

IAEA Workshop On the Experimental Nuclear Reaction Database (EXFOR) 6 – 10 October 2014



Title

Vladimir Varlamov

"Some problems of photonuclear data compilation and evaluation".

10/10/2014







Photon-induced reaction data, primarily total and partial photoneutron reaction cross sections, are of essential importance for basic research and a variety of applications in nuclear physics, astrophysics and many other related fields.

The majority of widely used data are those obtained with various experimental methods in the energy range of Giant Dipole Resonance. They form the foundation for investigation of fundamental features of electromagnetic interactions of atomic nuclei.

For many reasons, however, data obtained with the different methods are not consistent to each other.







Experimental photonuclear data research is complicated problem for several reasons:

• absence till now of intensive monoenergetic photon beams (experimentalists are forced to use various methods for production of conditions of quasimonoenergetic photon beams or to use special mathematical methods for unfolding the results obtained by bremsstrahlung);

• photoneutron reaction contribution as main part of Giant Dipole Resonance (experimentalists are forced to detect neutrons using detectors with low efficiency and not enough accurate methods for measurements of neutron energies);

• **direct methods** of neutron registration lead to frequently lost contributions of accompanying (because low values of correspondent reaction thresholds) protons;

• alternative activation methods have many restrictions concern the properties of final nucleus decay.







As a result we have many data with significant systematic uncertainties obtained using various experimental measurement methods and/or data processing procedures.

Therefore there are many problems of photonuclear data compilation and evaluation.

I would like to speak about the more important things concern photonuclear reaction cross section data.







Principal problem for EXFOR compilation: definition of the total REACTION (SF1(SF2,SF3)SF4,,SIG).

The first problem is relatively simple – the total photoneutron reactions

for the yield reaction $(\gamma,\mathbf{n}) + 2(\gamma,2\mathbf{n}) + 3(\gamma,3\mathbf{n})$

(SF1(G,X)0-NN-1,SIG)

in many articles (primarily eastern) description (γ, Xn) is used, **but in many others (primarily western)** - (γ, Sn) is used;

for the total photoneutron reaction (SF1(G,X)0-NN-1,UNW,SIG) $(\gamma,n) + (\gamma,2n) + (\gamma,3n)$

correspondingly (primarily eastern) (γ, Sn) is used, but (primarily western)

 (γ, tot) is used.

Therefore the EXFOR compiler should be very careful in definition what does total reaction (γ, Sn) means in reality.

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Partial photoneutron reactions









Partial reactions as combinations in reality.

There are two main different methods for photonuclear reaction identification:

- the direct identification of the reaction via final nucleus SF4 using method of induced activity (for example in reactions (γ ,n) and (γ ,2n) final nuclei are different having different spectra of de-excitation γ -quanta); therefore definite SF4 \rightarrow SF3.

- not direct identification using detection of outgoing particles SF3 and their multiplicity sorting (for example (γ ,n) and (γ ,2n) are separated by measurement of different neutron energies; therefore definite SF3 \rightarrow SF4:

But there is serious problem: in many cases at the same energies reactions with different outgoing particles can occur:

- for detection of 1 neutron we have not definitely (γ,n) reaction but a sum of reactions $(\gamma,n) + (\gamma,np)$ and more maybe $(\gamma,n) + (\gamma,np) + (\gamma,n2p)$;
- for detection of 2 neutrons we have not definitely $(\gamma,2n)$ reaction but a sum of reactions $(\gamma,2n) + (\gamma,2np)$ and more maybe $(\gamma,2n) + (\gamma,2np) + (\gamma,2n2p)$;
- for detection of 1 proton we have not definitely (γ, p) but $(\gamma, p) + (\gamma, pn) + \dots$ etc.







Principal problem for EXFOR compilation: definition of the partial REACTION (SF1(SF2,SF3)SF4,,SIG).

The second problem – partial photoneutron reactions

• in many cases using direct neutron detection the following partial reactions are investigated in reality because low thresholds of (γ,np) and $(\gamma,2np)$ reactions:

not (γ,n)	(SF1(G,N)SF4,,SIG)
but $(\gamma,n) + (\gamma,np)$	$[(SF1(G,N)SF4,,SIG)+[(SF1(G,N+P)SF4_{p},,SIG)]$
not (γ,2n)	(SF1(G,2N)SF4,,SIG)
but $(\gamma,2n) + (\gamma,2np)$	$[(SF1(G,2N)SF4,,SIG)+[(SF1(G,2N+P)SF4_{p},,SIG)] etc.$
not (γ,p)	(SF1(G,P)SF4,,SIG)
but $(\gamma,p) + (\gamma,pn)$	$[(SF1(G,P)SF4,SIG)+[(SF1(G,N+P)SF4_n,SIG)]]$ etc.

but

• in all cases using activation method namely (γ,n) , (γ,np) , $(\gamma,2n)$, $(\gamma,2n)$, $(\gamma,2np)$, (γ,p) , $(\gamma,2p)$ etc. reactions are investigated separately.

Therefore the EXFOR compiler should be very careful in partial reaction definition.

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The majority of photonuclear reaction cross section data were obtained using two types of photon beams:

- electron bremsstrahlung γ–quanta (betatrons or microtrons, Moscow, Saratov (Russia), Melbourne (Australia),...;

- quasimonoenergetic photons from positron annihilation in flight (linac, Livermore (USA), Saclay (France),..., General Atomic (USA), Pennsilvania (USA), Giessen (Germany),...

Significant difference in cross section data obtaining conditions leads to serious problems for both compilation and evaluation.







Electron bremsstrahlung photons

$$Y(E_{jm}) = \frac{N(E_{jm})}{\varepsilon D(E_{jm})} = \alpha \int_{Eth}^{Ejm} W(E_{jm}, k) \sigma(k) dk,$$

where $\sigma(k)$ is cross section at photon energy k of reaction with threshold E_{th} ; $W(E_{jm},k)$ is electron bremsstrahlung spectrum; $N(E_{jm})$ is reaction event number, $D(E_{jm})$ is γ -dose, ε is detector efficiency.

