TWO- AND THREE-BODY COLLINEAR DISINTEGRATIONS OF HEAVY NUCLEI AT MODERATE EXCITATION ENERGIES

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Introduction

After the discovery of the fission phenomenon, its binary concept was suggested by L. Meitner, O. Frisch, and N. Bohr in close analogy with the biological process of cell multiplication. Everybody was impressed by the very early paper of Bohr and Wheeler entitled "The Mechanism of Fission" and by how much of the fission process they could explain and predict just a few months after the fission was discovered. They used singular "Mechanism" assuming it to be universal. Up to now it was used to discuss experimental data on fission of various nuclei at different excitation energies. In fact, the detection of two massive collinear fragments seems to confirm the two-body character of the nuclear fission reaction. However, this experimental result is a necessary but not sufficient signature of the two-body kinematics. Being consistent with the experimental observation of the two collinear fragments, one could consider a more general case when the formation of the third fragment of very low energy is possible. The nuclear fission reaction should be written then as

$$^{M_0}Z_0 \rightarrow ^{M_1}Z_1 + ^{M_3}Z_3 + ^{M_2}Z_2, \tag{1}$$

where M_k and Z_k are the mass numbers and electric charges of the initial fissioning nucleus and of three fragments. Making use of the momentum balance of the moving fragments as a condition for the third one to be at rest, one obtains

$$M_1/M_2 = \sqrt{Z_2/Z_1}. \tag{2}$$

It is rather difficult to satisfy eq. (2) for the mass-asymmetric fission process that confirms the two-body character of low-energy nuclear disintegrations. However, eq. (2) can be satisfied in the strictly symmetric ($M_1 = M_2$ and $Z_1 = Z_2$) spontaneous decay of superheavy nuclei that does not permit to identify unambiguously the two-body mechanism of the nuclear fission reaction in this case.

General analysis of possible variants of the reaction leads to the conclusion that a variety of the nuclear disintegration processes could exist. A well-known binary fission into mass-asymmetric fragments corresponds to the case of $Z_3 = 0$. For small values of Z_3 the separation of two fragments is accompanied by the appearance of the slow charged particle. In the case of large Z_3 close to Z_0 the incomplete fission is possible with the production of two intermediate mass fragments and a massive residual nucleus at rest. When all the three charges in reaction (1) are comparable with each other, a collinear tripartition process of the heavy nucleus takes place.

Below we try to trace the experimental development in the fission study with accelerator beams which could support the concept of several fission mechanisms in heavy nuclei at moderate excitation energies.

Experiments with measurement of two kinetic energies ($2E$) of complementary fragments

Our experimental study of fission started using a bremsstrahlung beam of the PNPI electron synchrotron with the help of a back-to-back ionization chamber which could measure two kinetic energies of complementary fission fragments in coincidence [1–3]. Experiments with deuteron and α-particle beams were also performed [4], in which silicon surface-barrier detectors were used instead of the ionization chamber. As a result, it was proved that the mass-asymmetric fission process dominates for six investigated compound nuclei: ^{232}Th, ^{234}Pa, ^{236}U, ^{238}U, ^{240}Np, and ^{242}Pu. Total kinetic energies (E) of the mass-symmetric fission fragments had identical distributions with a constant $< E >$ value in fission processes of ^{238}U nuclei induced by bremsstrahlung beams with maximum energies $E_{\gamma max} = 17.5$, 30, 50, and 70 MeV. The obtained experimental results rejected the possibility of collinear tripartition process and corroborated the two-body character of nuclear disintegration. However, the experimental data on mean kinetic energies of light and heavy groups of fission fragments allowed to determine the mass region of fissioning nuclei where the condition (2) might be fulfilled and the collinear tripartition process might be observable. As seen from Fig. 1, fission process becomes predominantly mass-symmetric for nuclei with masses larger than 257 ± 3 u. Prior to 1970, all the experiments on spontaneous and neutron induced fission showed asymmetric mass distributions. The first observation of strongly enhanced mass-symmetric fission was reported for spontaneous fission of ^{257}Fm nuclei in 1970 and confirmed in 1971 by J.P.Balagna et al. (Phys. Rev. Lett., 1971. V.27. P.45). It was identified as a conventional fission process with two massive detectable fragments.

