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WEAK K-HINDRANCE MANIFESTED IN ALPHA DECAY OF THE ^{178m2}Hf ISOMER

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Слабый K-запрет, обнаруженный при альфа-распаде изомера ^{178m2}Hf

Проведен эксперимент по наблюдению ветви альфа-эмиссии при распаде изомера ^{178m2}Hf и определен парциальный период полураспада, равный $(2,5 \pm 0,5) \cdot 10^{10}$ лет. Сделан вывод, что α-распад сильно задержан благодаря центробежному барьеру, возникающему из-за высокого спина данного изомера. Дополнительный анализ показывает, однако, что K-запрет в α-распаде проявляется относительно слабо, несмотря на сильное влияние спинового запрета.

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Weak K-Hindrance Manifested in Alpha Decay of the ^{178m2}Hf Isomer

An experiment has been performed to detect the alpha-emission mode in ^{178m2}Hf isomer decay and a partial half-life of $(2.5 \pm 0.5) \cdot 10^{10}$ years was measured. It was concluded that α decay is strongly retarded by the centrifugal barrier arising due to the high spin of this isomeric state. Additional analysis shows, however, that the K-hindrance in this α decay is relatively weak, despite the strong manifestation of spin hindrance.

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

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Electromagnetic decay of the noted 31-year-lived isomer ^{178m2}Hf has been studied extensively (see Ref. [1] and references therein). The level structure of this isotope was also examined using spectroscopic techniques in many reaction studies, such as those of Refs. [2, 3]. Until now, however, α decay of this isomer has not been observed.

A scheme of the process according to the tabulated data of Refs. [4, 5] is given in Fig. 1. A maximum energy release in the isomer α decay corresponds to the transition from the 16^+ isomeric level to the ground-state ($I^\pi = 0^+$) level in the daughter ^{174}Yb nucleus. The value of $Q_\alpha = 4.53$ MeV allows abundant α decay with a half-life, $T_{1/2,}$ on the order of days, much shorter than the real $T_{1/2}$ due to electromagnetic decay. However, the isomer-to-ground state α decay spans a 16-unit change in angular momentum and thus should be strongly suppressed by the centrifugal barrier. Additional structure hindrance may arise due to changes of the K quantum number as $\Delta K = 16$ for this transition. One can see in Fig. 1 that the total Q_α for decay of the ground and first isomeric ($m1$) states are relatively low and should correspond to very long α-decay half-lives according to the known systematics.

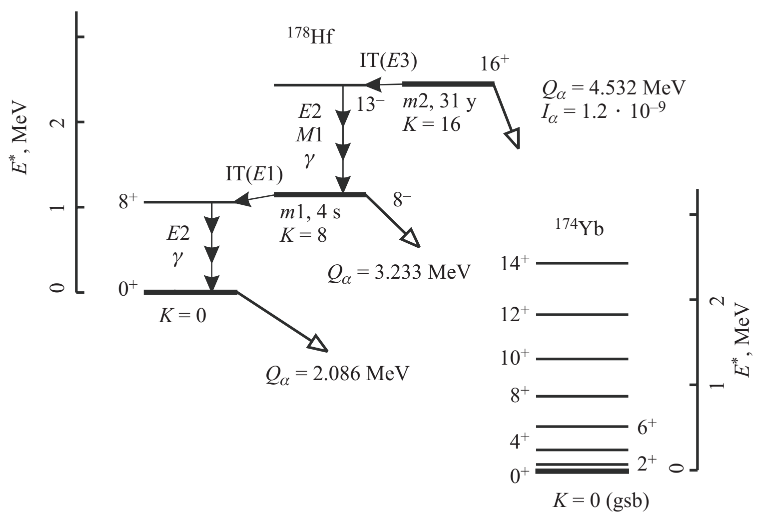

Fig. 1. Alpha-decay scheme for ^{178}Hf nuclei in the ground and isomeric states. The Q_α values were deduced from the nuclear mass tables [4] and the level energies and angular momenta from Ref. [5]. The presently measured absolute intensity is given for α decay of the $m2$ level

For mid-Z elements, alpha decay is typically observed for short-lived neutron-deficient isotopes. Considering elements ranging from Nd to Pb and isotopes thereof bounded by the magic numbers $N > 82$ and $Z \leqslant 82$, α-decay energies E_α to specific daughter states are known. In many cases the corresponding partial α-decay half-lives have been measured [5]. The hafnium isotopes lie in the center of this range of nuclides, being characterized by well-deformed axially symmetric prolate shapes, and it can be expected that semi-empirical systematics should apply to them.