Because the spectrum of bremsstrahlung γ -quanta is continuous the only investigated reaction yield – folding of cross section with γ -quanta spectrum can be measured.



There is important to point out: the resonances in cross section are reflecting in the reaction yield energy dependence as breaks (or kinks).

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Reaction cross section $\sigma(k)$ is the result of solution of the inverse task of unfolding

$$Y(E_{jm}) = \frac{N(E_{jm})}{\varepsilon D(E_{jm})} = \alpha \int_{Eth}^{Ejm} W(E_{jm}, k) \sigma(k) dk,$$

with definite apparatus function effective γ -spectrum obtained using various methods:



Some important things:

- apparatus function has narrow and clear localized line;
- apparatus function has complex (not ideal, for example, Gauss line) shape that can produce some additional uncertainties in cross section shape, magnitude and position.

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Quasimonoenergetic annihilation photons (an alternative to procedure of solving inverse ill-post task)

Three step measurements (example for experimental data for reaction ${}^{63}Cu(\gamma,n){}^{62}Cu$ (L0013,R.E.Sund+,J,PR,176,1366,1968):

1) yield $Y_{e+}(E_j)$ of reaction induced by photons from positron's both bremsstrahlung and annihilation;

- 2) yield $Y_{e_i}(E_i)$ of reaction induced by photons from electron bremsstrahlung;
- 3) subtraction of bremsstrahlung "tail": $Y_{e+}(E_j) Y_{e-}(E_j) = Y(E_j) \approx \sigma(k)$.



3) subtraction "1) – 2)":

 $\mathbf{Y}_{e^+}(\mathbf{E}_j) - \mathbf{Y}_{e^-}(\mathbf{E}_j) = \mathbf{Y}(\mathbf{E}_j) \approx \sigma(\mathbf{k});$

2) measurement of Y_e.(E_j) using electrons;

1) measurement of $Y_{e+}(E_j)$ using positrons.







Some important things:

• in each concrete experiment apparatus function is obtained individually because directly depends on both measured results (yields) and their normalization;

• positron annihilation in flight occurs in many steps (bremsstrahlung production by electrons ($e^- + A \rightarrow A + e^- + \gamma$), pair production by bremsstrahlung photons ($\gamma + A \rightarrow A + e^- + e^+$), positron annihilation ($e^+ + e^- \rightarrow 2\gamma$); therefore number of quasimonoenergetic photons and hence measured yields statistical uncertainty and hence normalization accuracy are small.

Principal problem for EXFOR compilation and evaluation:

the difference between two reaction yields Ye+ - Yeis again the only yield Y but not cross section σ .

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More correctly: the result of subtraction

Ye+ - Ye-

can be interpreted as cross section $Y \approx \sigma$ but with very low energy resolution typical to resolution of yield and not equal to the width of annihilation line:









So data obtained using quasimonoenergetic annihilation photons should be strongly over-smoothed (real energy resolution is several (4 -6) times worse than declared one) in comparison with estimation based on calculated with of annihilation line in photon spectrum.

The reason for that is quite simple. The difference procedure used is oriented to bremsstrahlung "tail" subtracting but not for high resolution obtaining: both experimental results Ye+ and Ye- have bad resolution determined by sum of large number of bremsstrahlung photons and small number of annihilation photons.

Therefore the resolution of difference Ye+ - Ye- = Y $\approx \sigma$ can not be attributed only to annihilation line.









Smoothing





Taking into account the photonuclear experiment apparatus function



$$\Sigma = \begin{bmatrix} \Delta Y_1^2 & \dots & \Delta Y_2^2 & \dots & \\ & \Delta Y_2^2 & \dots & \\ & & \dots & \\ 0 & & \dots & \Delta Y_n^2 \end{bmatrix}$$

At condition $M \| Ry - U\sigma \| = \min$ the vector of solution exists

$$R = U(\Sigma^{-1/2}A)^{-}\Sigma^{-1/2} = U(A^{*}\Sigma^{-1}A)^{-}A^{*}\Sigma^{-1}$$

$$\stackrel{\land}{\sigma} = Ry = RA\sigma + R\nu = U\sigma + R\nu$$

$$G = R\Sigma R^{*} = U(A^{*}\Sigma^{-1}A)^{-}U^{*}.$$

ゝ

σ is interpreted as result disturbed by noise Rν of measurement of cross section by apparatus U with apparatus function of needed quality, for example gauss line with definite resolution and G is the matrix of uncertainties. 10/10/2014 17

17 Reduction method



Photon energy, MeV

Cross section, mb







Principal problem for EXFOR compilation: definition of the REACTION (SF1(SF2,SF3)SF4,,,,,BRS) and INC-SOURCE (MPH) or (QMPH) and (BRST).

The problems in energy resolution mentioned lead to problems of compilation data for

REACTION and INC-SOURCE

because data obtained using quasimonoenergetic annihilation photons beams in reality

is not BRST

but in reality also

is not MPH or QMPH.

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19 BRST–MPH







Additional principal problem for EXFOR compilation of data obtained using bremsstrahlung: definition of the REACTION (SF1(SF2,SF3)SF4,,,,) INC-SOURCE (KINDT).

> For data obtained using bremsstrahlung INC-SOURCE (BRST) is used in combination with REACTION (SF1(SF2,SF3)SF4,,,,,BRS)

At the same time there are many experiments carried out using bremsstrahlung but in conditions of complete kinematics: energies of all outgoing particles are measured and therefore the energy of γ -quanta induced reaction also can be obtained definitely.

In such cases

not

REACTION (SF1(SF2,SF3)SF4,,,,BRS) INC-SOURCE (BRST)

but

REACTION (SF1(SF2,SF3)SF4,,,,) INC-SOURCE (KINDT)

should be used.







Additional principal problem for EXFOR evaluation of data obtained using quasimonoenergetic annihilation photons – disagreements of data from different laboratories.

Citation from S.S.Dietrich and B.L.Berman "Atlas of photoneutron cross section obtained with monoenergetic photons", Atomic Data and Nuclear Data Tables, 38 (1988) 199- 338:

"No attempt is made...to chose between two sets of data for the same nucleus measured at different laboratories or to compromise between them by presenting a set of recommended intermediate values...