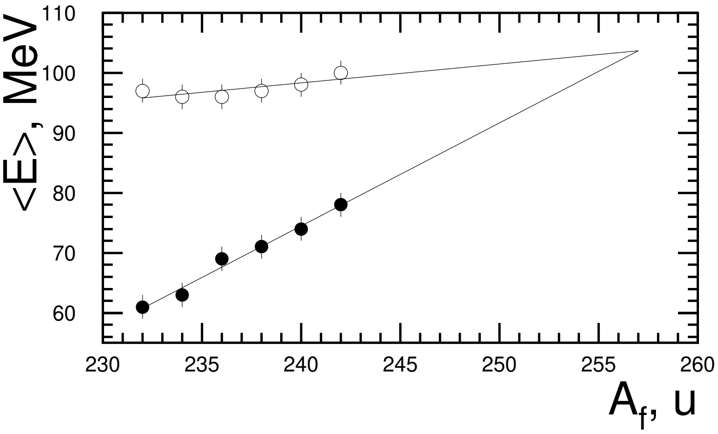

Fig. 1. Dependence of the average kinetic energy on the mass of the fissioning nucleus. Open circles – light-mass fragment group, solid circles – heavy-mass fragment group.

Incomplete fission process

When the sum of the masses and the sum of the charges of the two detectable collinear fragments are smaller than the mass and the charge of the fissioning nucleus, respectively, it is an unambiguous token of the incomplete fission process. Such a process was never mentioned in modern scientific literature, so it could not be recognized in experiments performed at moderate excitation energies. The processes of the incomplete fission were registered as ternary fission of ^{238}U (R.H.Iyer and J.W.Cobble, Phys. Rev., 1968. V.172. P.1186; K.W.MacMurdo and J.W.Cobble, Phys. Rev., 1969. V.182. P.1303) and ^{232}Th (T.C.Roginski et al., Phys. Rev., 1971. V.C4. P.1361) induced by intermediate energy ^{4}He and ^{3}He ions. The first of above mentioned papers contains evidence of ternary fission of an excited heavy nucleus into three fragments of comparable sizes. This result was obtained from a radiochemical study of fragments of ^{238}U fission induced by 20–120 MeV helium ions. The absolute formation cross sections for ^{24}Na, ^{28}Mg, ^{31}Si, ^{38}S, ^{47}Ca, ^{56}Mn, and ^{66}Ni in the fission induced by 39 ± 1 MeV helium ions determined clearly the transition region between binary and ternary fission, the crossover occurring near $A = 47$ u. The corresponding data for ^{183}Ta and ^{184}Ta and upper limits for ^{199}Au, ^{209}Pb, and ^{212}Pb also indicated the absence of any possible complementary heavy fragments. The fission nature of the light ternary fragments was confirmed by the forward-to-backward recoil properties. In our opinion, the results obtained in the cited above papers could be also interpreted in terms of the collinear tripartition process. For example, in the case of the experiment carried out by R.H.Iyer and J.W.Cobble the reaction could be

$$^{238}_{92}\mathrm{U} +^{4}_{2}\mathrm{He}\,(T = 39\ \mathrm{MeV}) \rightarrow^{28}_{12}\mathrm{Mg} +^{178}_{70}\mathrm{Yb} +^{28}_{12}\mathrm{Mg} + 8n. \qquad (3)$$

The known nuclear masses were used to calculate the energy release $Q = 64.7$ MeV in reaction (3). Because of the momentum compensation of the incoming α-particle only a half of its initial kinetic energy is available for the $^{28}_{12}$Mg kinetic energy:

$$E_{Mg} = (2Q + T)/4 = 42\ \ \mathrm{MeV}.$$

This value is in agreement with experimental measurements of R.H.Iyer and J.W.Cobble. The paper of B.A.Bochagov et al. [4] was cited by K.W.MacMurdo and J.W.Cobble who discussed the difference between the experiments with ^{3}He and ^{4}He ions.