The Geiger–Nuttall plot [6] is shown in Fig. 2 for even-Z nuclei, evidencing a nearly linear dependence of the log of the α-decay half-life $T^\alpha_{1/2}$ on the square root of Q_α. There is some curvature to the plots, but this is natural when an exponential function covers so many orders of magnitude. The systematic behavior seen in Fig. 2 is valuable for estimating the half-life of a nuclide for which Q_α is known, but $T^\alpha_{1/2}$ has not yet been measured. This is the initial basis for a prediction of the α-decay half-life of ^{178m2}Hf. The Geiger–Nuttall systematics, however, reflects well-allowed decays and does not account for significant structure or angular momentum hindrances. In the decay of ^{178m2}Hf, the high spin of the initial state should strongly influence $T^\alpha_{1/2}$.

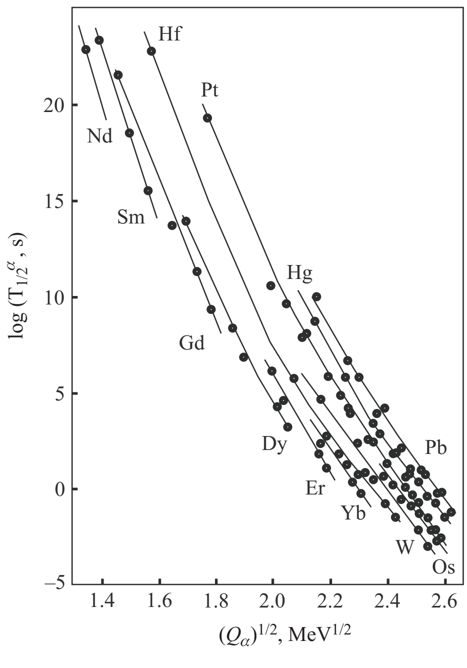

Fig. 2. The Geiger–Nuttall systematics of α-decay half-lives for nuclei in the range of $Z = 60-82$

Hindrance factors in α decay of odd-mass nuclei were discussed in Ref. [7], being defined as a ratio of the measured $T_{1/2}$ value to an expected magnitude that was found by referring to known values for α transitions without spin change, typically in neighboring even–even nuclei. Obviously, for simplicity this approach combines together hindrances arising due to different physical reasons. However, in principle, one may distinguish «macroscopic» and «spectroscopic» hindrances. The first hindrance arises in the case of particle emission with nonzero orbital momentum as reflected by a centrifugal barrier penetration factor. The second one reflects a structure hindrance due to the rearrangement of single-particle orbits and of the nuclear spin orientation.

For electromagnetic decay, structure hindrances are typically isolated in reference to the standard decay rate theoretically predicted for transitions of known energy and multipolarity. In deformed nuclei such «K-hindrances» were determined successfully for many transitions and the reduced hindrance factors were systematized. The latter parameter describes, in definition, the retardation factor reduced to one unit of the $(\Delta K - \lambda)$ value, where λ is a multipolarity of the transition.

In the present work, we extend the similar scheme to α decay. The ^{178m2}Hf α decay seems ideal for this development because it is characterized by strong changes of both spin I and K quantum numbers in α transitions to the yrast band of ^{174}Yb. Such transitions are selected as they possess the highest Q_α values (see Figs. 1 and 2). In general conception, our approach involves several steps: the use of the empirical systematics of Fig. 2 to estimate α-decay half-lives without hindrances, next construction of a systematic description for the spin hindrance originating from the centrifugal barrier, and then a comparison of the finally «predicted» half-life with the measured one. Via the latter comparison, the K-hindrance factor should be revealed.