When agreement and consistency between measurements are not one usually (but not always) is best advised to weight more recent data more heavily than earlier data".







Systematics:

many experimental data data for partial photonuclear reaction cross sections obtained in period 1962 - 1986 (the majority was obtained at Livermore (USA) and Saclay (France)), are published in

Atlas of Photoneutron cross sections obtained with monoenergetic photons (S.S.Dietrich, B.L.Berman. Atom. Data and Nucl. Data Tables, 38 (1988) 199)

Berman's library - EXFOR entries L0001 – L0059 (~ 174 nuclei sets)





¹³³Cs



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¹⁵⁹Tb



xn-sn-n-2n-3n



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Main problem for 20 nuclei investigated in both Labs:

 $(\gamma, 1n)$ cross sections are larger at Saclay but those for $(\gamma, 2n)$ - at Livermore.

V.V.Varlamov, N.N.Peskov, D.S.Rudenko, M.E.Stepanov. Consistent Evaluation of Photoneutron Reaction Cross Sections Using Data Obtained in Experiments with Quasimonoenergetic Annihilation Photon Beams at Livermore (USA) and Saclay (France) in Articles Translated from Journal Yadernye Konstanty (Nuclear Constants). INDC(CCP)-440, IAEA NDS, Vienna, Austria, 2004, pp. 37 – 85.





Main objective criterium for data reliability

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The natural and physically reliable energy dependence of F₂ should be

- Below the $(\gamma, 2n)$ reaction threshold B2n only the $(\gamma, 1n)$ reaction is possible: $F_2 = 0$;

- Above B2n both $(\gamma, 1n)$ and $(\gamma, 2n)$ reactions are possible, F₂ increases due to competition between decreasing $\sigma(\gamma, 1n)$ and increasing $\sigma(\gamma, 2n)$, going to the theoretical limit of 0.50, but never reach it because of a high-energy part in

- Above the B3n threshold the (γ, 3n) reaction is also possible, F₂ decreases due to a 3 $\sigma(\gamma, 3n)$.

 \mathbf{F}_2



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Some examples of Livermore data



Physically not reliable negative cross section values are correlated with physically forbidden values $F_2 > 0.50$

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26 F₂ - examples



There are additional physically natural criteria: $F_1 = \sigma(\gamma, 1n) / \sigma(\gamma, xn) < 1.00$ $F_3 = \sigma(\gamma, 3n) / \sigma(\gamma, xn) < 0.33$ etc.

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" $F_2 > 0.50$ " correlates with negative $\sigma(\gamma, n)$ values.

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New experimentally-theoretical method of evaluation

using combined model of photonuclear reactions:

- initial data – experimental neutron yield reaction (γ, xn) cross section;

- sorting neutrons for multiplicity based on theoretical model.

Theoretically calculated transitional multiplicity functions $F_i^{theor} = \sigma^{theor}(\gamma, in) / \sigma^{theor}(\gamma, xn)$ are used for cross section evaluation by following way

 $\sigma^{\text{eval}}(\gamma, \text{in}) = F_i^{\text{theor}}(\gamma, \text{in}) \bullet \sigma^{\text{exp}}(\gamma, \text{xn}).$

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Our new approach for evaluation of partial traction cross sections

 $\sigma^{\text{eval}}(\gamma, \text{in}) = F_i^{\text{theor}}(\gamma, \text{in}) \bullet \sigma^{\text{exp}}(\gamma, \text{xn}).$

means:

- i) the competition of partial reactions $(\gamma, 1n)$, $(\gamma, 2n)$ and $(\gamma, 3n)$ is in accordance with equations of model;
- ii) the sum of evaluated partial reaction cross sections

 $\sigma^{\text{theor}}(\gamma, xn) = \sigma^{\text{theor}}(\gamma, 1n) + 2\sigma^{\text{theor}}(\gamma, 2n) + 3\sigma^{\text{theor}}(\gamma, 3n)$

is equal to the experimental $\sigma^{exp}(\gamma, xn)$.





Disagreements – ¹¹⁵**In**



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Noticeable differences between evaluated and experimental data







Reaction	Integrated cross section σ ^{int} , MeV∙mb				
	Livermore	Evaluation	Saclay]	
(γ, xn)	3187	3200	3194]	
(γ, Sn)	2300 >	2383 <	2557	Up for 4 %!	Down for 9 %!
(γ, 1n)	1413	1642 <	1936	Up for 16 %!	Down for 19 %!
(γ, 2n)	887 -	< 714 >	605	Down for 24 %!	Up for 15 %!
(γ, 3n)	46	26	16	$\sigma^{int}(\gamma, 2n)/\sigma^{int}(\gamma, 1n)$	$\sigma^{int}(\gamma, 2n)/\sigma^{int}(\gamma, 1n)$
10/10/	/2014			decreased for 31 %.	increased for 27 %.

Disagreements - Tb







Independent test – activity method: identification of reaction using not outgoing neutrons but final nucleus









181**Ta**

Decays of ¹⁸¹Ta(γ , 1n) and ¹⁸¹Ta(γ , 2n) reactions final nucleus differ significantly:

¹⁸¹ Ta(γ , 1n) ¹⁸⁰ Ta, T _{1/2} =	8.154	hour,	E = 93.326	кэВ
			E = 103.557	кэВ
¹⁸¹ Ta(γ , 2n) ¹⁷⁹ Ta, $T_{1/2}$ =	1.82	year,	E = 63.0	кэВ

The comparison of ratios of reaction yields Y and integrated cross sections σ^{int} obtained for experimental and evaluated data for ¹⁸¹Ta for E^{int} = 65 MeV.

