MASS AND CHARGE DISTRIBUTIONS FROM PHOTON-INDUCED FISSION: COMPARISON WITH EXPERIMENTAL DATA AND YIELDS FROM NEUTRON INDUCED-FISSION

J.-C. David[1i], M.-L. Giacri-Mauborgne[1], D. Ridikas[1]

[1] DSM/DAPNIA/SPhN, CEA/Saclay, F-91191 Gif-sur-Yvette cedex, France

Introduction

When fission reactions take place in a material, the total activation can be estimated only if the mass and charge distributions of the fission products are known. Moreover, some of these produced nuclei will decay by delayed neutron emission, and a good knowledge of their characteristics might be very important for practical purposes. For actinides these distributions are well known for neutron-induced fission, what is not the case for photon-induced fission. We note that most frequently those photons are produced as bremsstrahlung radiation by electrons, and that, for example, one third of all existing accelerators in the world are electron linacs used in radiotherapy (that is about 5000) [1]. Therefore, a good knowledge of photofission yields becomes an important requirement for the operation of such installations. Many other reasons urge us to study photofission. The photofission process can be used to produce radioactive beams (ALTO project in Orsay-France) or neutrons (Hybrid reactors), to identify nuclear material (non-proliferation project) or characterize nuclear waste by a non-destructive way. Finally it could be also employed for the nuclear waste transmutation (with very high intensity electron beams).

Which code for which observables?

The knowledge of the γ-fission means the knowledge of several observables. The first one is, of course, the photon absorption cross section (σ_{abs}), the only input channel observable. For the output channel much more observables are available to characterize the reaction. The particle emission cross section ($\sigma(\gamma,xn)$, $\sigma(\gamma,xp)$) and the fission cross section tell us, quantitatively, how the nucleus deexcites. The fission yields give information about the charge and mass distribution of the fission products. Most of these fission products are radioactive, thus during that decay process the delayed neutrons, delayed photons and the activation products are the last observables of the photofission reaction. All these observables depend on the γ-energy and on the target nucleus.

At Los Alamos (LANL) the CINDER'90 [2] activation code was developed to obtain the activation products and associated delayed neutrons, created by neutron-induced fissions. For different reasons (the homeland security project, for example) the same code will be extended also for photon-induced reactions, including photofission. However, the data required has to be completed. These data are the cross sections (absorption, particle emission and fission), and the fission yields. Some data come from the IAEA data base, but are not sufficient (about 160 nuclei are available and more than 600 nuclei are needed). In addition the IAEA data base does not provide the fission yields. The GNASH code [3] could complete the data base, but it was dedicated to neutron reactions, and it would be very hard and time consuming work to use it for photon reactions. Another code, HMS-ALICE [4], is easier to use and is a good candidate to provide the required CINDER'90 inputs, but, up to now, the fission yields are not available.

So, we decided at CEA-Saclay to use and test a code from GSI which is known to give good results for spallation process, where a nucleus is also excited and deexcite via evaporation and/or fission. This code gives us all observables, cross section and fission yields. In fact, we use two codes from GSI, since we split the modelization in two main parts: the input channel with the γ absorption and nucleus excitation, and the output channel, that is the nucleus deexcitation. The γ excitation of the nucleus is based on the giant dipole resonance principally, but also on the giant quadrupole resonances. The absorption cross section is the sum of these components, each of them determined from empirical systematics [5]. The nucleus deexcitation is performed with the ABLA code [6] based on a statistical model, where fission is in competition with particle emission. In other words, the complete code provides neutron (proton) emission and fission cross sections, and also fission yields. Multi-chance fissions are taken into account as well.

Results

To check the validity of the GSI codes and to try to find a possible link between γ-fission and n-fission, we compare our theoretical results to available data. These data are the cross sections (absorption, particle evaporation and fission), the fission yields, the delayed neutrons and isotopic distributions (cumulative ones). We will focus on Uranium and Plutonium, since they are the nuclei experimentally investigated most of the time.

Cross sections

In figures 1 to 3 we compare our predictions with the IAEA data for different type of cross sections in the case of ^{235}U (fig.1), ^{238}U (fig.2) and ^{239}Pu (fig.3). The blue curves, called *GSInew*, require some explanations. The difference between *GSI* and *GSInew* is in the giant dipole resonance cross section parameterization. In *GSInew* the dependence on the deformation parameter has been changed according to Peter Möller systematics [7] and an effort has been made to reproduce $\sigma_{abs}(E_\gamma)$ for ^{238}U.

