E15-2013-93

S. A. Karamian [*]

Z-DEPENDENCE OF THE (γ, α) REACTION YIELD

Submitted to «Ядерная физика»

---

[*] E-mail: karamian@nrmail.jinr.ru

Карамян С. А. E15-2013-93

Зависимость выхода реакции (γ, α) от Z-ядра

Реакции с фотонами могут служить источником данных о внутренних связях и корреляциях нуклонов в тяжелых ядрах. Движение нуклонов оказывается только слегка возмущенным под действием электромагнитного поля, в отличие от реакций с сильно взаимодействующими частицами. Выход реакций (γ, α) можно использовать для проверки теоретических моделей, предполагающих полную α-кластеризацию ядер или присутствие в них мультикварковых объектов. При граничной энергии тормозного излучения 23 МэВ на разных ядрах мишеней измерены относительные выходы реакций (γ, n), (γ, p) и (γ, α). Доказано, что вероятность (γ, α)-процесса намного меньше, чем (γ, p), несмотря на практически сходные значения порогового и спинового факторов в реакциях обоих типов. Альфа-кластеризация не подтверждена. Обсуждены механизмы эмиссии частиц в ядерных реакциях, и прояснены некоторые детали.

Работа выполнена в Лаборатории ядерных реакций им. Г. Н. Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2013

Karamian S. A. E15-2013-93

Z-Dependence of the (γ, α) Reaction Yield

Reactions induced by photons may be used as a probe of 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 multiquark 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 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.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna, 2013

INTRODUCTION

The status of nucleons within a nuclear matter is not yet well clarified despite great interest for decades. The simplest assumption that nucleons conserve their individual properties the same as in vacuum has been questioned from different points of view. An array of ideas has been suggested: to replace the nucleons by quasiparticles, or by interacting bosons inside the nucleus, either to suppose the complete alpha-clustering in nuclear matter. In the past decade, an idea of short range nucleon–nucleon correlations with formation of a quark bag attracts an attention, and it is being tested in reactions at GeV energies [1]. The «cumulative effect» was known even earlier. The models mentioned above find some successful application in simulation of different processes. Additional tests in experiments are yet required. We propose now to use the reactions of nucleons and α-particle emission induced by bremsstrahlung at moderate endpoint energy of about 20–30 MeV. At higher energies, the consequent emission of nucleons becomes probable, or even dominates, and this makes it difficult to deduce the regularities in the yield of different-type reactions.

Recently, there have been reached [2] evidences for regular threshold dependence of the yields of photon-induced reactions. Abundant data are available in literature on the yields of the (γ, n), (γ, p) and partially (γ, d) reactions, while individual attempts of [3–7] to observe the (γ, α) yields demonstrate a significant scattering of the resulted values. Relative yields were systematized [2] versus the $(E_e - E_{\rm th} - B_C)$ parameter containing an excess of the end-point energy E_e above the sum of reaction threshold $E_{\rm th}$ and Coulomb barrier B_C for particle emission.

The B_C values are calculated using a 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 decreasing with the growth of the aforementioned effective threshold parameter of $(E_{\rm th} + B_C)$ at the moderate end-point energy $E_e \leqslant 30$ MeV. For (γ, α) reactions, the systematic data collection has not yet been available in the domain of heavy and medium weight targets. Therefore, experimental studies of the (γ, α) reactions are relevant for exploration of the nuclear-process mechanism in some details. In particular, a comparison of the nucleon and alpha emission rates may clarify the questionable point about the nucleon–nucleon correlation status. Complete alpha-clustering in heavy nuclei could at least be tested.

1. EXPERIMENTAL POSSIBILITIES AND SUCCESSFUL MEASUREMENTS

Shortcomings in the (γ, α) reaction yields could be covered in regular measurements with 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 form of enriched isotopes, the convenient decay properties for detection of the product activity and reasonably soft background restrictions. Inspecting the Nuclide Chart from $A = 109$ to 207, we 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 spectroscopic data were taken into account following the Nuclear Data Sheets and Table of Isotopes. The most favorable cases are distinguished and characterized in Table 1.