Ratios		Evaluation		
	Saclay	Livermore	Activity	F _{1,2,3}
of cross sections $\sigma(\gamma,2n)/\sigma(\gamma,n)$	0.36 (797/2190)	0.67 (887/1316)		0.49 (958/1956)
of yields Y(γ,2n)/Y(γ,n)	0.24	0.42	0.34 ± 0.07	0.33
of cross sections $\sigma(\gamma,3n)/\sigma(\gamma,n)$	0.063 (137/2190)			0.055 (107/1956)

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Comparison with data near threshold obtained using quasimonochromatic laser Compton-backscattering γ-rays








Possible reasons for clear systematic disagreements

The same neutron multiplicity sorting by neutron kinetic energy measurement was used in both Labs based on supposition that one neutron from (γ , 1n) reaction has energy larger than both neutrons from reaction (γ , 2n)

but experimental methods for neutron energy measurements were different:

- at Saclay the large Gd-loaded liquid scintillator was used ("suffered from a high background rate, made up largely of 1n-events, which introduced larger uncertainties in the background subtraction and pile-up corrections" – citation from B.L.Berman and S.C.Fultz, Rev.Mod.Phys., 47, 713 (1975));

- at Livermore so-called "ring-ratio" method was used (concentric rings of counters in paraffin moderator): low-energy neutrons (from reaction (γ , 2n)) should have enough time for moderation in the way to inner ring but high-energy neutrons (from reaction (γ , 1n)) should go to the outer ring passing inner ring (due to multiple scattering high energy-neutron could return to inner ring).







SUMMARY

Problems for EXFOR photonuclear data compilers:

• definitions of total reactions are not simple and depends on author's preferences;

• definitions of partial reactions can be complicated because presences of proton contributions into neutron reaction and vice versa of neutron contributions into proton reaction;

• there are many problems in definition of INC-SOURCE: using bremsstrahlung the energy of initial γ -quantum can be obtained accurate by complete kinematics (KINDT);

• there is crazy problem of compilation (and at the same time for evaluation) of data obtained using quasimonoenergetic annihilation photons: if Ye+ - Ye- = $Y \approx \sigma$, EN-RSL should be re-estimated; if Ye+ - Ye- = $Y \neq \sigma$, data should be recompiled,...

Problems for photonuclear data evaluators:

• shapes of cross sections Ye+ - Ye- = Y $\approx \sigma$ obtained using quasimonoenergetic annihilation photons are very doubtful because in reality those are the only reaction yields (energy resolution is much more worse than the width of annihilation line);

• shapes and values of partial reaction cross sections Ye+ - Ye- = $Y \approx \sigma$ obtained using quasimonoenergetic annihilation photons are very doubtful because those are not satisfy to physically objective criteria of data reliability.



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Thanks!

THANKS A LOT FOR ATTENTION!

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Many efforts:

E. Wolynec and M.N.Martins, Rev.Bras.Fis., 17, 56 (1987)

B.L. Berman et. al., Phys.Rev., C36, 1286 (1987)

V.V.Varlamov, et. al., INDC(CCP)-440, IAEA NDS, 37 (2004).

Contradictive recommendations: to multiply Livermore data, to divide Saclay data, to recalculate Saclay data for putting them into consistency with Livermore data.

With the aim to find objective criteria we investigated many sums, differences and ratios of various cross sections and at lately found out very simple, clear and physically objective criteria for data reliability – for presence (or absence) of systematic errors.











Analogous erroneous moving some number of neutrons from one decay channel to another.

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Systematics of (γ ,Sn) reaction cross section ratios "All/Livermore" for ~ 500 data sets (V.V.Varlamov, B.S.Ishkhanov. Study of Consistency Between (γ, xn) , $[(\gamma, 1n) + (\gamma, 1n1p)]$ and $(\gamma, 2n)$ Reaction Cross Sections Using Data Systematics. Vienna, Austria. INDC(CCP) - 433, 2002)



42 (γ, xn) systematics

12:







Model

B.S.Ishkhanov, V.N.Orlin. Physics of Particles and Nuclei, 38, 232 (2007), Physics of Atomic Nuclei, 71, 493 (2008):

semiclassical exciton preequilibrium model of photonuclear reaction based on the Fermi gas densities with taking into account effects of nucleus deformation and effects of Giant Dipole Resonance isospin splitting.

Model was tested on experimental data for neutron yield (γ, xn) reaction.

M.B. Chadwick et al., Phys. Rev. C 44, 814 (1991) – analogous model.



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Decay channels competition



Competition







The IAEA Co-ordinated Research Project on Compilation and Evaluation of Photonuclear Data for Applications.

"Handbook on photonuclear data for applications. Cross-sections and spectra" Final report of a co-ordinated research project 1996 – 1999. IAEA-TECDOC-1178, 2000.

IAEA Photonuclear Data Library (<u>https://www-nds.iaea.org/photonuclear/</u>).

Blokhin A.I., Nuclear Data Center, IPPE, Obninsk, Russia Chadwick M.B., Los Alamos National Laboratory, USA Fukahori T., Nuclear Data Center, JAERI, Japan Han Y., Nuclear Data Evaluation Laboratory, KAERI, Korea Lee Y.-O., Nuclear Data Evaluation Laboratory, KAERI, Korea Martins M.N., Instituto de Fizika, Universidade de Sao Paulo, Brazil Mughabhab S.F., Brookhaven National Laboratory, USA Oblozinsky P., IAEA Nuclear Data Section, Austria Varlamov V.V., Centre for Photonuclear Experiments Data, Moscow, Russia Yu B., China Nuclear Data Center, AEI, Beiging, China Zhang J., China Nuclear Data Center, AEI, Beiging, China







The IAEA Co-ordinated Research Project on Compilation and Evaluation of Photonuclear Data for Applications (1996 – 1999).

The major steps in an evaluations for 164 isotopes of 48 elements (from ²H to ²⁴¹Pu); consist of:

• Based on experimental information, evaluate the photoabsorption cross section (in many cases $\sigma(\gamma, Sn)$ were used instead of $\sigma(\gamma, abs)$), which is usually taken as an input to the subsequent nuclear reaction calculation.

• Calculate the $(\gamma, 1n)$, $(\gamma, 2n)$, $(\gamma, 1p)$ etc. excitation functions, and compare against available data. If different experiments are discrepant with one another, establish methods to assess which experiment is most likely to be accurate.

• If the calculated excitation functions disagree with measured values, consider studying the sensitivity of model calculations to some of input parameters.

• When an acceptable representation of measured data is obtained, use the model calculations to predict cross sections, and emission spectra.