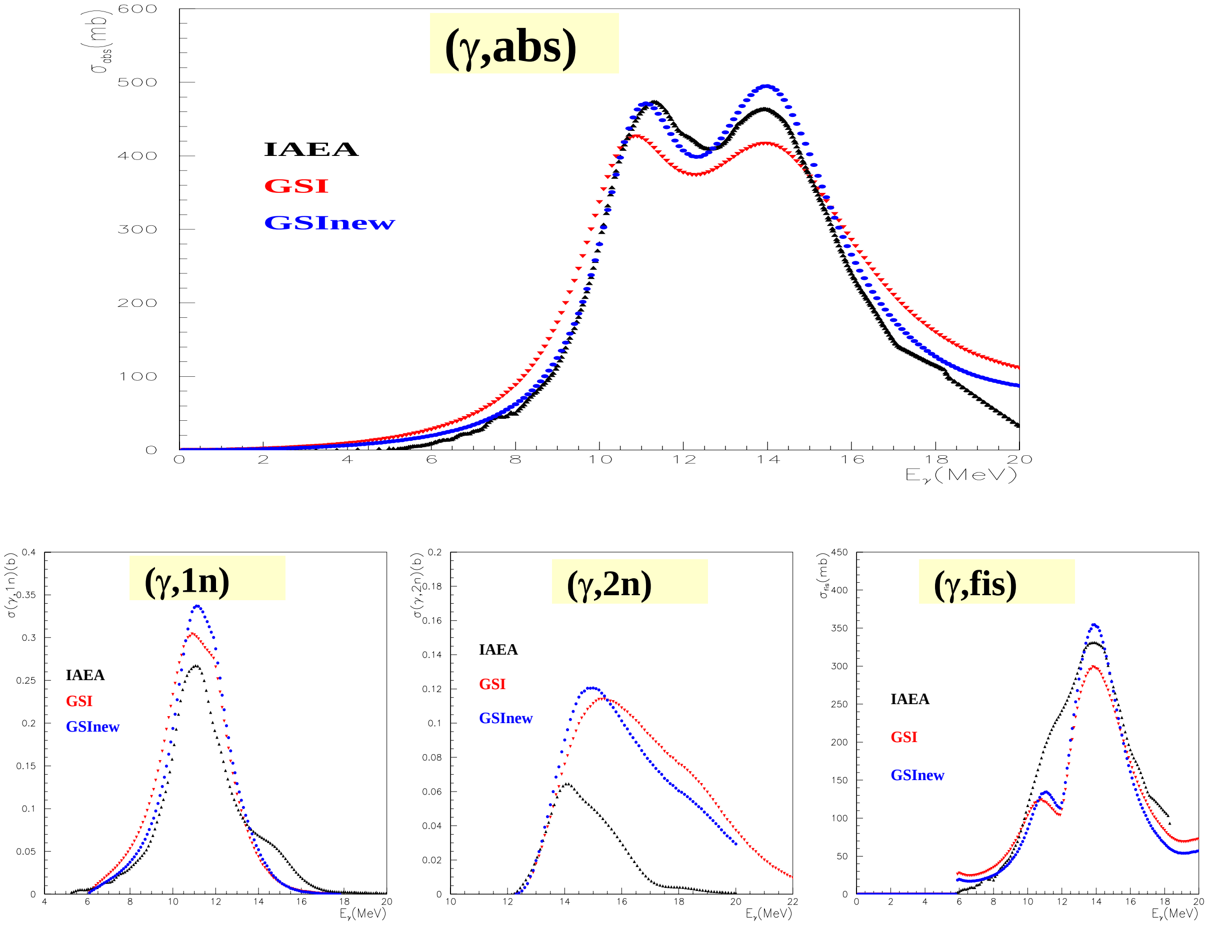

Figure 1: Cross sections for the reaction γ + ^{235}U.

Figure 1 is dedicated to ^{235}U. The results are good for the $\sigma_{abs}(E_\gamma)$ with good shapes and the right absolute values. For the $\sigma(\gamma,1n)$, $\sigma(\gamma,2n)$ and σ_{fis}, the shapes are roughly good except for σ_{fis} around 12 MeV, and $\sigma(\gamma,xn)$ beyond 14 MeV, and the absolute values are often too high for particle emission, and especially for $\sigma(\gamma,2n)$. The overestimation between 10 and 13 MeV of neutron evaporation leads to an underestimation of the fission cross section, since $\sigma_{abs}(E_\gamma)$, well reproduced, can be considered as the sum of $\sigma(\gamma,1n)$, $\sigma(\gamma,2n)$ and σ_{fis} in this case.