The mass numbers of potential targets correspond to the domain of the medium-weight species up to the alpha-decaying nuclides. The activities shown in Table 1 are chosen because they are characterized by the relatively intense γ

Table 1. Encounter data for the (γ, α) experiment

Target	Abundance, %	Product	Halflife	E_γ, 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 (γ, 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 (γ, n)
^{143}Nd	12.2	^{139}Ce	138 d	166	—
^{145}Nd	8.3	^{141}Ce	32.5 d	145	^{141}Nd from (γ, 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 (γ, 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 $(\gamma, 2n)$
^{193}Ir	62.7	^{189}Re	24.3 h	245	^{189}Ir from $(\gamma, 2n)$
^{203}Tl	29.5	^{199}Au	75.3 h	158	—
^{207}Pb	22.1	^{203}Hg	46.6 d	279	^{203}Pb from (γ, n)

lines convenient for detection at moderate halflives. The major problem of such experiments would be the presence of the background radiation generated by the isobaric nuclides due to the same transition at the same daughter nuclide but after ε instead of β^- decay. This internal physical background cannot be eliminated by a better shielding of the detector. Fortunately, in several cases, the background is absent; they must be considered the best for reliable detection of the (γ, α) reaction and for the yield measurements. For some other targets, the experimental conditions could be improved by using enriched isotopes because the background is created typically due to reactions with auxiliary isotopes in the target. A possibility to distinguish the reaction products by their halflives sometimes is productive.

In the present experiments, the (γ, α) yields were successfully measured in seven cases. The majority of targets were metal foils of natural isotopic composition, except for the ^{109}Ag, ^{113}Cd, and ^{176}Yb enriched isotopes. The Al foils were used to pack the target materials for irradiation. Full weight of them was typically about 0.2–0.5 g. The $^{\rm nat}$Pb target was taken in amount of 10 times greater for detection of (γ, α) very low yield, $\sim 10^{-6}$. Fortunately, in this case the advantageous possibility could be exploited. The (γ, α) and (γ, n) reactions were determined by the same γ line at 279 keV emitted in decay of both ^{203}Hg and ^{203}Pb products. These nuclides were distinguished by their halflives: 46.6 and 2.16 d, correspondingly, allowing complete decay of the latter activity for successful measurements of the former one. Such options seem even convenient for the measurement of the (γ, α) to (γ, n) or (γ, p) yield ratios.

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 the electrons. After irradiation for about 5 hours with electrons at the beam intensity of 10 μA, the induced activity measurements were continued over one week and in some cases even longer, up to months. The gamma spectra were taken using the HPGe 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 spectroscopic measurements are resulted in observation of the γ lines of the following products of (γ, α) reactions: ^{105}Rh, ^{109}Pd, ^{111}Ag, ^{115}Cd, ^{172}Er, ^{177}Lu, and ^{203}Hg, despite the great activity of other radionuclides produced in more abundant reactions. A number of radioactive-product atoms could be evaluated from the measured count rate in the characteristic γ lines. The standard program for gamma-spectra processing was resulted in energy position and area for each well-resolved γ line.

Decay schemes for radionuclides were taken from Nuclear Data Sheets. The standard formalism was used for calculation of the time-efficiency factors affecting the accumulation and decay of radioactive products. Similar method was applied for the detection of (γ, p) products. The (γ, n) reactions were abundantly manifested with the corresponding activities in all measured γ spectra. Spectrometry with fine energy resolution allows accurate isolation of the lines belonged to (γ, p) and (γ, α) products. Finally, 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 given in Tables 2 and 3 for (γ, α) and (γ, p) reactions, respectively. Calibration to the yield of the most probable reaction excludes the influence of the total photon-absorption cross section. The giant dipole resonance parameters are varied for different targets due to dependence on the mass number and because of possible microscopic effects. The (γ, n) reaction is practically used as a spectator in these measurements and serves to find other reaction probabilities.

As follows from Table 2, the (γ, α) relative yields are typically lower than 10^{-4} in measured cases, and the upper limit for ^{189}Re product also does not

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

Target	Product	Halflife	E_γ, keV	Relative yield: $(\gamma, \alpha)/(\gamma, n)$	Threshold parameter: $(E_{\rm th} + B_C)$, MeV
^{109}Ag	^{105}Rh	35.4 h	319	$(1.5 \pm 0.3) \cdot 10^{-4}$	13.56
^{113}Cd	^{109}Pd	13.7 h	88	$(2.4 \pm 0.3) \cdot 10^{-4}$	14.32
^{115}In	^{111}Ag	7.45 d	342	$(4.5 \pm 0.5) \cdot 10^{-5}$	14.45
^{119}Sn	^{115}Cd	53.46 h	528	$(3.9 \pm 0.4) \cdot 10^{-5}$	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	$\leqslant 2.8 \cdot 10^{-4}$	15.84
^{207}Pb	^{203}Hg	46.6 d	279	$(1.7 \pm 0.2) \cdot 10^{-6}$	17.51