A centrifugal barrier arises for any α transition with spin change. For isolation of a corresponding retardation factor, one has to examine experimental data on α decay for the case when the Kquantum number makes no effect. Recall that K is defined as the projection of vector I to the symmetry axis. It does not exist, for instance, in near-magic spherical nuclei and in nuclei with non-axial shapes. Thus, for evaluation of the spin hindrance, we assume that K does not exist at all and the α-decay rate for transitions between initial I_i and final I_f states may be expressed as:

$$R = \frac{N_{\rm at}\ln 2}{T^{\alpha}_{1/2}(Q^{if}_{\alpha})} \sum_{m=-I_f}^{I_f} \frac{1}{F(|I_i - m|)}, \tag{1}$$

where Q^{if}_{α} is the transition energy; m is the projection of $\mathbf{I}_f$ along the direction of $\mathbf{I}_i$; $N_{\rm at}$ is the number of decaying nuclei and $T^{\alpha}_{1/2}(Q^{if}_{\alpha})$ corresponds to the value from Fig. 2 that assumes zero-spin change and depends only on Q^{if}_{α}. The

$F(|I_i - m|) = F(\Delta I)$ is defined as the spin-hindrance factor and depends only on the difference in spins between initial and final states. If $I_i = 0$, the sum in Eq. (1) may be replaced by the spin volume factor $(2I_f + 1)$ divided by $F(I_f)$. For simplicity, the notation $F(\Delta I) = F(\ell)$ may be used.

Quantitative data exist in the literature on the relative intensities for branches of α decay for ground states of even–even nuclei that reach levels in the ground-state band of the daughter nuclide. The initial and final states are characterized by $K = 0$ and it was therefore possible to isolate the spin-hindrance factor at $\Delta I = I_f$. Data [5] for ^{230}Th and ^{238}Pu were analyzed using Eq. (1) and the corresponding Geiger–Nuttall curves for these elements. With this procedure, values of the spin-hindrance function $F(\ell)$ were characterized up to $\ell = 8$.

For α decay of ^{178m2}Hf, the spin difference may, in principle, be as high as ΔI = 16, so the $F(\ell)$ values extracted from Th and Pu data are insufficient. To extend $F(\ell)$ to higher spin differences, data was used for α decay of the high-spin isomers ^{211m}Po (25/2$^+$), ^{212m}Po (18^+) and ^{214m2}Rn (8^+). These nuclei demonstrate other cases of pure manifestation of spin hindrance in α decay without structure and K-hindrance factors. They are near-magic nuclei and the K quantum number does not exist at all, so their α decays proceed without structure retardation.

For each of these nuclei, a ratio was taken between the isomer's half-life for α decay giving a specific ΔI_m and the ground-state α-decay half-life for a specific ΔI_g. This ratio of half-lives was then related to the difference between the spin changes caused by the isomer and ground-state α decays to determine the corresponding $F(\ell)$ value. The difference between spin changes was not so large for the odd-A ^{211}Po nucleus: even though a branch of the isomer's α decay provides as much as $\Delta I = 12$, the ground-state α decay causes $\Delta I = 4$. However, for the even–even nuclei ^{212}Po and ^{214}Rn the spin difference is equal to the spin released in the isomer decay because there is no spin change in α decays of the respective ground states. In this manner, Eq. (1) was used to obtain $F(\ell)$ for ℓ up to 18, being new values, and for $\ell \leqslant 8$ to confirm those found earlier.

Figure 3 shows the extracted $F(\ell)$ function. The magnitude of this hindrance is very high for ℓ up to 16, of importance for the estimation of α decay of the $I^\pi = 16^+$ ^{178m2}Hf isomer. By analogy with standard practices for electromagnetic decay, one can introduce a reduced hindrance factor f for α decay according to $F(\ell) = f^\ell$, given in the top panel of Fig. 3; despite the scatter the points exhibit a more or less regular behavior that suggests a choice of $f = 6.8$ for high ℓ numbers. Thus, the necessary components are now available to obtain a reasonable estimate of the α-decay half-life of the ^{178m2}Hf isomer. Everything is based on empirical information, making reliable the below-estimated values.

The partial α-decay half-lives, $T_{1/2}^{\alpha,f}$, are given in the Table for the transitions from the $I_i^\pi = 16^+$ ^{178}Hf isomeric state to levels in the ground-state band of