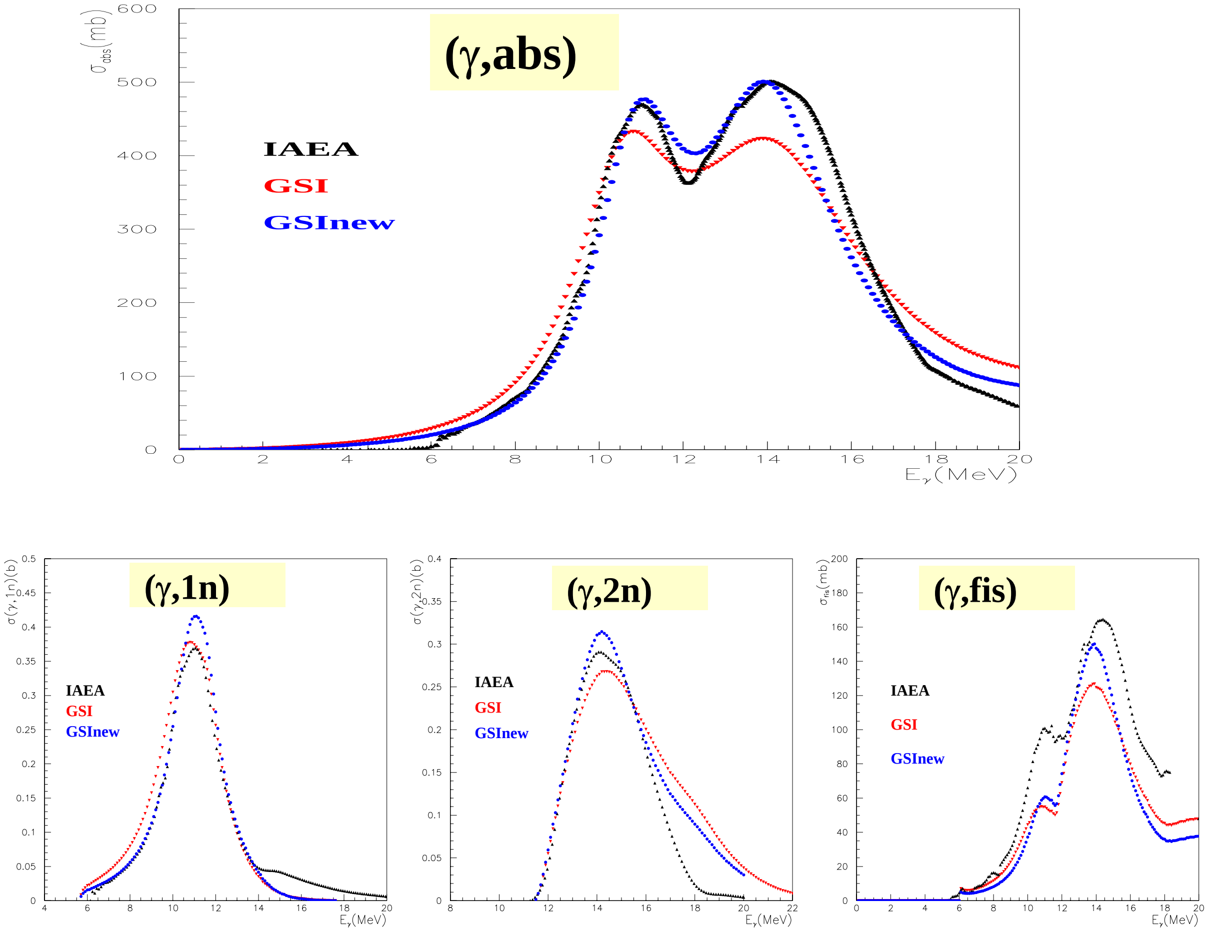

Figure 2: Cross sections for the reaction γ + ^{238}U.

For ^{238}U, figure 2, the conclusions are the same: quite good results, but the competition between evaporation/fission is not always in right proportions.

Unfortunately, the cross sections comparisons for ^{239}Pu, figure 3, are not so good as the previous ones for Uranium. Here the competition between neutron evaporation and fission is even worse, with too little evaporation, and too strong fission contribution. On the other hand the fission cross section shapes are not too bad (compared to the absolute values) except the valley between 11 and 14 MeV, for the σ_{abs}, being too deep.

These preliminary results on the cross sections show, first, that although σ_{abs} are quite good, some improvement might be needed (see ^{239}Pu), and secondly, we have to take care on the evaporation/fission competition. The ABLA code was developed for spallation process where the excitation energies are higher than γ energies considered in this work. So some effects, important at low energy, are perhaps not included in the model. Nevertheless, possible effects, which can be taken into account in the model, still need to be examined. Actually, we used for this study the default values for the input parameters, except the nuclear reduced friction coefficient which is supposed to be 2.10^{21} s^{-1} for the spallation process, and which is equal to 0 here for γ-fission due to the low energies. This value, 0, give us better results for σ_{fis} than the default value, and is usually used for low excitation energies.

