# MEASUREMENTS AND ANALYSES OF THE PHOTONUCLEAR REACTION YIELDS

S. A. KARAMIAN

*FLNR, Joint Institute for Nuclear Research, Dubna, 141980, Russia*

Reactions induced by photons may be used as a probe for bonds and correlations between nucleons inside a heavy target nucleus. Electromagnetic radiation perturbs the nucleons only slightly unlike the influence of strongly interacting particles. The yield of (γ, α) reactions could be used to test theoretical models assuming a complete α-clustering, or multi-quark objects in heavy nuclei. Relative yields of (γ, *n*), (γ, *p*), and (γ, α) reactions have been measured at the bremsstrahlung end-point energy of 23 MeV with several targets. Much lower probability of (γ, α) compared to (γ, *p*) reactions is proved despite practically similar threshold and spin factors for both types of reactions. Alpha clustering in heavy targets is not supported. The mechanism of particle release in nuclear reactions is discussed and some details are clarified.

## 1. Introduction

The status of nucleons within a nuclear matter isn't yet well clarified point despite great interest since the decades. Simplest assumption that nucleons conserve their individual properties, same as in vacuum, was under criticism from different points of view. There is known a spectrum of ideas: on necessity to replace the nucleons by quasi-particles, on interacting bosons inside the nucleus and on complete alpha-clustering in nuclear matter. Over recent decade, an idea of short range nucleon-nucleon correlations with formation of a quark bag attracts an attention, and it is tested in reactions at GeV energies [1]. The mentioned above models also find some application and reach a success in simulation of different processes. The additional tests in experiments are yet relevant.

We propose now to use the reactions induced by bremsstrahlung at moderate end-point energy of about (20 – 30) MeV. At higher energies, the consequent emission of nucleons becomes probable, or even dominates, and that makes difficult to deduce the regularities in the yield of different-type reactions.

Recently, there are presented [2] the evidences for regular threshold dependence of the yields of photon-induced reactions. Relative yields were systematized versus the ($E_e$ – $E_{th}$ – $B_c$) parameter containing an excess of the end-point energy $E_e$ above the sum of reaction threshold $E_{th}$ and Coulomb barrier $B_c$ for particle emission. Emission of α-particles [3 – 7] is now under the scope. The $B_c$ values are calculated using widely used expression given in [8]. The different-reaction yields are normalized to the yield of the most abundant (γ, *n*) reaction and they show a regular growth with the increase of the mentioned threshold-excess parameter at moderate end-point energy $E_e \leq 30$ MeV.

For (γ, α) reactions, the systematic data collection is not available at the domain of heavy and medium weight targets. Existing results of some experiments [3–7] unfortunately demonstrate the scattering of the yield values by several orders of magnitude. Therefore, experimental studies of the (γ, α) reactions must be productive for understanding of the nuclear processes mechanism. In particular, a comparison of the nucleon and alpha emission rates could clarify the questionable point about nucleon-nucleon correlation status. At least, an idea of complete alpha clustering in heavy nuclei must be probed.

## 2. Experimental possibilities and successive measurements

Shortcomings in the (γ, α) reaction yields could be covered with regular measurements using the reliable activation technique. The relatively low yield of the (γ, α) reaction in combination with a great background due to the most abundant reactions: (γ, *n*), (γ, γ’) and (γ, *p*), make the experiment moderately complicated. The following requirements must be satisfied: the target species available in a form of the enriched isotopes, the convenient properties for detection of the product activity and reasonably soft background restrictions. Inspecting the Nuclide Chart from A =100 to 208 we have found the most promising cases for successful detection of the (γ, α) product by activation method with γ-spectroscopy measurements of the induced activity. The tabular data were taken into account by Nucl. Data Sheets and Table of Isotopes, and the most favorable cases are distinguished. They are listed and characterized in

Table 1. The mass numbers of potential targets correspond to the domain of heavy nuclides but out of the alpha-radioactive nuclide range. The activities listed in Table 1 are chosen because they are characterized by the relatively intense $\gamma$-lines convenient for detection at moderate halflife.