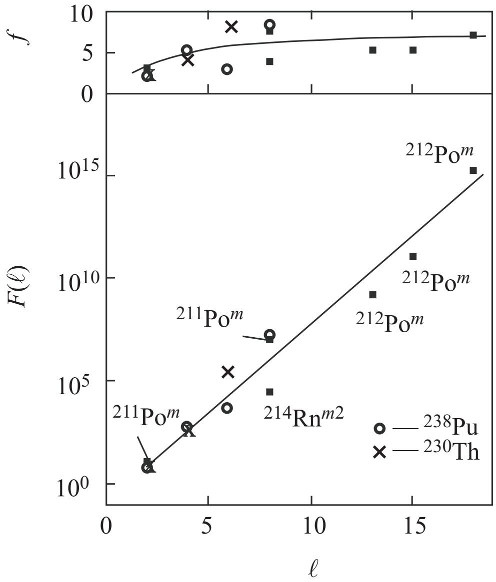

Fig. 3. Spin hindrance $F(\ell)$ versus angular momentum taken by the emitted α particle (bottom panel) and the reduced hindrance f (top panel). Three points marked as ^{212m}Po correspond to different branches of its α decay

^{174}Yb with $I_f^\pi = 0^+{-}14^+$. The parity selection rule is satisfied because all levels have positive parity and even I_i and I_f values. The values in the Table were obtained using Eq. (1) and in the summation the dominant contribution comes from the term corresponding to the minimum ΔI. This occurs because $F(\ell)$ is a very steep function, as shown in Fig. 3. The shortest half-life is expected for decay that reaches the 6^+ level of the daughter, giving the optimum product of $T_{1/2}^{\alpha,f}(Q_\alpha^{if})$ and $F(\ell)$. For higher I_f, the $F(\ell)$ function is drastically reduced while $T_{1/2}^{\alpha,f}(Q_\alpha^{if})$ increases greatly and the reverse happens for lower I_f. Using the partial half-lives, the total α-decay half-life is defined by a combination of partial ones to be $T_{1/2}^{\alpha} = 6.4 \cdot 10^7$ y. The α decay represents a low-intensity branch compared to electromagnetic decay of the isomer, thus requiring a sensitive measurement.

A source of ^{178m2}Hf containing about $3.5 \cdot 10^{13}$ atoms with isomeric nuclei was prepared about 10 years ago as part of a production series [8] carried out at the Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, using the ^{176}Yb(^{4}He, $2n$)^{178m2}Hf reaction. The hafnium fraction was chemically isolated from the enriched ^{176}Yb material after exposing it to a 36-MeV ^{4}He-ion beam produced by the U200 cyclotron. The hafnium fraction containing ^{178m2}Hf activity was deposited onto a Be foil and formed a hafnium oxide layer; no hafnium carrier was used in the chemical preparation. The thickness of the layer was small, definitely allowing small energy losses for transmission of α particles. The only hafnium in the material was that produced by nuclear reactions within the high-purity and highly-enriched ^{176}Yb target material. The purity of the hafnium material was guaranteed by the procedures described in Ref. [8], including neutron activation analysis.

Estimated partial half-lives for α decay of the ^{178m2}Hf isomer to levels in the ground-state band of ^{174}Yb. The calculations are discussed in the text

Transition $I_i \to I_f$	E_α, MeV	$T^{\alpha,f}_{1/2}$, y
$16^+ \to 0^+$	4.43	$8.6 \cdot 10^{10}$
$16^+ \to 2^+$	4.35	$3.0 \cdot 10^{9}$
$16^+ \to 4^+$	4.18	$3.4 \cdot 10^{8}$
$16^+ \to 6^+$	3.91	$1.2 \cdot 10^{8}$
$16^+ \to 8^+$	3.56	$2.8 \cdot 10^{8}$
$16^+ \to 10^+$	3.12	$2.7 \cdot 10^{9}$
$16^+ \to 12^+$	2.61	$7.2 \cdot 10^{10}$
$16^+ \to 14^+$	2.03	$5.5 \cdot 10^{13}$

The source included nuclei in the 8^- ^{178m1}Hf isomeric state in equilibrium, being at a level of about $4 \cdot 10^{-9}$ compared to the number of ^{178m2}Hf nuclei present. No significant contribution from $m1$ activity to α emission can be expected.

The first measurement was performed using a Si surface barrier (SSB) detector since this would provide spectroscopic information from any detected signal. The ^{178m2}Hf source was placed in a vacuum chamber at a distance of 10 mm in front of the SSB detector that had an active area with 14-mm diameter. The efficiency of the detector in this geometry was calibrated using a $^{\rm nat}$U sample and the energy resolution was found to be better than 50 keV using α lines of ^{234}U and ^{238}U. The ^{178m2}Hf source was kept for two weeks in the chamber and the chamber was pumped twice per week by a dry vacuum system. A spectrum of α particles was collected during this time and a background spectrum was also measured during a two-week period under identical conditions, but without the source.

