (1962) 385

Γ_n = 5.15 eV
Γ_γ = 0.47 eV
Γ = 5.62 eV
Δ_D = 0.47 eV

$\Rightarrow$ Importance of multiple scattering for $\Gamma_n > \Gamma_\gamma$

Universal curve #1

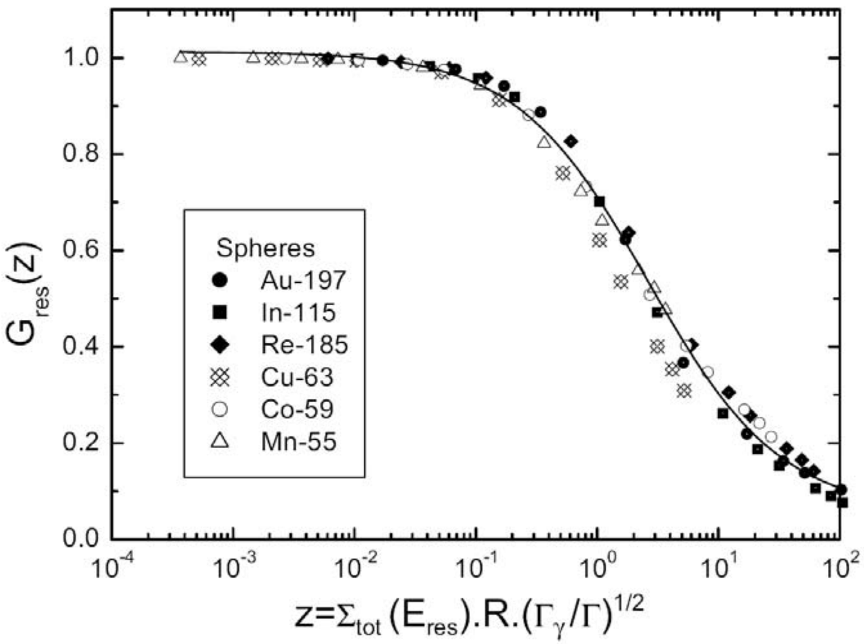
E. Martinho et al., "Universal curve of epithermal neutron self-shielding factors in foils, wires and spheres", J. Appl. Rad. And Isot., 58 (2003) 371

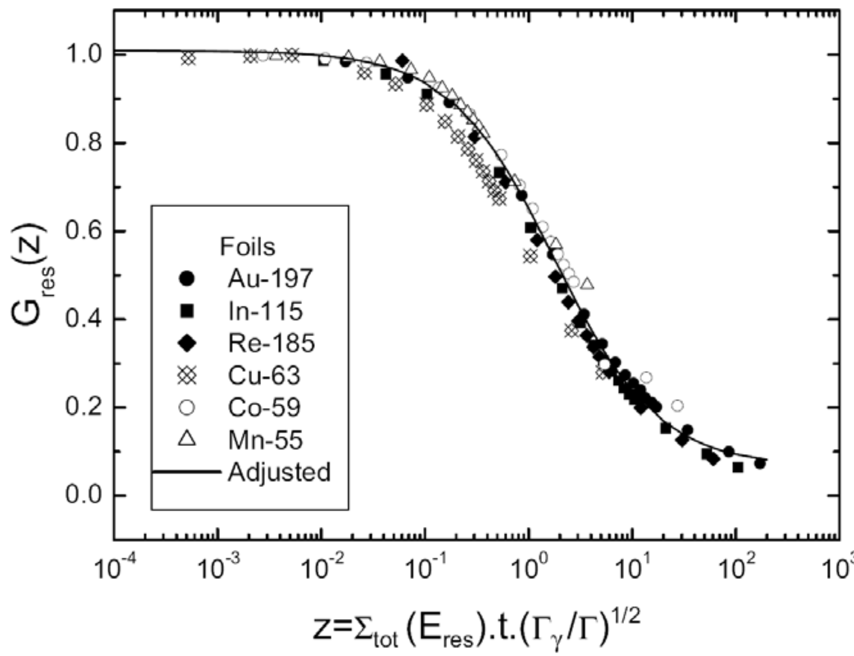
Express G_R as a function of the variable $z = \Sigma_t(E_r)\sqrt{\frac{\Gamma_\gamma}{\Gamma}}\,y$