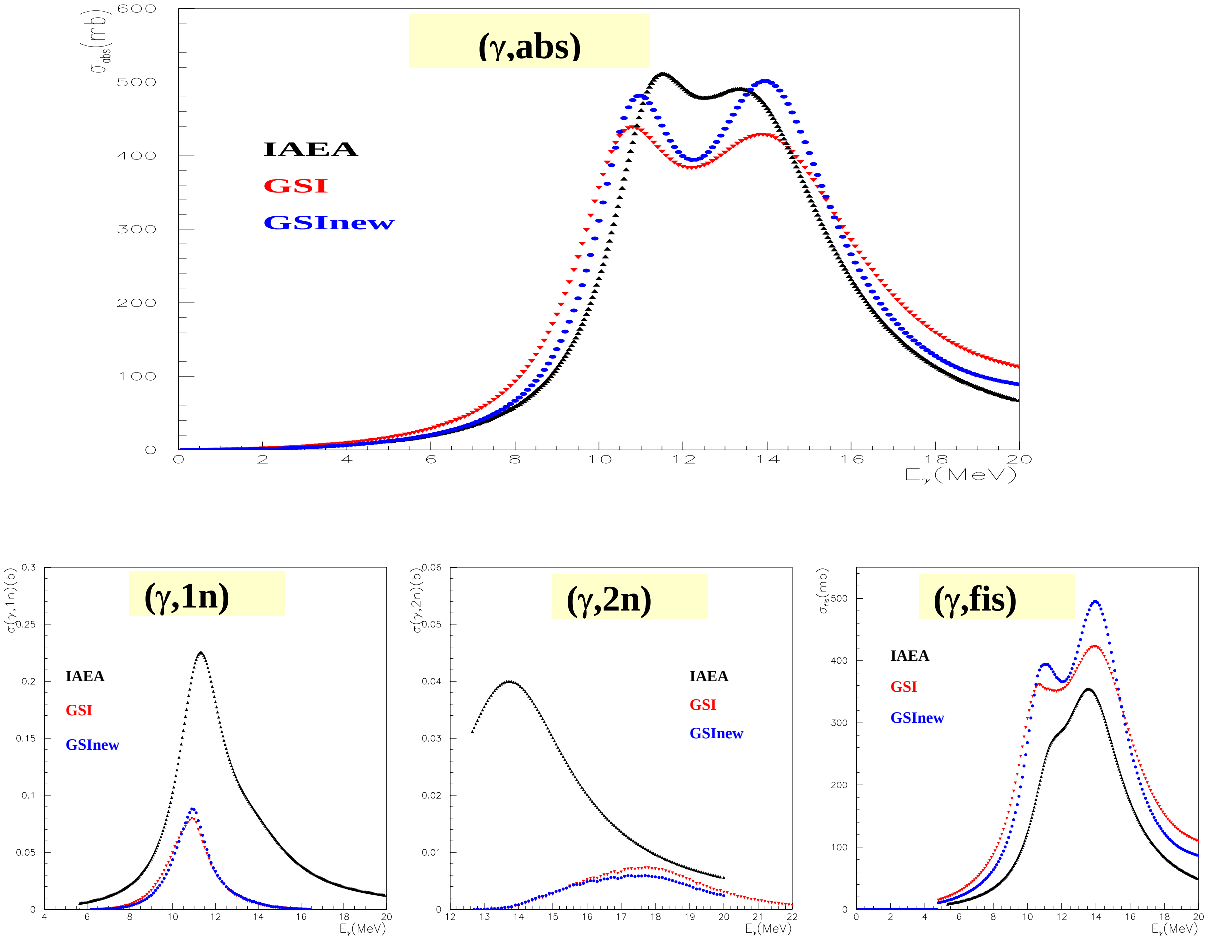

Figure 3: Cross sections for the reaction γ + ^{239}Pu.

Fission yields

Figures 4 and 5 show results for mass and charge fission yields for Uranium. Figure 6 shows results for a number of isotopic fission yields, because one point corresponds to a defined mass and charge.

For Uranium, we compare our predictions to 25 MeV bremsstrahlung data. So we used for our calculations a bremsstrahlung spectrum to be able to compare exactly the same observables. We add results of three monoenergetic photon calculations, 10-15-20 MeV, for the mass distribution (on the left). The idea was to know if we could reproduce the bremsstrahlung data by monoenergetic photons, and if yes, at which energies. It seems it is possible here with 15 MeV photons. The most interesting result is that our calculations (green line) reproduce rather well the data (red points). The only slight differences are in the valley. For the charge distributions (on the right), the data are scarce, but the agreement is still good.