Single events were observed in both spectra. An α-energy range from 2.0–4.5 MeV was selected so as to cover all branches listed in the Table for assumed α decays of ^{178m2}Hf to ^{174}Yb ground-state band levels. The total number of events occurring within the selected range was obtained for the «effect» spectrum (with source) and the «background» spectrum (without source). Subtracting the «background» number from the «effect» value gave $N_\alpha = (-17 \pm 25)$ as the number of α decays from the ^{178m2}Hf source during the two-week period. Correcting to the detector efficiency, one obtains an upper limit for α emission from the source at $\leqslant 1$ alpha per 3 hours. This corresponds to a lower limit for the α-decay half-life of ^{178m2}Hf of $6.6 \cdot 10^9$ y and a K-hindrance factor > 100.

No statistically-significant emission, above background, was detected within the entire range of energies from 2 to 9 MeV. The background level must be attributed to contamination of the vacuum chamber, etc., which were composed of regular technical materials like stainless steel. It was considered most productive to employ a more sensitive, albeit non-spectroscopic, detection method rather than to attempt an improvement of sensitivity with SSBs by increasing the acquisition time, installing multiple detectors or preparing a more purified vacuum chamber.

A method for low-background charged-particle detection has been known for decades based on the use of solid-state track detectors. This approach is well-developed and has been calibrated by many groups, including JINR, Dubna (see, for instance, Refs. [9, 10]). In the present experiment for α detection, CR-39 foils were used, being of special production for nuclear track detection by «Track Analysis Systems Ltd., UK». These detector-quality foils were produced from very pure materials and contain no α-active contaminants. Thus, in measurements with these foils the only source of background could be by penetration of radon gas. Track detector foils are provided with a clean polyethylene film stuck to the detector surface in order to exclude radon. The film is removed prior to use. During experiments, the foils must remain isolated from the surrounding air. Thus, the ^{178m2}Hf source was pressed between two clean CR-39 foils and carefully wrapped by plastic foil. Additionally the sandwich was sealed for months within a plastic box. It is known that plastic materials contain less contamination from U and Th (producing radon) and metal packing should not be used.

Tracks due to α particles appeared in the detector foil after its exposure to the source and after etching the foil to develop those tracks. The number of tracks was counted by visual registration using an optical microscope. This method can be described as high-efficiency and low-background. The foil facing the source serves as an α detector with integral efficiency of about 80% from 2π, according to the previous calibrations. The rear-positioned foil was useful mostly for isolation of the detection area from the backgrounds.

After 7-month's exposure to the source, the surfaces of the exposed CR-39 detector foils showed moderate damage. A rough spot was present on the foil past etching, seen directly by the naked eye in the region where the active material was placed. This diffuse damage was interpreted as being caused by a high flux of low-energy electrons emitted from the source. Tracks of α particles were nevertheless successfully identified and counted via the microscope, but it was decided to attempt a reduction of the electron-induced damage to the foil surface.

Additional series of studies were carried out in which detector foils of CR-39 were exposed for shorter periods of 1 and 3.5 months in contact with the hafnium source to reduce their exposure to electrons. The etching time was slightly shortened as well to minimize the development of the diffuse damage spot. Under these conditions the degree of damage on the surface of the detector foil due to electrons was significantly reduced. Tracks of α particles were observed as clear images and in accordance with their standard configuration. The total number of tracks within the area in contact with the active spot of the source was integrated and was consistent with the number found in the first experiment with CR-39 when normalized for time. The region in the facing foils away from the source, without exposure to α particles, was also examined to determine the level of background in the detector.

Complementary background measurements were performed with similar CR-39 foils kept in the same environment and placed in contact with various non-α-active foils including a Be foil similar to that which served as substrate for the hafnium source. A measurement was also made of the background without any material in contact with a detector. In all cases, the background track densities were statistically identical and also identical to the value obtained from the detector foil used with the hafnium sample (facing the sample), but away from the active spot.