$y = R$ for wires

$y = R$ for spheres

$y = t$ for foils





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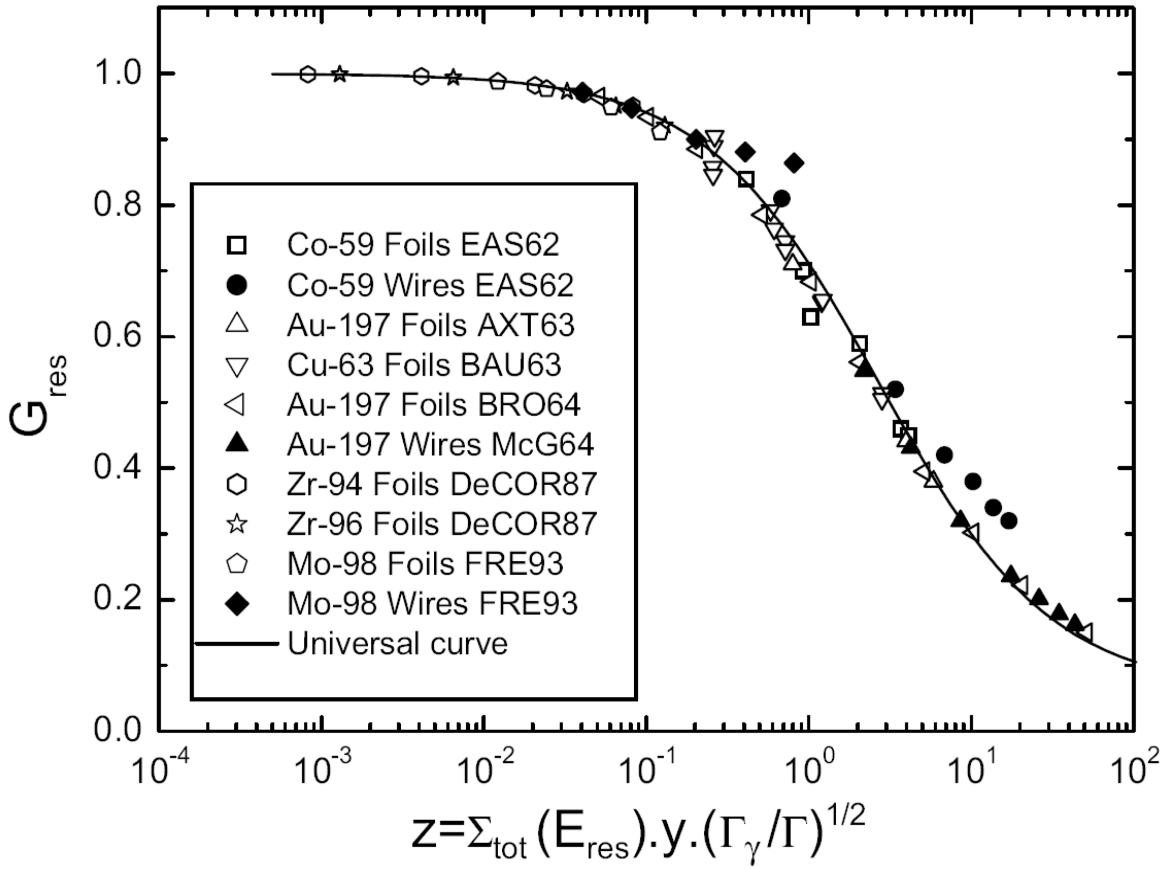
Universal curve #2

$$G_{res} = \frac{A_1 - A_2}{1 + (z/z_0)^p} + A_2$$

$$z = \Sigma_t(E_r)\sqrt{\frac{\Gamma_\gamma}{\Gamma}}\, y$$



$y = 2R$ for wires

$y = R$ for spheres

$y = 1.5t$ for foils

Summary

- **The importance of the resonance structure for NAA**
  - Thermal capture cross section
  - 1/v behaviour of cross sections
  - Westcott g_w - factor, $g_w(T)$
  - Self-shielding and multiple scattering corrections, G_R
- **A first estimate of the correction factor for self-shielding and scattering is given by E. Martinho et al.**
- **More accurate correction factors can be calculated by analytical expressions and Monte Carlo simulations if all relevant effects are accounted for**
  - Resonance structure (e.g. MCNP use probability tables)
  - Neutron scattering
  - Doppler effect
- **Solution:**

F. De Corte et al., J. Radioanal. and Nucl. Chem., 179 (1994) 93

"In general, the best way to solve the problem of self-shielding is to avoid it "