Reasons to search for collinear tripartition of heavy nuclei induced by 1 GeV protons

Measurements of the angular and energy distributions of the neutrons in coincidence with α-particles accompanying two massive fragments of the spontaneous fission of ^{252}Cf showed that nuclear-unstable species were formed. They turned out to be ^{5}He in the ground state and ^{6}He* in the excited states [5]. Neutron excessive He nuclei disintegrated in times comparable with separation time of the two massive fragments:

$$^5\mathrm{He} \rightarrow \alpha + n \qquad \text{and} \qquad ^6\mathrm{He}^* \rightarrow \alpha + 2n.$$

These processes contribute noticeably to the energy spectrum of α-particles measured in coincidence with two massive fragments of the spontaneous fission of ^{252}Cf. This example shows that the third fission fragment in the collinear tripartition process might be a nuclear-unstable object. Such a property together with a low-velocity of this object requires the identification of the third fragment by the thoroughly analyzed kinematics of the two detected fragments. Besides, the existence of the third unstable fragment could be observed via its decay products. This was the goal of the experiment on the disintegration of ^{238}U nuclei loaded into nuclear emulsion. The experiment was performed using the proton beams with energies of 0.46–9 GeV at Dubna and Gatchina [6]. Fig. 2 shows the experimental dependence of the average folding angle between the two massive fragments on the number of accompanying charged particles for the ^{238}U disintegration induced by 1 GeV protons. At any available charged particle multiplicity one can find the events with two massive fragments separating collinearly in nuclear emulsion. Thus the experiments with 1 GeV protons have shown that there were some reasons for studying the collinear three-body disintegration of the ^{238}U nuclei.

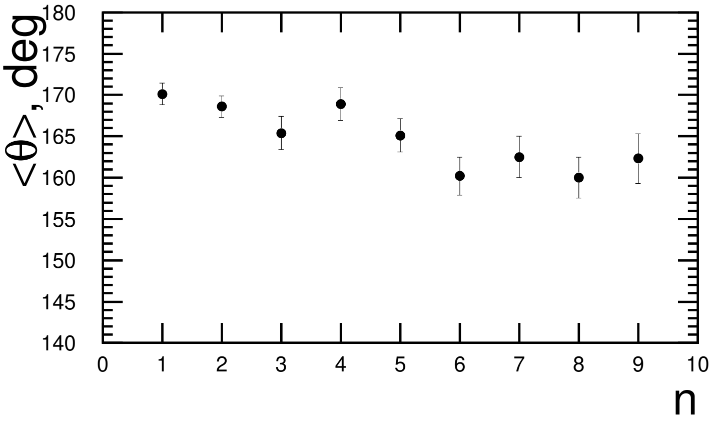

Fig. 2. Experimental dependence of the average folding angle $< \Theta >$ between the two massive fragments on the number n of accompanying charged particles for ^{238}U disintegration induced by 1 GeV protons. The average folding angle for coplanar ternary fission induced by 1 GeV protons was found to be 119° ± 1°.

It is clear, however, that the formation of the three fragments of comparable masses in the nuclear disintegration is not necessarily accompanied by the collinear separation of the two of them. Assuming that all the three fragments should have similar properties, one can try to detect all of them. In the experiment [7] on the ^{238}U disintegration induced by 1 GeV protons the detection of three-prong events was performed using the backing free 200 μm layers of nuclear emulsion. In order to improve the detection efficiency of the three-prong events related to the fragments of comparable masses, the sensitivity threshold of nuclear emulsion was increased up to the values of ionization losses for the charge $Z = 10$. This allowed to increase considerably the total proton flux at sufficient transparency of the nuclear emulsion layers. For $3.43 \cdot 10^5$ two-prong events, 133 three-prong events were detected, all three tracks belonging to the fragments of comparable masses. None of four- or more-prong events were observed. After correcting for the scanning efficiency one could find the ratio of the probabilities for ^{238}U fission into three and two massive fragments to be equal to $(4.7 \pm 0.5) \cdot 10^{-4}$. The total probability for the three-prong events is 20 times lower than that for the events with two massive fragments and high multiplicity of accompanying charged particles. Among 399 angles between two of the three tracks it was not found any exceeding the value of Θ=165°. These quantitative experimental estimates related to the coplanar three-body disintegrations allowed us to choose the geometry for investigation of the collinear three-body splitting with the help of the angle-velocity-energy correlation spectrometer (AVECS).

Experiments with the $(2E, 2V)$ measurements

Designed and constructed at PNPI, the AVECS device was among a few instruments of that type in the world (see, for example, Y.S.Kim et al., Nucl. Instr. Meth., 1993. V.A329. P.403). A large amount of information can be obtained with such coincidence spectrometer that considerably compensates the inconvenience of the equipment complexity.