The major problem of such experiments would be a presence of the background radiation generated by the isobaric nuclides due to the same transition at the same daughter nuclide but after $\varepsilon$- instead of $\beta^-$- decay. This internal physical background couldn't be excluded by a better shielding of the detector, or so. Fortunately, in several cases, the background is absent and they must be considered as the best for reliable detection of the $(\gamma, \alpha)$ reaction and for the yield measurements. For some other targets, the experimental conditions could be improved by using of the enriched isotopes because the background is created typically by reactions with auxiliary isotopes in a target. In our experiment, the $(\gamma, \alpha)$ yields were successfully measured in four cases and three more are resulted in the upper limits for the yield. The targets in majority were taken in a form of metal foils of natural isotopic composition, except the $^{176}$Yb 95% enriched ytterbium oxide powder packed in the envelope made of Al foil. Full weight of the target materials was typically about (0.2 - 0.5) g.

Table 1. Encounter data for the $(\gamma, \alpha)$ experiment

| Target | Abundance, % | Product | Halflife | $E_\gamma$, keV | Background |
|---|---|---|---|---|---|
| $^{109}$Ag | 48.2 | $^{105}$Rh | 35.4 h | 319 | $^{105}$Ag from $(\gamma, 2n)$ |
| $^{113}$Cd | 12.2 | $^{109}$Pd | 13.7 h | 88 | $^{109}$Cd from $(\gamma, n)$ |
| $^{115}$In | 95.7 | $^{111}$Ag | 7.45 d | 342 | – |
| $^{119}$Sn | 8.6 | $^{115}$Cd | 53.4 h | 528 | – |
| $^{137}$Ba | 11.2 | $^{133}$Xe | 5.25 d | 81 | $^{133m}$Ba from $(\gamma, n)$ |
| $^{143}$Nd | 12.2 | $^{139}$Ce | 138 d | 166 | – |
| $^{145}$Nd | 8.3 | $^{141}$Ce | 32.5 d | 145 | $^{141}$Nd from $(\gamma, n)$ |
| $^{153}$Eu | 52.2 | $^{149}$Pm | 53.1 h | 286 | $^{149}$Eu from $(\gamma, 2n)$ |
| $^{160}$Gd | 21.9 | $^{156}$Sm | 9.4 h | 204 | – |
| $^{163}$Dy | 24.9 | $^{159}$Gd | 18.5 h | 364 | $^{159}$Dy from $(\gamma, n)$ |
| $^{176}$Yb | 12.8 | $^{172}$Er | 49.3 h | 407 | – |
| $^{176}$Lu | 2.6 | $^{172}$Tm | 63.6 h | 1094 | $^{172}$Lu from $(\gamma, 3n)$ |
| $^{181}$Ta | 100 | $^{177}$Lu | 6.65 d | 208 | – |
| $^{187}$Re | 62.6 | $^{183}$Ta | 5.1 d | 246 | $^{183}$Re from $(\gamma, 2\mathrm{n})$ |
| $^{193}$Ir | 62.7 | $^{189}$Re | 24.3 h | 245 | $^{189}$Ir from $(\gamma, 2\mathrm{n})$ |
| $^{203}$Tl | 29.5 | $^{199}$Au | 75.3 h | 158 | – |
| $^{207}$Pb | 22.1 | $^{203}$Hg | 46.6 d | 279 | $^{203}$Pb from $(\gamma, \mathrm{n})$ |

The bremsstrahlung radiation was generated by 23 MeV electron beam and used for targets activation downstream the 3 mm W converter and 25 mm Al radiator for stopping of the electrons. Past irradiation during about 5 hours at the electron beam intensity of 10 μA, the induced activity measurements were continued over one week and in some cases even longer, up to one month. The gamma spectra were taken using HP Ge detector with energy resolution better 1.8 keV by the $^{60}$Co lines. The set of standard sources was used for energy and efficiency calibration of the detector. Series of the spectroscopic measurements were resulted in observation of the γ-lines belonged to the products of (γ, α) reactions in In, Sn, $^{176}$Yb, and Ta targets, despite a great activity of other radionuclides produced in more abundant reactions.