Convert the calculated results into the ENDF-6 format.







The item

"in many cases $\sigma(\gamma, Sn)$ were used instead of $\sigma(\gamma, abs)$ "

can be the reason for serious loss of data reliability.

 $\begin{aligned} \sigma(\gamma, abs) &= \sigma(\gamma, 1n) + \sigma(\gamma, 1n1p) + \sigma(\gamma, 2n) + \sigma(\gamma, 2np) + \sigma(\gamma, 3n) + \ldots + \sigma(\gamma, F) + \\ \sigma(\gamma, 1p) + \sigma(\gamma, 1d) + \sigma(\gamma, 1d1p) + \ldots + \sigma(\gamma, 1\alpha) \approx \\ \sigma(\gamma, Sn) + \sigma(\gamma, charged particles) \end{aligned}$

Because $\sigma(\gamma, Sn) = \sigma(\gamma, xn) - \sigma(\gamma, 2n)$ large systematic errors in $\sigma(\gamma, 2n)$ lead to systematic errors in $\sigma(\gamma, Sn)$ and correspondingly to those in data for partial reaction cross sections evaluated on the base of using $\sigma(\gamma, Sn)$.





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Disagreements with Utsunomiya data obtained using quasimonochromatic laser Compton-backscattering γ-rays



Similar disagreements: experimental cross section for ⁹¹Zr (triangles) in comparison with our evaluation (line)

Current situation

with regards to photonuclear (photoneutron) reaction data obtained for energies of Giant Dipole Resonance (up to about 50 MeV)

- there are many experimental data obtained by various methods in various laboratories;
- the majority of partial photoneutron reactions (primarily (γ , 1n), (γ , 2n), (γ , 3n)) cross sections has been obtained at Livermore and Saclay using the method of neutron multiplicity sorting;
- generally there are enough small (~ 12 %) disagreements between neutron yield reaction (γ , xn) cross sections;
- in many cases there are significant (up to 100 %) disagreements between partial photoneutron reaction cross sections;
- those disagreements are clear systematic: as a rule (γ , 1n) reaction cross sections are larger at Saclay but (γ , 2n) reaction cross sections are larger at Livermore;
- in many cases both Saclay and Livermore data are not satisfied the new criteria of data reliability;
- in many cases both Saclay and Livermore data contradict with new data obtained using alternative methods without neutron multiplicity sorting;
- in many cases IAEA CRP evaluations agree with Saclay or Livermore data and disagree with alternative experiments data and evaluations based on new reliability criteria.

Shortcomings of CRP evaluations: needs to update existing databases (IAEA photonuclear data library

- In many cases $\sigma(\gamma, Sn)$ was used instead of $\sigma(\gamma, abs)$ systematic errors in $\sigma(\gamma, Sn)$ are 1. different for different nuclei;
- 2. In many cases evaluations have been done in order to model accurately Saclay data and are not satisfied new data reliability criteria;
- Many experimental data have been obtained after 2000 year using not only neutron 3. multiplicity sorting method;
- 4. Some new advanced theoretical models have been developed till now;
- 5. Evaluations have not been done (though experimental data exist in Berman's EXFOR library) for 37 isotopes for which data are needed not only for applications but for basic research (not only nuclear physics but nuclear astrophysics) also:

³H, ³He, ^{6,7}Li, ^{10,11}B, ¹⁴C, ¹⁹F, ⁴⁵Sc, ⁷⁵As, ^{76,78,80,82}Se, ⁸⁹Y, ¹⁰³Rh, ¹¹⁵In, ¹³⁸Ba, ¹³⁹La, ^{140,142}Ce, ^{142,143,144,145,146,148,150}Nd, ¹⁵³Eu, ¹⁶⁰Gd, ¹⁷⁵Lu, ^{186,188,189,190,192}Os, ²³⁷Np.

Some citations from IAEA CRP "Recommendations to Users and Evaluators" (IAEA-TECDOC-1178, 2000, page 60):

• "In cases where more than one evaluation exists... we recommend that users also study the sensitivity of their results to the use of other evaluations...".

• "The IAEA Photonuclear Data Libraries... have been produced through extensive recent research activities. While they have been generally tested against available... data, additional validation work is desirable... . Any discrepancies that are found, when related to the evaluators, may lead to further improvements in the evaluations".

• "We recommend that laboratories undertake new evaluations for cases where only one choice was available".

• "Additional experiments are needed to better understood photonuclear reaction physics. In particular, there still exists only few measurements of emission spectra of secondary particles from monochromatic photon-induced reactions."

Conclusions:

- The IAEA CRP (1996 1999) played important role in photonuclear reactions research and applications (systematics, many evaluations, digital data library, etc.);
- Evaluations carried out have several definite shortcomings (choice of data for modeling (primarily (γ ,abs) or (γ ,Sn) data of Saclay), disagreements with data reliability criteria, many omitted data);
- Many new data were obtained using various methods for period of time from 2000 year (using activity method yields of partial reactions were obtained up to 7 outgoing neutrons);
- Many advanced nuclear models were developed for that period of time;
- New Coordinated Research Project (or a smaller-scale Data Development Project) looks be useful for improving situation for photonuclear data;
- Similar to IAEA CRP (1996 1999) new Project should coordinate the efforts of experimentalists, theoreticians and evaluators;
- The first person for discussions about future certainly is Mark Chadwick (USA LANL).

The IAEA Co-ordinated Research Project on Compilation and Evaluation of Photonuclear Data for Applications (1996 – 1999):

- evaluations for 164 isotopes of 48 elements (from ²H to ²⁴¹Pu);
- using various nuclear modeling codes
 - GNASH (Los Alamos),
 - -ALICE-F and MCPHOTO (Tokai),
 - GUNF and GLUNF (Beijing),
 - XCFISS (Obninsk);
- using as initial experimental data for photoabsorption cross section $\sigma(\gamma, abs) = \sigma(\gamma, 1n) + \sigma(\gamma, 1n1p) + \sigma(\gamma, 2n) + \sigma(\gamma, 2np) + \sigma(\gamma, 3n) + ... + \sigma(\gamma, F) + \sigma(\gamma, 1p) + \sigma(\gamma, 1d) + \sigma(\gamma, 1d1p) + ... + \sigma(\gamma, 1\alpha) \approx \sigma(\gamma, Sn) + \sigma(\gamma, charged particles);$
- in many cases $\sigma(\gamma, Sn)$ were used instead of $\sigma(\gamma, abs)$;
- in many cases evaluations have been done in order to model accurately Saclay (γ , Sn) data.