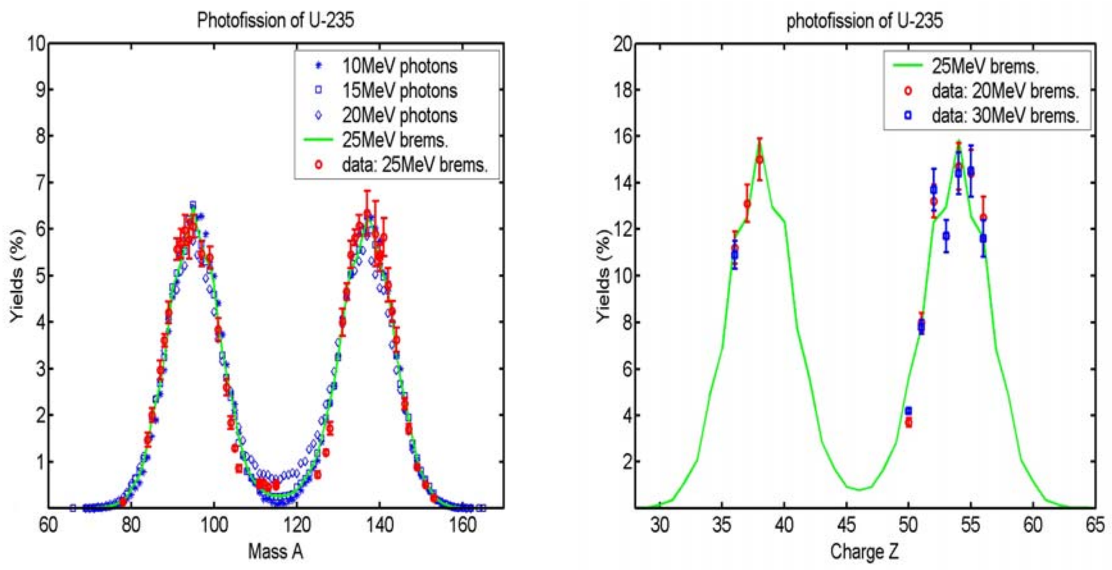

Figure 4: Fission yields for the reaction $\gamma_{\text{bremsstrahlung}}$ + ^{235}U. Data for 25 MeV bremsstrahlung (left side) come from [8], and for 20 and 30 MeV bremsstrahlung (right side) from [9].

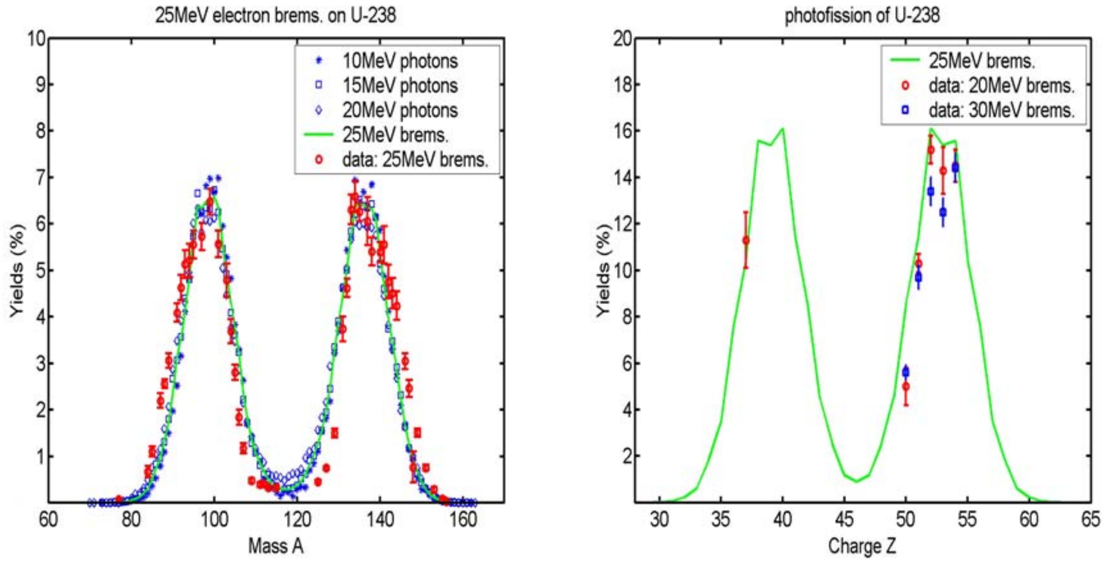

Figure 5: Same as fig.4, but for $\gamma_{\text{bremsstrahlung}}$ + ^{238}U.

The yields in figure 6 for ^{239}Pu are also good. Here the yields are cumulative and we have had to use the CINDER'90 evolution code after GSI codes to obtain them. So, we conclude that the fission yields in term of mass distributions are well reproduced by the ABLA code.