Table 3. Measured yields of the (γ, p) reactions

Target	Reaction	Product	Halflife	E_γ, keV	Relative yield: $(\gamma, p)/(\gamma, n)$	$(E_{\rm th} + B_C)$, MeV
$^{\rm nat}$Cd	^{112}Cd (γ, p)	^{111}Ag	7.45 d	342	$(1.15 \pm 0.15) \cdot 10^{-2}$	14.83
	^{113}Cd (γ, p)	^{112}Ag	3.12 h	617	$(1.00 \pm 0.15) \cdot 10^{-2}$	14.89
	^{114}Cd (γ, p)	^{113}Ag	5.37 h	299	$(0.98 \pm 0.15) \cdot 10^{-2}$	15.40
$^{\rm nat}$Sn	^{118}Sn (γ, p)	^{117g}In	43.2 min	553	$(4.9 \pm 0.5) \cdot 10^{-3}$	15.42
		^{117m}In	116 min	315		15.74
	^{116}Sn (γ, p)	^{115m}In	4.49 h	336	$(5.1 \pm 0.7) \cdot 10^{-3}$	15.07
	^{114}Sn (γ, p)	^{113m}In	1.66 h	392	$(8.8 \pm 1.0) \cdot 10^{-3}$	14.32
^{176}Yb	^{174}Yb (γ, p)	^{173}Tm	8.24 h	399	$(0.75 \pm 0.15) \cdot 10^{-3}$	15.72
$^{\rm nat}$Hf	^{178}Hf (γ, p)	^{177g}Lu	6.65 d	208	$(1.8 \pm 0.4) \cdot 10^{-3}$	15.33

contradict that. The long-lived ^{115m}Cd and ^{177m}Lu isomers could add no noticeable contribution to the total yield of the reactions because the isomer yield was suppressed by orders of magnitude [2] due to the great values of their spins: 11/2 and 23/2, correspondingly. In the cases of ^{113}In and ^{115}In, the isomer spin is conversely lower than that of the ground state and the isomers collect the major part of the reaction strength, they are selected in Table 3.

The (γ, p) products sometimes have been observed at the spectra measured for detection of the (γ, α) reactions and in some cases the special irradiations have been carried out for the yield of (γ, p). The measured values are given in Table 3 and it is clear that the (γ, p)-to-(γ, n) ratio appears typically at a level of $10^{-2} - 10^{-3}$. This result does not contradict the known data compiled, for instance, in [2]. Higher probability of proton emission did allow reliable detection of (γ, p) reactions in many works, unlike the deficit of data for the (γ, α) yields. The explanation of the observed low (γ, α)-to-(γ, p) ratios for medium-weight targets is given below.

2. PROBABILITY OF ALPHA PARTICLE EMISSION

The role of (γ, α) reactions in nucleosynthesis at stellar conditions cannot be discussed here because this topic is too special and should be treated separately. We are interested solely in 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) in ratio to (γ, n) reactions are plotted versus the threshold parameter $(E_{\text{th}} + B_C)$. In accordance to the regularity established in [2], the (γ, p) yield decreases systematically with the growth of the threshold parameter $(E_{\text{th}} + B_C)$. The behavior of (γ, α) yields is not identical to (γ, p), even though $(E_{\text{th}} + B_C)$ values are comparable. The Coulomb barrier is higher for alphas, while E_{th} is reduced because of the great binding energy of four nucleons in α particle.

The scattering of points corresponding to (γ, α) is stronger manifested which could probably be explained by the individual properties of the targets. The slope of yield versus the threshold parameter is not as steep as for protons. Furthermore, the most significant peculiarity is visible in the absolute magnitude of relative yield which is lower for α emission by two orders of magnitude. This would be difficult to understand within threshold dependence of the photon induced yields. Indeed, for explanation, one could assume a lower penetrability factor for alphas due to their heavier mass. But this is not applicable to the case of reactions well above the barrier. In the present studies, the excitation energy exceeds the barrier by 6–10 MeV. In addition, the lower penetrability must be accompanied with a steeper decrease of the yield versus the growing threshold. Just the opposite trend is observed in the pattern shown in Fig 1.