A number of produced $^{111}$Ag, $^{115}$Cd, $^{172}$Er, and $^{177}$Lu atoms could be evaluated from the measured γ-line counts using the standard program for gamma spectra processing. The decay schemes of radionuclides are taken from Nuclear Data Sheets, and the standard formalism is used for evaluation of the time-efficiency factors accounting the accumulation and decay of radioactive products. Similar method has been applied for the detection of (γ, *p*) products. The (γ, *n*) reactions were abundantly manifested by corresponding activities in all measured γ-spectra. Therefore, it was natural to calibrate the observed yields of (γ, *p*) and (γ, α) processes to the yield of the most abundant (γ, *n*) reaction. The ratios are reduced in Tables 2 and 3 for (γ, α) and (γ, *p*) reactions, respectively.

Table 2. Experimental results for the yield of (γ, α) reactions

| Target | Product | Halflife | $E_\gamma$, keV | Relative yield: (γ,α)/(γ,n) | Threshold parameter: $(E_{th}+B_c)$, MeV |
|---|---|---|---|---|---|
| $^{109}$Ag | $^{105}$Rh | 35.4 h | 319 | $\leq 3.5\cdot 10^{-4}$ | 13.56 |
| $^{113}$Cd | $^{109}$Pd | 13.7 h | 88 | $\leq 2.4\cdot 10^{-4}$ | 14.32 |
| $^{115}$In | $^{111}$Ag | 7.45 d | 342 | $(3.1\pm 0.5)\cdot 10^{-5}$ | 14.45 |
| $^{119}$Sn | $^{115}$Cd | 53.46 h | 528 | $(2.9 \pm 0.4)\cdot 10^{-5}$ | 15.31 |
| $^{176}$Yb | $^{172}$Er | 49.3 h | 407 | $(0.4 \pm 0.1)\cdot 10^{-5}$ | 14.75 |
| $^{181}$Ta | $^{177}$Lu | 6.65 d | 208 | $(0.70 \pm 0.12)\cdot 10^{-5}$ | 14.51 |
| $^{193}$Ir | $^{189}$Re | 24.3 h | 245 | $\leq 2.8\cdot 10^{-4}$ | 15.84 |

Accurate measurements were carried out also for Ag, Cd, and Ir targets, but the (γ, α) yields could only be estimated in a form of the upper limit. This is due to the presence of internal background as was explained above. As follows from Table 2, the (γ, α) relative yields are as low as about $10^{-5}$ in all successfully measured cases, and the upper limits at a level of $10^{-4}$ also do not contradict that. The long-lived $^{115m}$Cd and $^{177m}$Lu isomers could not add a noticeable contribution into the total yield of the reactions because the isomer yield is suppressed by orders of magnitude [2] due to the great values of their spins: 11/2 and 23/2, correspondingly. Finally, the low value, of about $10^{-5}$, for ($\gamma$, $\alpha$)-to-($\gamma$, $n$) ratio is evidently proved in the series of experiments with the medium-weight target nuclei.