At first, the pulse-height defect (PHD) in the silicon detectors has been measured with 8–17 MeV ^{19}F, 4–18 MeV ^{27}Al, 7–25 MeV ^{40}Ca, and 6–38 MeV ^{58}Ni ions. The ions were obtained by elastic scattering of the ^{252}Cf spontaneous fission fragments on appropriate scatterers. Simultaneous measurements of the detector response and the time-of-flight of a given ion made it possible to determine the PHD for every event. A very thin surface layer could account for the origin of the main part of the PHD [8].

In the optimal final version AVECS comprised a vacuum chamber with two time-of-flight tubes. At the end of each tube at the distance of 70 cm a mosaic of eight silicon detectors was placed. Thin 200–500 μg/cm^2 targets made of ^{238}U, ^{209}Bi, ^{197}Au, ^{184}W, ^{nat}Sm, or a spontaneously fissioning ^{252}Cf source could be placed in the centre of the chamber. The ^{252}Cf source was used for the energy and time calibration. One of the arms was fixed, the other could be turned in the horizontal plane coplanar to the proton beam. An independent time-start setup for time-of-flight measurements was located at the fixed arm close to the target. Experiments were performed in the proton beam of the PNPI synchrocyclotron with the energy of 1 GeV and intensity of (2–5)×10^{11} protons per second for two types of geometric configurations: "collinear" and "non-collinear". In the "collinear" configuration (Fig. 3) two massive complementary fragments were detected within narrow cones whose common axis was orthogonal to the proton beam direction. In the "non-collinear" configuration the movable arm was turned by 10° downstream. In both runs kinetic energies E_i and times of flight T_i of the fragments were measured simultaneously, it allowed us to determine their masses $M_i \sim E_i T_i^2$ and momenta $P_i \sim E_i T_i$. The latter, together with appropriate angular resolution, made it possible to study event-by-event the vector sum

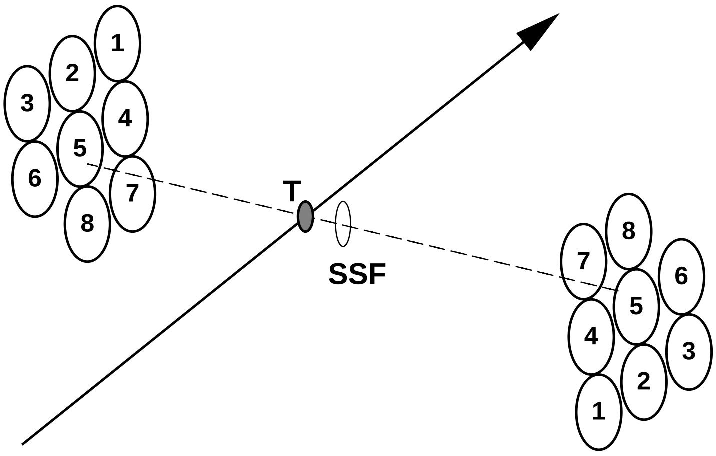

Fig. 3. Sketch of the AVECS device for $(2E, 2V)$ measurements. T – target, SSF – start signal film.

$$\vec{P} = \vec{P}_1 + \vec{P}_2, \tag{4}$$

where $\vec{P}_i$ are the fragment momenta. Momentum distributions were studied for different values of the nucleon losses, defined as a difference between the mass of the target nucleus M_0 and the sum of the measured fragment masses:

$$\Delta M = M_0 - (M_1 + M_2). \tag{5}$$

We selected the events of collinear configuration with $\Delta M \geq 75$ u (^{238}U target) which might correspond to disintegration of the nucleus into three fragments of comparable masses. Among the total of 2.2×10^4 events, 250 were found which satisfy the condition $\Delta M \geq 75$ u. Fig. 4 shows the momentum plot (P_1, P_2) for these events. Then the limitation on the kinetic energy of the third fragment was introduced as a deviation from the momentum balance ($P_1 = P_2$) of the two complementary collinear fragments. Events with $E_3 \leq 2$ MeV are shown by crosses in Fig. 4. The rest of the events had the third fragment moving but its kinetic energy was, as a rule, smaller than the energies of the two other fragments. Since the main interest of the investigation was a search for the events satisfying relation (2), 58 events with $P_1 = P_2$ were used to construct the mass plot (M_1, M_2) shown in Fig. 5. As the masses of the complementary fragments are widely dispersed, it is impossible to satisfy relation (2) for all the detected events. Only those shown by crosses in Fig. 5 may be described according to eq. (2).