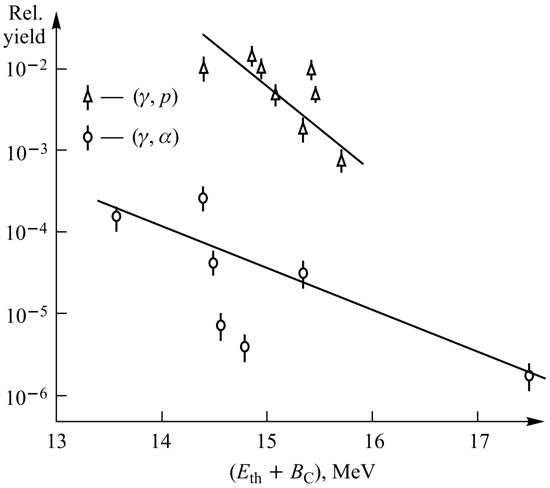

Fig. 1. Relative yields of (γ, p) and (γ, α) in ratio to (γ, n) reactions at $E_e = 23$ MeV versus threshold parameter value. Straight lines guide for eyes

Given all this, we suppose that there is a direct correlation of the $(\gamma,\ \alpha)$ yield with the target atomic number Z as is shown in Fig. 2. The regular function appears and may support the idea that α-emission probability involves a preformation factor decreasing with Z for heavy nuclides. In absolute value, this factor is of about 10^{-2}. In general, the idea of the preformation factor and even the magnitude look similar to the typical assumptions used in theory of radioactive alpha decay. When the medium-energy photon is absorbed by a nucleus this perturbs the intrinsic status of nucleons only slightly and the analogy to α decay seems applicable. The necessity of the preformation factor follows from the ratio of (γ, α) to (γ, p) yields. They could not be explained by the effect of different

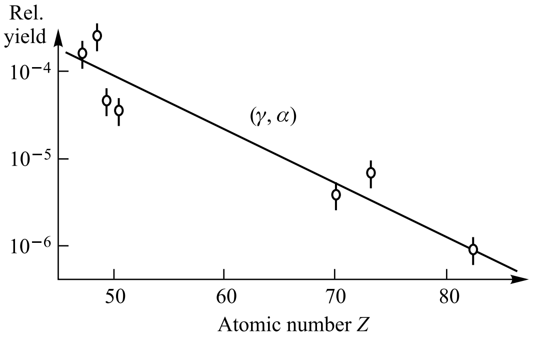

Fig. 2. Z-dependence of the (γ, α)-reaction yield. The solid curve guides for eyes

thresholds, even after reliable corrections, like common calibration and others. Probably, an explanation with the preformation factor is the only possible, and it reflects the qualitative difference of the composite α particle emission, unlike to single proton ready for release. Therefore, the nucleons remain uncorrelated as independent particles in a nucleus reducing the probability of both (γ, α) reaction and α decay. For the first step, alpha must be formed and then emitted. Clearly, the pre-equilibrium and direct mechanisms dominate in the energy range of 100–1000 MeV, but not with 23 MeV photons.

Low magnitude of the preformation factor indicates absence of α clusters in the target nucleus prior to the reaction. This conclusion contradicts the models involving a complete α-clustering inside the nuclear matter. Clusters may permanently exist in light nuclei, but not in $A \geqslant 100$ species. Other points are open for the additional analysis, for instance, the idea of nucleon–nucleon correlations with formation of the quark bags instead of the nucleon gas in the bound nuclei. Without theoretical analysis, it would be difficult to say whether our result denies the presence of multiquark objects inside a nucleus, or just indicates some restricted probability for the short range nucleon–nucleon correlations.

3. TO MECHANISM OF PARTICLE EMISSION

Let us discuss now possible consequences of the present results for interpretation of nuclear reactions, in general. Excitation functions and spectral-angular characteristics for particles emitted in nuclear reactions have been widely studied for many decades but the processes developed inside a nucleus and preceding the emission typically remain hidden. Many authors suppose that the mechanism details are hardly possible to specify. Traditionally, 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. There is a bit of a mystery in 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 truly direct mechanism, a kind of knock-out, when an individual nucleon is kicked out due to the momentum transfer from the projectile. Direct knock-out requires the presence of the particle ready for emission and of the impact momentum. Definitely, these conditions are not common. For instance, the initiating photon carries in the nucleus an 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 preformation factor. Similar factor was used in α-decay theory at zero excitation of a nucleus and also for treatment of the pre-equilibrium α emission [9] at high energies $\geqslant 100$ MeV. It seems to be the general property for α-particle release from the nucleus. But in reactions induced by heavy ions at moderate energy of 10 MeV/nucleon, α particles are emitted with a great probability higher than that for protons, as is well known since 50 years [10]. The alpha particle emission is characterized by high cross section of about 40% of total reaction cross section and emission velocity reaches the double velocity of the projectile ion. Relatively low energy of collision does not allow one to apply the theory of pre-equilibrium emission and to get a great probability of the process, either to explain the double velocity of alphas.