Table 3. Measured yields of the ($\gamma$, p) reactions

| Target | Reaction | Product | Halflife | $E_\gamma$, keV | Relative yield: ($\gamma$, p)/($\gamma$,n) | ($E_{th}$+$B_c$), MeV |
|---|---|---|---|---|---|---|
| $^{nat}$Cd | $^{112}$Cd ($\gamma$, p) | $^{111}$Ag | 7.45 d | 342 | $(1.15\pm0.15)10^{-2}$ | 14.83 |
|  | $^{113}$Cd ($\gamma$, p) | $^{112}$Ag | 3.12 h | 617 | $(1.00\pm0.15)10^{-2}$ | 14.89 |
|  | $^{114}$Cd ($\gamma$, p) | $^{113}$Ag | 5.37 h | 299 | $(0.98\pm0.15)10^{-2}$ | 15.40 |
| $^{nat}$Sn | $^{118}$Sn ($\gamma$, p) | $^{117g}$In | 43.2 min | 553 | $(4.9\pm0.5)10^{-3}$ | 15.42 |
|  |  | $^{117m}$In | 116 min | 315 |  | 15.74 |
| $^{176}$Yb (95%) | $^{174}$Yb ($\gamma$, p) | $^{173}$Tm | 8.24 h | 399 | $(0.75\pm0.15)10^{-3}$ | 15.72 |
| $^{nat}$Hf | $^{178}$Hf ($\gamma$, p) | $^{177g}$Lu | 6.65 d | 208 | $(1.8\pm0.4)10^{-3}$ | 15.33 |

The ($\gamma$, $p$) products sometimes have been observed at the spectra measured for detection of the ($\gamma$, $\alpha$) reactions and in some cases the special irradiations have been carried out for the yield of ($\gamma$, $p$). The measured values are reduced in Table 3 and it is clear that the ($\gamma$, $p$)-to-($\gamma$, $n$) ratio appears typically at a level of $10^{-3}$. This result does not contradict the known data: see, for instance, a compilation in [2]. Higher probability of proton emission did allow reliable measurements of the ($\gamma$, $p$) yields in published works, unlike the deficit for the ($\gamma$, $\alpha$) yields.

## 3. Discussion of the reaction mechanism

A role of ($\gamma$, $\alpha$) reactions in nucleosynthesis at stellar conditions is out of discussion here because such special topics must be developed and described elsewhere. We are interested for the nuclear-physics conclusions. From the yields of many reactions, it is possible to deduce some regular trends after analyses. In fig. 1, the relative yields of (γ, α) and (γ, *p*) reactions are plotted versus the threshold parameter ($E_{th}$ + $B_c$). In accordance with the regularity established in [2], the (γ, *p*) yield decreases systematically with the threshold growth. Such a trend is not as evident for the (γ, α) yield. The scattering of points corresponding to (γ, α) is probably explained by the individual properties of targets. The latter factor makes, obviously, lower significance for the (γ, *p*) reactions. In general, two types of reactions could be compared evaluating the geometric mean values of the yields.

The corresponding mean values are shown in fig. 1 by the dashed lines, and it is clear that the probability of the (γ, *p*) reaction exceeds the one for (γ, α) by a factor of about 350, in mean

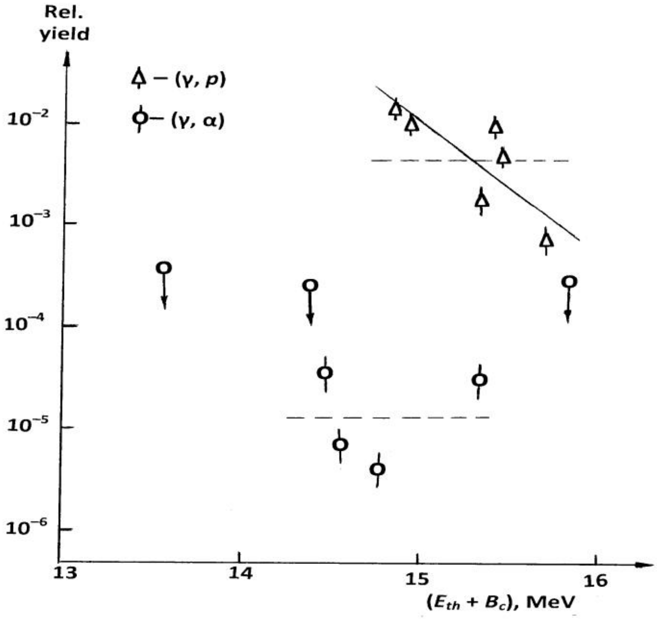

Figure 1. Relative yields of (γ, p) and (γ, α) in ratio to (γ, n) reactions at $E_e$=23 MeV versus threshold parameter value.