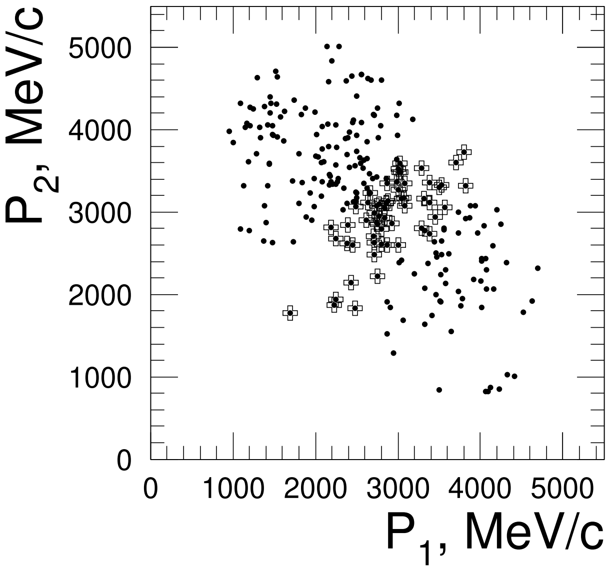

Fig. 4. (P_1, P_2)-plot for collinear disintegration with $\Delta M \geq 75$ u. Crosses – events with balanced momenta, $E_3 \leq 2$ MeV.

Collinear tripartition of heavy nuclei

Twenty six events shown by crosses in Fig. 5 can be used to evaluate the relative yield of the collinear tripartition of heavy nuclei compared with binary fission. For the reaction ^{238}U + p (1 GeV) the probability turned out to be $\geq 10^{-3}$ for $M_3 = \Delta M \geq 75$ u. This value is two times larger than $5 \cdot 10^{-4}$ found for the coplanar ternary fission in the same nuclear disintegration. The experimental search for the collinear tripartition in the spontaneous fission of ^{252}Cf gave an upper limit on the level of 10^{-5} for $M_3 = \Delta M \geq 32$ u. Thus we have to conclude that the collinear tripartition is a process which can be induced in heavy nuclei at intermediate excitation energies. It is a disintegration of the nucleus into two fission fragments together with a significant neck in between. The third inner fragment is formed by the double rupture of the neck. The momentum imbalance of the two detected fragments arises due to the momentum of the third inner fragment. Despite the possible large value of this momentum, the kinetic energy of the third fragment turns out to be lower, as a rule, than the energies of the detected fragments due to its large mass. In several cases this energy was found to be zero. This means that the third fragment is at rest, the two others being detected in the experiment as moving. These events look like usual binary fission of a heavy nucleus into two fragments. In order to get a tentative description of the collinear tripartition process it is necessary to determine the electric charges of all the three fragments which were not measured in the experiment. There are several possibilities to estimate the charges [9,10]. We used relation (2) for homogeneous matter distribution in the target nucleus to compose Table for a tentative description of the collinear tripartition in the reaction ^{238}U + p (1 GeV) [10].

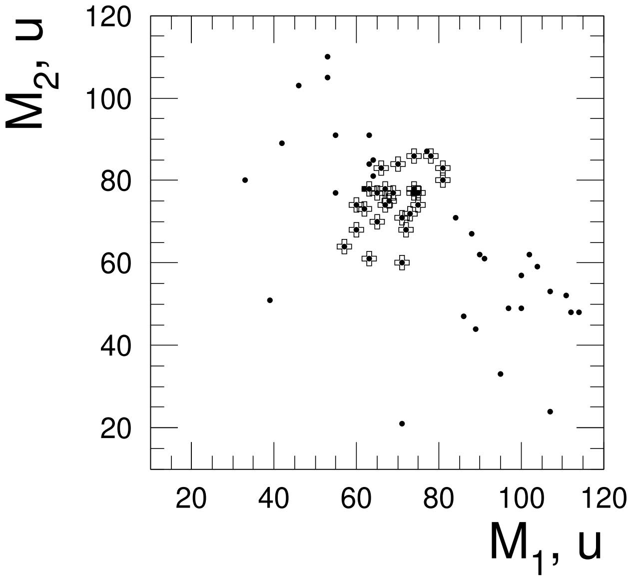

Fig. 5. (M_1, M_2)-plot for events with balanced momenta. Crosses – disintegration events for which the relation $M_1/M_2 = \sqrt{Z_2/Z_1}$ is fulfilled .