The background track density was measured with good statistical accuracy and was then used to define the excess of counts due to the presence of the ^{178m2}Hf activity. The three exposure runs covered a total duration of about one year and the results were integrated to deduce an excess count rate due to α activity of the source. The total activity was deduced after account of the efficiency factor to be of 2.1 α/day. During the measurement period, the number of ^{178m2}Hf nuclei was about $2.8 \cdot 10^{13}$, so that the α-decay half-life was found to be

$$T^{\alpha}_{1/2} = (2.5 + 0.5) \cdot 10^{10} \text{ y}. \tag{2}$$

This value significantly improves the estimation of Ref. [11] where only an upper limit was indicated for the α-decay rate of ^{178m2}Hf corresponding to $T^{\alpha}_{1/2} > 6 \cdot 10^{8}$ y. The present measurement error exceeds a 10% statistical error since systematical errors could not be excluded, for instance, due to the uncertainty in the efficiency of detection. Spectral information on ^{178m2}Hf α decay is based only on the theoretical values given in the Table as the experiment did not allow groups to be distinguished in the α spectrum.

The measured $T^{\alpha}_{1/2}$ is larger than the estimated value by a factor of 390. One can interpret this as a manifestation of K-hindrance in α decay since this effect was not included in the calculations. A K-hindrance of only $4 \cdot 10^{2}$ is quite low for α decay of ^{178m2}Hf. For example, one of the dominant decay branches seen in the Table reaches the 6^+ level in ^{174}Yb via a transition with degree of K forbiddenness $\nu = (\Delta K - \Delta I) = 6$. In comparison, the corresponding spin hindrance $F(\ell = 6) \sim 10^5$ is seen in Fig. 3. This suggests that in reality K-hindrance is weakly manifested in α decay and the K quantum number plays a relatively small role.

A theoretical analysis of α-decay half-lives is given in Refs. [12, 13]. For the ground state of ^{178}Hf, $T^{\alpha}_{1/2} \sim 5 \cdot 10^{23}$ y was found [12], corresponding to $Q_\alpha = 2.08$ MeV. The measured value for the ^{178m2}Hf isomer is much shorter, $T^{\alpha}_{1/2} = 2.5 \cdot 10^{10}$ y. This is the sustained manifestation of the gain in Q_α value due to the 2.446-MeV excitation energy of the metastable state as shown in Fig. 1. The retardation of α decay by angular momentum creates a factor of many orders of magnitude and, after accounting for this effect, a relatively weak K-hindrance was deduced from the experimental $T^{\alpha}_{1/2}$ value.

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REFERENCES

1. *Smith M. B., Walker P. M., Ball G. C. et al.* // Phys. Rev. C. 2003. V. 68. P. 031302(R).

2. *Mullins S. M., Dracoulis G., Byrne A. P. et al.* // Phys. Lett. B. 1997. V. 393. P. 279; Ibid. V. 400. P. 401.

3. *Aprahamian A., de Haan R. C., Börner H. G. et al.* // Phys. Rev. C. 2002. V. 65. P. 031301(R).

4. *Möller P., Nix J. R., Myers W. D. et al.* // Atom. Nucl. Data Tables. 1995. V. 59. P. 185.

5. Table of Isotopes, Eighth Edition / Ed. by Firestone R. B., Shirley V. S., Baglin C. M. et al. N. Y.: John Wiley and Sons, 1996.

6. *Geiger H., Nuttall J. M.* // Phil. Mag. 1911. V. 22. P. 613.

7. *Schmorak M. R.* // Nucl. Data Sheets. 1980. V. 31. P. 283.

8. *Oganessian Yu. Ts., Karamian S. A., Gangrsky Yu. P. et al.* // Proc. of the Intern. School-Seminar «Heavy-Ion Physics», Dubna, Russia. Dubna: JINR, 1993. V. I. P. 365.

9. *Tretyakova S. P., Zamiatnin Yu. S., Kovantsev V. N. et al.* // Z. Phys. A. 1989. V. 333. P. 349.

10. *Karamian S. A., Grüner F., Assmann W. et. al.* // Nucl. Instr. Meth. B. V. 193. P. 144.

11. *Van Klinken J., Venema W. Z., Janssens R. V. F. et al.* // Nucl. Phys. A. V. 339. P. 189.

12. *Xu C., Ren Z.* // Phys. Rev. C. 2004. V. 69. P. 024614.

13. *Denisov V. Yu., Ikezoe H.* // Phys. Rev. C. 2005. V. 72. P. 064613.

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