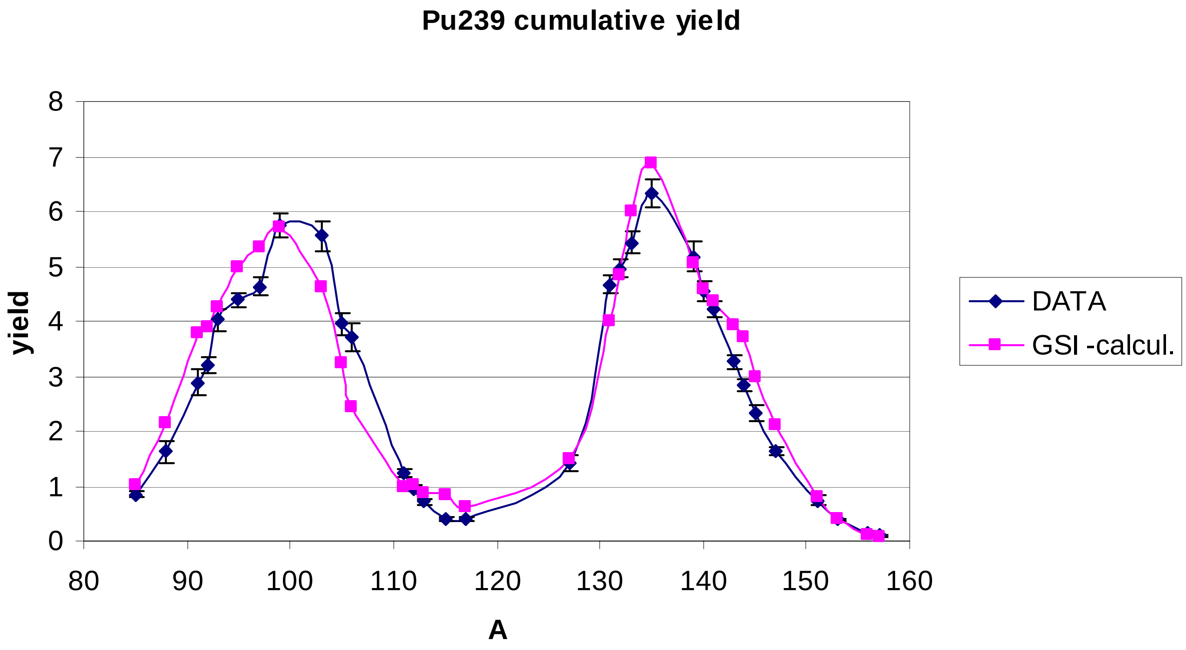

Figure 6: Cumulative fission yields for the reaction $\gamma_{\text{bremsstrahlung}}$ + ^{239}Pu. Data are drawn from [10].

Now with the figure 7 we tried to evaluate if the fission yields obtained with a 25 MeV bremsstrahlung spectrum (equivalent to 15 MeV photons) could have similarities with fission yields obtained with neutrons. We plot the bremsstrahlung data points for ^{235}U and the data for neutron-induced fission on ^{234}U and ^{235}U with thermal, fast neutrons (≈1MeV) and 14 MeV neutrons. We can conclude that a possible link exist with neutrons around 7 MeV, but this result has to be confirmed by other ways.

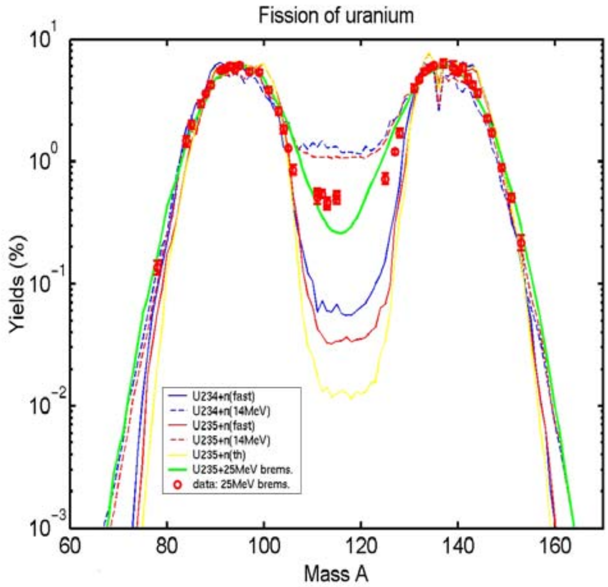

Figure 7: Fission yields comparisons between photofission and neutron-induced fission. Data red points: see figure 4. Data for neutron reaction come from Reference [11].