Theory

Semiclassical exiton preequilibrium model of photonuclear reaction based on the Fermi gas densities and taking into account the effects of nucleus deformation and of GDR isospin splitting. Bohr description of $\sigma(y, lpkn)$:

$$\sigma(\gamma, lpkn; E_{\gamma}) = \sum_{i} \sigma_{\Gamma ДP}^{(i)}(E_{\gamma}) W_{\Gamma ДP}^{(i)}(l, k, E_{\gamma}) + \sigma_{K Д}(E_{\gamma}) W_{K Д}(l, k, E_{\gamma}),$$

 σ^{i} - one of 4 components (2 isospins - T_{0} and T_{0} + 1 and 2 directions of vibration), σ_{GDR} - Lorenz lines with $\Gamma^{\downarrow}_{pe3} \approx GI(a_{0}/R_{0})[E_{pe3} - \Delta(Z, N)\delta_{TT_{2}}]^{2}$,

where

$$I(\xi) = \left[1 - 3\xi(1 + \pi^2\xi^2/3)/(1 + \pi^2\xi^2)\right]/(1 + \pi^2\xi^2)$$

W - decay probabilities (recurrent):

$$\begin{split} W(l,k,E;dp,dn,m) &= \hbar \sum_{\substack{j=n,p \\ \Delta m'=2}} \sum_{\substack{m'=m \\ \Delta m'=2}}^{m-2} \frac{D(m',E;dp,dn,m)}{\Gamma^{\uparrow}(E;dp,dn,m') + \Gamma^{\downarrow}(E;dp,dn,m')} \\ &\times \int_{0}^{E-B_{j}} \lambda_{j}(\varepsilon_{j},E;dp,dn,m') W(l_{j},k_{j},U_{j};dp_{j},dn_{j},m') d\varepsilon_{j} + \\ &+ D(\bar{m},E;dp,dn,m) P(l,k,E;dp,dn), \end{split}$$

xn-sn-n-2n-3n

We need the objective criteria for data reliability

Main problem - criteria

¹⁵⁹Tb(γ,n)

Ratios	Experiments			Evaluation
	Saclay	Livermore	Activity	F _{1,2,3}
of cross sections $\sigma(\gamma,2n)/\sigma(\gamma,n)$	0.36 (797.4/2189.5)	0.67 (887.0/1315.7)		0.49 (958.3/1956.3)
of yields Y(γ,2n)/Y(γ,n)	0.24	0.42	0.34 ± 0.07	0.33 *)
of cross sections $\sigma(\gamma,3n)/\sigma(\gamma,n)$	0.063 (137.4/2189.5)			0.055 (107.3/1956.3)
of yields Y(γ,3n)/Y(γ,n)	0.02		0.023 - 0.025**)	0.018*)

Special investigation - E.Wolinec and M.N. Martin. Revista Brasiera de Fisica, 17, 56 (1987):

that was shown that results of measurements for ¹⁸¹Ta of $\sigma(e, xn)$ and $\sigma(e, 1n)$ reaction cross sections (measured directly by activation method for 98.3 keV γ -ray line from decay of final nucleus ¹⁸⁰Ta \rightarrow ¹⁸⁰Hf) recalculated using virtual photon spectra into correspondent $\sigma(\gamma, Sn)$ and $\sigma(\gamma, 1n)$ reaction cross sections lead to

agreement with Livermore data but not with Saclay ones.

1) So it was shown that the reason is incorrect procedure for neutron multiplicity sorting used at Saclay – $(\gamma, 2n)$ data are underestimated but $(\gamma, 1n)$ vise versa overestimated because of error in the neutron multiplicity sorting.

2) The neutron multiplicity sorting at Livermore is correct.

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F features

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 $(\gamma,\,1n)$ and $(\gamma,\,2n)$ reaction data

← before

and

🗲 after

joint correction procedure of recalculation from incorrect Saclay data to correct Livermore ones

Data for 19 nuclei (⁵¹V, ⁷⁵As, ⁸⁹Y, ⁹⁰Zr, ¹¹⁵In, ^{116,117,118,120,124}Sn, ¹²⁷I, ¹³³Cs, ¹⁵⁹Tb, ¹⁶⁵Ho, ¹⁸¹Ta, ¹⁹⁷Au, ²⁰⁸Pb, ²³²Th, ²³⁸U) were corrected (V.V.Varlamov et al. (J,YK,2003,(1-2),48,2003), put into EXFOR library – M0635 and added later by data for 4 actinides (²³²Th, ²³⁸U, ²³⁷Np, ²³⁹Pu) – M0722.

The problem seemed to be solved: it was recommended to use "good" Livermore data and do not use "bad" Saclay ones, **but...**

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Correction of both (γ , 1n) and (γ , 2n) reaction cross sections of Saclay

 $(\gamma,n) - (\gamma,2n)$ correction

Resolution 65

The simple method for Saclay data correction and putting those into consistency with Livermore data:

Total photoneutron reaction cross section in GDR energy region

 $(\gamma, \mathbf{xn}) = (\gamma, \mathbf{1n}) + 2(\gamma, \mathbf{2n}).$

Ratio R ("Saclay/Livermore" normalization) for all reactions cross sections

$$\mathbf{R} = \sigma_{S}^{n} / \sigma_{L}^{n} = \sigma_{S}^{n} / \sigma_{L}^{n} = \sigma_{S}^{2n} / \sigma_{L}^{2n} = (\sigma_{S}^{n} + 2\sigma_{S}^{2n}) / (\sigma_{L}^{n} + 2\sigma_{L}^{2n}),$$

$$\sigma_{S}^{n} = (\sigma_{S}^{n} + 2\sigma_{S}^{2n}) = \mathbf{R} \sigma_{L}^{n} = \mathbf{R} (\sigma_{L}^{n} + 2\sigma_{L}^{2n}).$$

Saclay corrected $\sigma_{S}^{2n}^{*}$ must be equal to Livermore corrected: $\sigma_{L}^{2n}^{*} = \mathbf{R}\sigma_{L}^{2n}$,

therefore: $\sigma_{L}^{2n} = \sigma_{S}^{2n} = R\sigma_{L}^{2n} = \sigma_{S}^{2n} + \frac{1}{2}(\sigma_{S}^{n} - R\sigma_{L}^{n})$.