This conclusion is not changed even if the regular decrease of the (γ, *p*) yields is taken into account as is shown by a solid line in fig. 1.

The mentioned factor of 350 could not be explained either by the threshold, or by the spin dependencies of the reaction abundance. There remain only qualitatively different features corresponded to the emission of a single proton, unlike the composite α particle. The pre-formation factor must probably be in account for alphas. Being in an absolute value near 1/350, this factor corresponds by an order of magnitude to the known pre-formation factors in α-decay of the ground-state nuclei. Therefore, inside a nucleus, the nucleons remain unperturbed before α emission both in (γ, α) reaction and in α-decay. Higher mass number of alphas compared to protons could influence the probability of penetration through the barrier, but in our case, the processes well above the barrier are detected: $(E_e - E_{th} - B_c) \approx 8 - 10$ MeV. Thus, a subbarrier penetration factor is not a real reason for lower probability of α emission compared to protons. A low magnitude of the pre-formation factor confirms an absence of α-clusters ready for the emission. This conclusion contradicts the models involving a complete α-clustering inside the nuclear matter. Clusters may permanently exist in light nuclei, but not in $A \geq 100$ species. Other points are open for the additional analysis, for instance, the idea on nucleon-nucleon correlations with formation of the quark bags instead of the nucleon gas (liquid) in the bound nuclei.

Without theoretical analysis, it would be difficult to say, whether our result cancels the presence of multi-quark objects inside a nucleus, or just means some restricted probability for the short-range nucleon-nucleon correlations.

Let’s discuss now possible consequences of the presented results for interpretation of nuclear reactions, in general approach. Excitation functions and spectral-angular characteristics for particles emitted in nuclear reactions were widely studied since many decades. But the processes developed inside a nucleus and preceding the emission remain typically buried. Many authors suppose that the mechanism details are hardly possible to specify. In general, the reactions are distributed over three classes: direct reactions, pre-equilibrium emission, and decay of the excited compound nucleus. This classification in itself is not very productive until their inherent peculiarities are well established. In principle, there is a little mysterious scenario, how a bound nucleon sitting at definite single-particle orbit transmits to the external space. Emission of the composite particles seems even more sophisticated. One could dream about truely direct mechanism, type of knock-out, when an individual nucleon is kicked out due to the momentum transfer from the projectile. Direct knock-out requires a presence of the particle ready for emission and a presence of the impact momentum. Definitely, these conditions are not common. For instance, the initiating photon carries in the insignificant momentum. In the interim, the emission from compound nucleus happens due to the momentum created randomly by statistical fluctuations.

It was explained above that (γ, α) reaction yield is suppressed due to the pre-formation factor. But in reactions induced by heavy ions, α-particles are emitted with a great probability, not lower than that for protons, as is well-known since 50 years [9]. The only explanation of this fact could be found in assumption that α-particle is formed due to an impact of heavy projectile through the mechanism of "internal coalescence". Schematic illustration is shown in fig. 2. Directed flow of nucleons is created by the projectile momentum and the group of nucleons is joined together with formation of the well-bound alpha cluster. In literature, there is assumed direct mechanism for alpha emission induced by heavy ions, but in reality, this appears as a two-step mechanism.