Delayed neutrons

The figure 8 plots the time behaviour of the delayed neutrons. Our calculation is carried out with a 25 MeV bremsstrahlung spectrum, and the data we have found deals also with bremsstrahlung spectra, but for electron energies of 8, 10 or 15 MeV. Nevertheless, the comparisons are quite good for ^{235}U. Similar results are also obtained for ^{238}U (not shown).

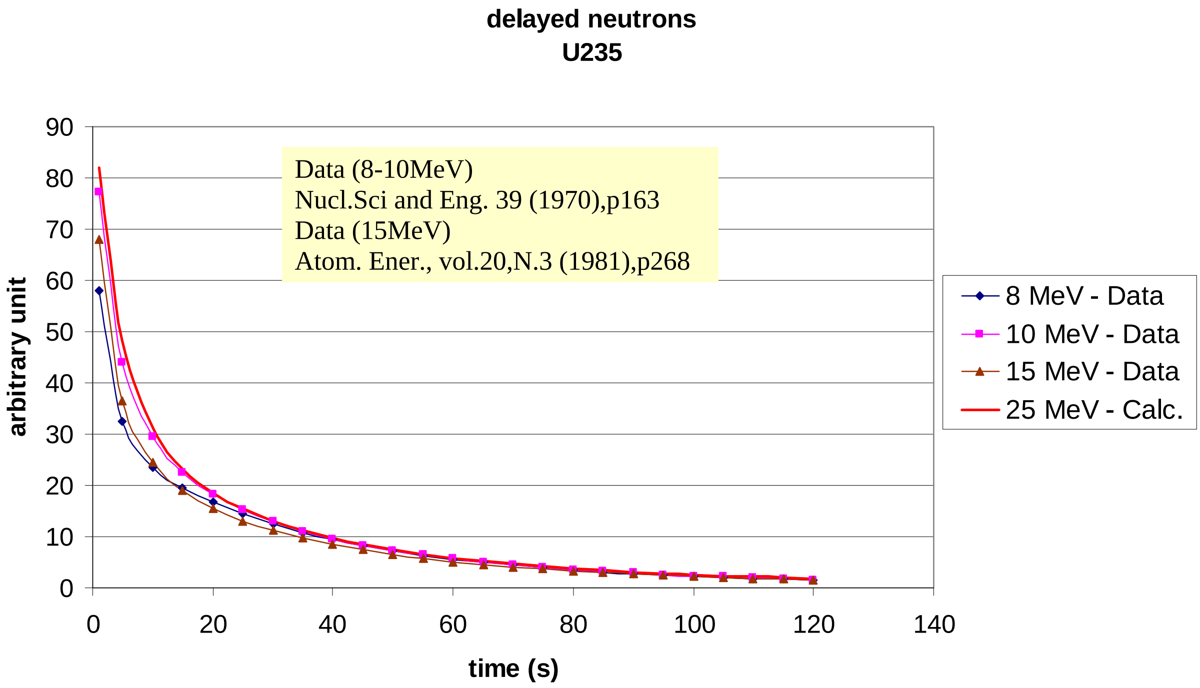

Figure 8: Time behaviour of the delayed neutrons for the reaction $\gamma_{bremsstrahlung}$ + ^{235}U.

Group	Half-life (s)	**e^-(25MeV) + ^{235}U (calculation)**	**e^-(15MeV) + ^{235}U (data)**	**n(fast) + ^{234}U (data)**	**n(14MeV) + ^{234}U (data)**
1	55.60	**0.063**	**0.054**	**0.052**	**0.050**
2	20.00	**0.237**	**0.200**	**0.256**	**0.151**
3	5.45	**0.294**	**0.152**	**0.213**	**0.137**
4	2.00	**0.358**	**0.369**	**0.350**	**0.281**
5	0.50	**0.109**	**0.139**	**0.057**	**0.046**
6	0.20	**0.009**	**0.086**	**0.009**	**0.009**
All		**1.070**	**1.000**	**0.937**	**0.674**

Table 1: Delayed neutrons yields for each group.

In the table 1, we show the yields of delayed neutrons for each group. Our calculation is done for ^{235}U and 25 MeV bremsstrahlung spectrum, and the data deal with 15 MeV bremsstrahlung spectrum, neutrons on ^{234}U, at ≈1 MeV and 14 MeV. The comparison shows that γ-fission results are not very far from neutron fission data. But the link we wished to establish between γ and neutron fission is not so clear as we could think from the very beginning (also see Fig. 7). Since most of the time the neutron-induced fission produces less delayed neutrons, except for some rare groups, the hypothesis which said a 7 MeV neutron-induced fission could be regarded as a 15 MeV photon-induced fission (roughly equivalent to a 25 MeV bremsstrahlung spectrum) seems to be wrong for the delayed neutrons.