Saclay (γ , 1n) reaction cross section part $\frac{1}{2}(\sigma^{n}S - R\sigma^{n}L)$ is "moved back" to Saclay (γ , 2n) reaction cross section $\sigma^{2n}S$.

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66 **"n-2n" correction formulae**

 S/S_1 are presented, where S were calculated for various laboratories data and S_1 - for Livermore QMA-data.

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Structurenes" S/S₁ ratios for (γ,xn) reaction cross section data

squares - BR-data (Moscow, Melbourne (Australia), other) - $\langle S/S_1 \rangle = 4.35$;

crosses - QMA-data (Saclay (France), Giessen (Germany), other) - $\langle S/S_I \rangle = 1.22$;

bows - Tagged Photons-data (Illinois (USA)) - $\langle S/S_1 \rangle = 4.22$

Structure systematic

Well-known data under discussion:

E.G.Fuller, H.Gerstenberg. Photonuclear Data - Abstracts Sheets 1955 - 1982. NBSIR 83-2742. U.S.A. National Bureau of Standards, 1986.

S.S.Dietrich, B.L.Berman. Atlas of Photoneutron Cross Sections Obtained with Monoenergetic Photons. Atomic Data and Nuclear Data Tables, 38 (1988) 199.

A.V.Varlamov, V.V.Varlamov, D.S.Rudenko, M.E.Stepanov. Atlas of Giant Dipole Resonances. Parameters and Graphs of Photonuclear Reaction Cross Sections. INDC(NDS)-394, IAEA NDS, Vienna, Austria, 1999.

V.V.Varlamov, V.V.Sapunenko, M.E.Stepanov. Photonuclear Data 1976 - 1995. Index. Moscow State University. Moscow, 1996 (bibliographic database URL (http://depni.sinp.msu.ru/cdfe/services/pnisearch.html).

International nuclear (including photonuclear) reaction data relational database (EXFOR):

I.N.Boboshin, V.V.Varlamov, E.M.Ivanov, S.V.Ivanov, N.N.Peskov, M.E.Stepanov, V.V.Chesnokov. Relational Nuclear Databases Upon the MSU INP CDFE Web-site and Nuclear Data Centres Network CDFE Activities. Report on the IAEA Consultant's Meeting on the Co-ordination of Nuclear Reaction Data Centres (Technical Aspects), 28 – 30 May 2001, Vienna, Austria. INDC(NDS)-427, IAEA NDS, Vienna, Austria, 2001, p. 49.

All data for quasimonoenergetic photons and many data for bremsstrahlung are included: URL (http://depni.sinp.msu.ru/cdfe/exfor/index.php):

EXFOR database

 $16O(\gamma,xn)$

In detailes: quasimonoenergetic data look like smoothed bremsstrahlung ones.

BR –QMA

Once more - ${}^{63}Cu(\gamma,n){}^{62}Cu$ reaction cross section in 3 steps QMA-experiment

- a) σ(k) ≈ Y(E_j) = Y_{e+}(E_j) Y_{e-}(E_j); must be additionally processed taking into account real apparatus function is needed ;
- b) Y_e(E_j) measured using electron bremsstrahlung must be processes by one of methods traditional for BRexperiments;
- c) $Y_{e+}(E_j)$ measured using photons from sum of positrons annihilation and bremsstrahlung must be processed also using appropriate apparatus function. ${}^{63}Cu(\gamma,n){}^{62}Cu$

Disagreements (Saclay/Livermore)

of amplitudes – absolute values – integrated cross sections

5 clear cases (from "Atlas..." of S.S.Dietrich and B.L.Berman, Atomic Data and Nuclear Data Tables, 38 (1988) 199) of σ^{int} disagreements for appropriate integration energy limits E_{γ}^{max} :

Nucleus	⁵¹ V	⁷⁵ As	⁹⁰ Zr	¹³³ Cs	¹⁶⁵ Ho
$E_{\gamma}^{\text{int-max}}$	27.8	26.2	25.9	24.2	26.8
(MeV)	27.8	29.5	27.6	29.5	28.9
$\sigma^{\text{int}} s / \sigma^{\text{int.}}_{L}$	689/654 = 1.06	1306/1130 ≥ 1.16	1309/1158 ≥ 1.13	2484/2505 ≈ <i>1</i>	3667/3385 ≥ 1.08

The values obtained at Saclay are higher than that obtained at Livermore for about 6 - 16 %.

Explanation of the reasons (B.L.Berman, et al., Phys.Rev., C36 (1987) 1286): "... an Livermore experiments error either in the photon flux determination or in the neutron detection efficiency or in both".

5 nuclei "S/L"






Significant disagreements for partial reaction (γ,n) and $(\gamma,2n)$ cross section between Saclay and Livermore data (integrated cross section ratios are presented).