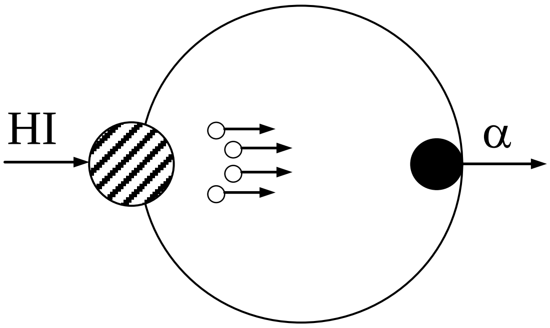

Figure 2. Schematic illustration of the internal coalescence mechanism for α-cluster formation by heavy projectile.

Even more paradoxical looks an idea that the nucleon emission from heavy targets also requires the preceding re-arrangement of the nucleon orbits. The nucleons sitting at the upmost orbits in the unexcited nucleus are characterized by a great value of the orbital momentum $l$, like 5, 6, and 7. The Fermi distribution is established past energy equilibration in the compound nucleus and the outer nucleons are also characterized by a high angular momentum. Individual nucleon must go through a stage of the orbital momentum exchange with other nucleons because the emission with $l$ = (5-7) is suppressed by the centrifugal barrier. There is described in the text books that transmission coefficients for neutrons are reasonably high only at $l$ = 0, 1, 2, and neutrons are emitted as *s*, *p*, and *d* waves. The conclusion follows that each neutron evaporation event must be, at least, a two-step process. The importance of the pre-arrangement of the nucleon momentum prior the emission is normally neglected in literature. But this may suppress an absolute rate of the emission reducing the statistical widths: $\Gamma_n$, $\Gamma_p$, $\Gamma_\alpha$ …

## 4. Summary

Yields of seven (γ, α) reactions are measured using the bremsstrahlung radiation at the end-point energy of 23 MeV, and pretty low probability of about $10^{-5}$ is deduced for the (γ, α)-to-(γ, *n*) yield ratio. Probably, the pre-formation factor for alphas must be in account. This result contradicts the observations of a great probability of alpha emission in reactions with heavy ions. Arisen puzzling could be resolved assuming the formation of α cluster due to the impact momentum of a heavy projectile through the mechanism of “internal coalescence”. Therefore, the reaction is going via two steps, and the truely direct mechanism is not supported. In many other cases, the re-arrangement of nucleons must precede the successful product emission. Moreover, the models describing a nucleus as the construction made of α clusters are not supported because the pre-formation factor is needed for treatment of both α-decay and (γ, α) reactions. Short-range nucleon-nucleon correlations leading to a formation of the multi-quark objects (quark bags) in nucleus could also influence the (γ, α) probability. This point must be additionally analyzed in theory for conclusive simulation of the experimental data.

## Acknowledgements

A.G. Belov and G.A. Bozhikov are gratefully acknowledged for the beam supply and for the assistance in γ spectroscopic measurements.

## References

1. V. I. Kukulin, *Phys. of Atomic Nuclei*, **74**, 1594 (2011).
2. S. A. Karamian, JINR E15–2012–65, Dubna 2012; Submitted to *Phys. of Atomic Nuclei*.
3. Y. M. Volkov, A. I. Ignatiev, G. A. Kolomenskii, et al., *Phys. of Atomic Nuclei*, **32,** 595, (1980).
4. B. S. Dolbilkin, Sh. Kan, T. Kim, et al., *Bull. RAS, Phys*. **55**, 967 (1991).
5. N. P. Dikiy, A. N. Dovbnya, Y. V. Lyashko, et al., Book of Abstracts of International Conference NUCLEUS 2007, Voronezh, 2007, p. 293.
6. I. N. Vishnevsky, V. A. Zheltonozhsky, I. M. Kadenko, et al., Ibid, p. 121.
7. S. A. Karamian, J. de Boer, Yu. Ts. Oganessian, et al., *Z. Phys.* A **356**, 23 (1996).
8. R. Bass, *Lect. Notes in Phys.*, **117**, 281 (1980).
9. H. C. Britt and A. R. Quinton, *Phys. Rev.*, **124**, 877 (1961).