Isotopic distributions

Finally we show on figure 9 an isotopic distribution of ^{39}Y. Our calculation and the data are again for the same reaction (see figure 8). Here we also added a calculation for a 14 MeV neutron-induced fission with the ABLA code (pink line). The differences between the theoretical results and the data can maybe explain the difficulties to draw a link between neutron and γ fission from the delayed neutrons, since these delayed neutrons come from the fission products.

The figure shows also that the ABLA code is consistent, because the results for γ and neutron-induced fission are similar. But we note also that, as we had concluded for the cross sections, a better use of the ABLA code, and perhaps its validity for the low energies, has to be investigated: the data for n(14MeV)+^{234}U are not well reproduced (theoretical results are translated by one mass unit compared to the data).

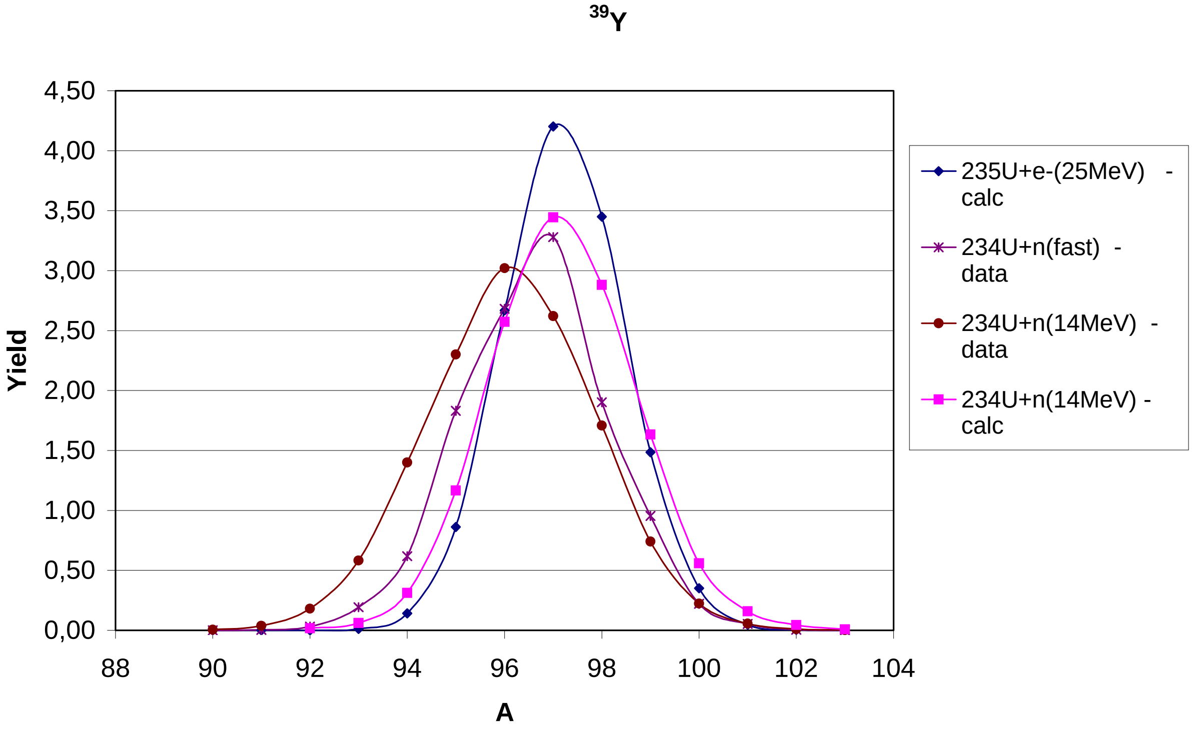

Figure 9: Isotopic distribution for the ^{39}Y element in the case of n+^{234}U and γ + ^{235}U.

Conclusions

A number of γ-induced reaction channels have been systematically investigated.

The photon-induced absorption cross sections are quite well reproduced by the GSI code based on giant dipole and quadrupole resonances. But there are some discrepancies for the particle emission and fission cross sections. These discrepancies depend on the incident photon energy and on the target nucleus, and the reason being the difficulty to get the right competition between evaporation and fission. Improvements can be done, which could also lead to get, perhaps, better results for isotopic distributions.

The fission yields obtained with ABLA are rather good. This is the strong point of this code for low energies. The delayed neutrons are roughly well reproduced, but an improvement of the evaporation/fission competition will be interesting for this observable as well.

We expected to show a link between neutron and photon-induced fission, but from this preliminary study no clear conclusions can be drawn. An exhaustive investigation will still have to be made.

Finally we would like to stress that the GSI model for photofission seems to be very encouraging.

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[i] jcdavid@cea.fr