Table

Tentative nuclear reaction	Total kinetic energy of fragments, MeV	Initial distance D_3, fm
^{78}Rb + $^{75-\kappa}$Cr + ^{86}Ga + κ	186 ± 4	34.2 ± 0.7
^{67}As + $^{98-\kappa}$Ga + ^{74}Ni + κ	145 ± 3	46.9 ± 1.0
^{65}Ge + $^{104-\kappa}$As + ^{70}Co + κ	156 ± 3	43.9 ± 0.9
^{63}Co + $^{115-\kappa}$Kr + ^{61}Cu + κ	137 ± 3	50.6 ± 1.0
^{74}Ga + $^{87-\kappa}$Se + ^{78}Co + κ	161 ± 3	42.9 ± 0.8
^{81}Ge + $^{75-\kappa}$Cu + ^{83}Ga + κ	175 ± 3	38.2 ± 0.7
^{67}Ge + $^{94-\kappa}$Rb + ^{78}V + κ	154 ± 3	44.7 ± 0.9

A small uncertain value κ denotes several cascade nucleons compensating the momentum of the incident proton. In order to calculate the initial distance between the outer fragments in the last column of the Table one needs to know the Coulomb energy of the three collinearly located charges. Using eq. (2) one obtains

$$U_3 = \frac{e^2}{D_3}[Z_1 Z_2 + Z_3\,(\sqrt{Z_1} + \sqrt{Z_2})^2]. \tag{6}$$

Equating U_3 defined by this expression to the measured total kinetic energy of the three fragments, one can obtain the D_3 value. The obtained D_3 values for the collinear tripartition of the ^{238}U nucleus, being dispersed considerably, exceed the double inter-fragment distance for the binary fission of the same nucleus at the lower excitation energies.

In conclusion, we would like to note that disintegration processes with more than three charged particles one of which is at rest are also possible. Recently it was shown (P.Singer et al., Contribution to the 3rd Int. Conf. on Dynamics Aspects of Nuclear Fission, Casta–Papiernika, Slovak Republic, 1996) that ^{10}Be nuclei in a rather high excited state of 3.37 MeV were produced when three charged bodies were registered in coincidence. Part of the γ-rays observed at this energy seems not to be affected by Doppler broadening, which cannot be explained within any current picture of the dynamics of the fissioning system near scission. We may propose the following nuclear disintegration which satisfies the experimental observation:

$$^{252}_{98}\mathrm{Cf} \to\ ^{128}_{50}\mathrm{Sn} +^{10}_{4}\mathrm{Be}^* +^{10}_{4}\mathrm{Be} +^{101}_{40}\mathrm{Zr} + 3n.$$

An excited up to 3.37 MeV $^{10}_{4}$Be* nucleus remains at rest while the second $^{10}_{4}$Be nucleus is accelerated by mutual Coulomb repulsion together with two massive fragments.

References

[1] *B.A.Bochagov, A.P.Komar, G.E.Solyakin.* // Zh. Eksp. Teor. Fiz., 1960. V.38. P.1374.

[2] *B.A.Bochagov, A.P.Komar, G.E.Solyakin, V.I.Fadeev.* // Atomnaya Energiya, 1961. V.110. P.540.

[3] *B.A.Bochagov, A.P.Komar, G.E.Solyakin.* // Zh. Eksp. Teor. Fiz., 1962. V.43. P.1611.

[4] *B.A.Bochagov, S.S.Vasiliev, G.G.Semenchuk, G.E.Solyakin.* // Yad. Fiz., 1965. V.1. P.461.

[5] *A.P.Graevsky, G.E.Solyakin.* // Yad. Fiz., 1973. V.18. P.720.

[6] *A.I.Obukhov, G.E.Solyakin.* // Pis'ma Zh. Eksp. Teor. Fiz., 1993. V.57. P.73.

[7] *A.A.Zhdanov, V.I.Zakharov, A.V.Kravtsov, G.E.Solyakin, N.P.Filatov.* // Pis'ma Zh. Eksp. Teor. Fiz., 1991. V.54. P.311.

[8] *S.A.Kassirov, G.G.Kovshevny, A.A.Kotov, V.R.Resnik, G.E.Solyakin, N.K.Terent'ev.* // Nucl. Instr. Meth., 1974. V.119. P.301.

[9] *G.E.Solyakin, A.V.Kravtsov.* // Phys. Rev., 1996. V.C54. P.1798.

[10] *G.E.Solyakin.* // Yad. Fiz., 1997. V.60. P.35.