	n	2 <i>n</i>	xn			
Nucleus	$\sigma^{int}{}_{s}(\gamma,n)/$	$\sigma^{int}{}_{s}(\gamma,2n)/$	$R^{int}(\gamma,xn)$	$\sigma^{int}_{S}(\gamma,n)/$	$\sigma^{int}{}_{s}(\gamma,2n)/$	$R^{int}(\gamma,xn)$
	$\sigma^{\text{int}}_{L}(\gamma,n),$	$\sigma^{\text{int}}_{S}(\gamma,2n),$	/25/	$\sigma^{\text{int}}_{L}(\gamma,n),$	$\sigma^{\text{int}}_{S}(\gamma,2n),$	/26/
	/1, 25/	/1, 25/		/26/	/26/	
	(= arb. units)	(= arb. units)	(arb. units)	(arb. units)	(arb. units)	(arb. units)
⁵¹ V				1.07	0.79	1.07
⁷⁵ As				1.21	1.22	1.21
⁸⁹ Y	1279/960 = 1.33	74/99 = 0.75	1.26	1.25	0.87	1.25
⁹⁰ Zr				1.26	0.73	1.26
¹¹⁵ In	1470/1354 = 1.09	278/508 = 0.55	0.94	0.97	0.76	0.97
¹¹⁶ Sn				1.10	0.92	1.10
¹¹⁷ Sn	1334/1380 = 0.97	220/476 = 0.46	1.01	1.02	0.93	1.02
¹¹⁸ Sn	1377/1302 = 1.06	258/531 = 0.59	1.06	1.07	0.86	1.07
¹²⁰ Sn	1371/1389= 0.98	399/673 = 0.75	0.99	1.00	0.86	1.00
¹²⁴ Sn	1056/1285 = 0.82	502/670 = 0.75	0.93	0.93	0.94	0.93
¹²⁷ I				1.34	1.07	1.34
^{133}Cs	1828/1475 = 1.24	328/503 = 0.65	1.11	1.10	0.88	1.10
¹⁵⁹ Tb	1936/1413 = 1.37	605/887 = 0.68	1.06	1.07	0.71	1.07
¹⁶⁵ Ho	2090/1735 = 1.20	766/744 = 1.03	1.14	1.20	1.05	1.20
¹⁸¹ Ta	2180/1300 = 1.68	790/881 = 0.90	1.22	1.25	0.89	1.25
¹⁹⁷ Au	2588/2190 = 1.18	479/777 = 0.62	1.00	1.00	0.69	1.00
²⁰⁸ Pb	2731/1776 = 1.54	328/860 = 0.38	1.30	1.21	0.77	1.21
²³² Th				0.84	0.69	0.84
²³⁸ U				0.76	0.79	0.76
	more higher	more lover <	< K>≈1.1 2			

While (γ,n) Saclay data are more higher than those from Livermore,

(γ,2n) data are, vise versa, more lower.

> "(γ ,n) – (γ ,2n)" discrepancies







Important results:

- clear data discrepancies force one to use data existed strongly individually;
- quasimonoenergetic photons-data are strongly (3 4 times) over-smoothed and must be additionally reprocessed to take into account real shape of apparatus function (effective photon spectrum);
- Livermore total photoneutron reaction (γ ,xn) cross sections have in general absolute values smaller then that obtained at various other laboratories; the reason: "... an Livermore experiments error either in the photon flux determination or in the neutron detection efficiency or in both"; therefore Livermore (γ ,xn) cross sections data of for 19 nuclei studied specially must be multiplied by appropriate coefficients $R^{int}(\gamma,xn)$ and for others – by $\langle R^{int}_{syst} \rangle = 1.12$ at least;
- Saclay partial photoneutron reactions (γ,n) and $(\gamma,2n)$ cross sections are not correct and consistent each other because of incorrect neutron multiplicity sorting procedure used and must be recalculated;
- Livermore neutron multiplicity sorting procedure at the same time is correct and therefore Livermore (γ,n) and $(\gamma,2n)$ cross sections are in consistence with each other and with (γ,xn) cross sections and both can be used but again only multiplied by coefficients $R^{int}(\gamma,xn)$ or $\langle R^{int}_{syst} \rangle$.

Important results

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3 important physical consequences:

- **GDR structure** (resonances with width ~ hundreds of keV) exists; BR-data look like preferable for GDR structure detailed study because QMA-data are strongly over-smoothed;
- E1 GDR decays dominantly statistically Saclay interpretation of high-energy tails of (γ,n) reaction cross sections as contributions of high-energy neutrons from GDR nonstatictical direct decay (those contributions evaluated to be about 17 - 30 %) because of small decreasing of (γ,n) reaction cross sections for energies higher than $(\gamma,2n)$ reaction threshold B(2n) looks like as very doubtful; Saclay (γ,n) data corrections described decrease those and put them into accordance with Livermore data: direct decay contributions are not more than 10 - 12 %;
- big extra integrated cross section $\sigma^{int}(\gamma, abs) \approx 1.3 1.5$ 60NZ/A (MeV•mb) became doubtfully being all due to effective mass of nucleon changing because of the effect of exchange forces; Saclay data correction described affects photoabsorption cross section evaluation using cross section data combinations (γ, abs) = $(\gamma, sn) + (\gamma, p)$ and $(\gamma, sn) = (\gamma, xn) - (\gamma, 2n)$; mistake in $(\gamma, 2n)$ reaction data produces the mistakes in both (γ, sn) and (γ, abs) reaction data; correction described do them noticeably smaller.

Physical consequences

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Neutron multiplicity sorting procedure test:

Twice measurement of 181 Ta(e,2n) 180 Ta cross section s(e,2n) = ${}^{1/2}$ (s(e,xn) - s(e,n)):

- 1. $\sigma_1(e,n)$ neutron multiplicity sorting measurement;
- σ₂(e,n) measurement of induced activity (decay ¹⁸⁰Ta → ¹⁸⁰Hf , 93.3 keV, Ge-Li).
 Mean-square ratio <σ₁(e,n)/σ₂(e,n)> = 1.057 ± 0.023 means high reliability of multiplicity sorting procedure.

Comparison of (e,n) and (γ ,n) data show that Saclay data for (γ ,2n) reaction are underestimated and correspondingly that for (γ ,n) reaction – vise versa overestimated.







Proposals for possible Coordinated Research Project (or a smaller-scale Data Development Project):

review the current situation with regards to gammaray data (starting from the GDR and extending to lower excitation energies down to 2-3 MeV)

- review available compilations

- discuss needs and ways to update existing databases (IAEA photonuclear data library)

- discuss needs for evaluation of existing data
- discuss/assess various theoretical approaches
- discuss need to coordinate efforts to perform new measurements

- discuss need to coordinate efforts to set up a dedicated database of compiled and evaluated gamma-ray data

- in case a CRP is advised, define the work to be carried out in the course of the project