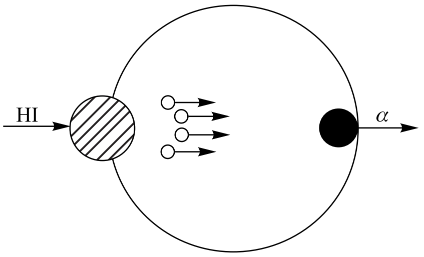

Fig. 3. Schematic illustration of the internal coalescence mechanism for α-cluster formation by heavy projectile

Reasonable explanations could be found if we assume that α particle is formed due to the impact of heavy projectile through the mechanism of «internal coalescence». The schematic illustration is shown in Fig. 3. Because of incompressibility of nuclear matter, a flow of nucleons to the forward direction is created by the projectile momentum. The nucleons are stripped from the bound orbits and move with the velocity up to double velocity of the projectile. Then, they are joined together with formation of the well-bound alpha cluster. In [10] and later, there is always assumed the direct mechanism for alpha emission induced by heavy ions, but in reality, this appears as a two-step mechanism because α clusters are not present in the target and must be created by the dynamical process of collision.

The idea that the nucleon emission from heavy nuclei also requires the preceding rearrangement of the nucleon orbits seems even more paradoxical. Within shell model of nuclei, the nucleons sitting at the upmost orbits at zero excitation are characterized by the great values 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 the great orbital 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. According textbook [11], the transmission coefficients T_l 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 neutron evaporation in major must be a two-step process. The importance of the prearrangement of the nucleon momentum prior the emission is normally neglected in literature. However, this may suppress an absolute rate of emission reducing statistical width Γ_n.

CONCLUSION

Yields of seven (γ,α) reactions were measured using the bremsstrahlung radiation with endpoint energy of 23 MeV in a high-sensitivity experiment stressed by the probability lower 10^{-4} to 10^{-6} for (γ,α)-to-(γ,n) ratio. Evidently, the alpha preformation factor must be responsible for the reduced probability in analogy to the known case of radioactive α decay. The pre-equilibrium α emission sometimes is also treated using preformation factor, but exciton model at high energy $\geqslant 100$ MeV makes in essence no similarity with the low-energy processes. Moreover, both low- and high-energy processes including our observations demonstrate the disagreement with the known since 50 years great probability of α emission in reactions with heavy ions at modest energy of about 10 MeV/amu. The puzzle could be resolved assuming formation of the alpha cluster due to impact momentum of a heavy projectile through the mechanism of «internal coalescence». Therefore, the reaction is going via two steps, and the truly direct mechanism does not seem viable. In many other cases, the rearrangement of nucleons must precede the successful product emission. This stage is commonly neglected in publications, but the corresponding effects could be found after developed studies. The model describing nucleus as a construction made of α clusters is not confirmed because probability of emission is suppressed according to the present experiment. Short-range nucleon–nucleon correlations leading to formation of the multiquark objects (quark bags) in nucleus could also influence the (γ,α) probability. This point requires an additional analysis for conclusive simulation of the data in theory.

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. *Kukulin V. I.* // Yad. Fiz. 2011. V. 74. P. 1594 (Phys. Atom. Nucl. 2011. V. 74. P. 1567).
2. *Karamian S. A.* JINR Preprint E15-2012-65. Dubna, 2012; Submitted to Phys. At. Nucl.
3. *Volkov Y. M. et al.* // Yad. Fiz. 1980. V. 32. P. 595 (Phys. At. Nucl. 1980. V. 32. P. 582).
4. *Dolbilkin B. S. et al.* // Bull. Russ. Acad. Sci. Phys. 1991. V. 55. P. 967.
5. *Dikiy N. P. et al.* // Book of Abstracts. Intern. Conf. «Nucleus 2007» / Ed. by A. K. Vlasnikov. St. Petersburg, 2007. P. 293.
6. *Vishnevsky I. N. et al.* // Ibid. P. 121.
7. *Karamian S. A. et al.* // Z. Phys. A. 1996. V. 356. P. 23.
8. *Bass R.* // Lect. Notes in Phys. 1980. V. 117. P. 281.
9. *Wu J. R., Chang C. C., Holgren H. D.* // Phys. Rev. C. 1979. V. 19. P. 698.
10. *Britt H. C., Quinton A. R.* // Phys. Rev. 1961. V. 124. P. 877.
11. *Blatt J. M., Weisskopf V. F.* Theoretical Nuclear Physics. N. Y., London: Wiley, 1952.

Received on August 26, 2013.

Корректор *Т. Е. Попеко*

Подписано в печать 15.11.2013.
Формат 60 × 90/16. Бумага офсетная. Печать офсетная.
Усл. печ. л. 0,93. Уч.-изд. л. 1,26. Тираж 225 экз. Заказ № 58117.

Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/