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Systematic Study of Decay Properties of Heaviest Elements[1]

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Abstract—Decay properties and stability of heaviest nuclei with $Z \leq 132$ are studied within the macro-microscopical approach for nuclear ground-state masses. We use phenomenological relations for the half-lives with respect to α-decay, β-decay and spontaneous fission. Our calculations demonstrate that the β-stable isotopes ^{291}Cn and ^{293}Cn with a half-life of about 100 years are the longest-living superheavy nuclei located on the first island of stability. We found the second island of stability of superheavy nuclei in the region of $Z \approx 124$ and $N \approx 198$. It is separated from the "continent" by the "gulf" of short-living nuclei with half-lives shorted than 1 μs.

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INTRODUCTION

Great success was achieved during the last twenty years in the experimental study of reactions leading to superheavy nuclei, their decay properties and structure. Near-barrier fusion reactions have been used up to now for the production of new superheavy (SH) elements in the "cold" [1, 2] and "hot" [3] combinations of colliding nuclei. In the cold fusion reactions based on the closed shell target nuclei lead and bismuth the fusion cross-section decreases very rapidly with increasing charge and mass of projectile. In more asymmetric (and "hotter") fusion reactions of ^{48}Ca with actinide targets the cross sections were found to be much larger [3, 4]. However, californium is the heaviest available target, which has been used in these experiments for the production of element number 118 [5].

Thus, to get SH elements with $Z > 118$ in fusion reactions, one should proceed to heavier than ^{48}Ca projectiles (^{50}Ti, ^{54}Cr, etc.). The corresponding cross sections for production of the elements 119 and 120 are predicted to be smaller by two orders of magnitude [6] as compared with ^{48}Ca induced fusion reactions leading to formation of the elements 114–116. Another limitation of the fusion reactions (both "cold" and "hot") for producing superheavy elements consists in the fact that they lead to neutron deficient isotopes having rather short lifetimes. The most stable superheavies are predicted to be located in the region of neutron-rich nuclei, which is unreachable by fusion reactions with stable beams.

In fact, the predicted magic numbers, especially for protons, are quite different within different theoretical approaches. The magic number $Z = 114$ was predicted in the earliest macro-microscopic calculations [7, 8] and confirmed later in Refs. [9, 10]. Fully microscopic approaches predict the proton shell closure at $Z = 120$ [11], $Z = 126$ [12], or $Z = 114$, 120, 126 [13] depending on the model details. The neutron magic number $N = 184$ is almost constantly predicted by different theoretical models. Knowledge of the decay modes and half-lives of nuclei in a very wide range of neutron and proton numbers (nuclear map) is necessary for such predictions and planning of the corresponding experiments. Multi-nucleon transfer reactions might be used for the synthesis of long-living heavy neutron-rich nuclei located on the island of stability [14, 15]. For example, at the present level of the experimental technics development, the registration of superheavy recoil in the focal plane of detectors is possible only if its half-life is larger than 1 μs. This experimental limitation makes some regions of the nuclear map currently inaccessible to experimental studies.

The main decay modes of heavy and superheavy nuclei are: α-decay, β-decay, and spontaneous fission (SF). The α-decay half-lives are usually calculated within the Viola–Seaborg formula (see, e.g., [16–18]), with semi-empirical relations (e.g., [19]) or using the WKB method. The key quantity in all these methods is the energy of α-decay, the uncertainty of which mainly influences the accuracy of the calculations. In this paper we apply a variant of the Viola–Seaborg relation for α-decay half-life. The estimation of β-decay half-life is more complicated, especially for nuclei close to the line of β-stability, where the half-life is strongly sensitive to the structure of the mother and daughter nuclei as well as to the corresponding Q-value. Systematic study of the β-decay half-life was performed, e.g., in Refs. [20, 21] restricting the analysis to allowed β-transitions. Phenomenological relations for β half-lives such as the well known Sargent law [22, 23] or more recent parametrizations [24] could be also very useful. The most uncertain quantity is the half-life of SF, in particular, because of fewer experimental data

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points in comparison with α- and β-decays. The main two methods applied to calculate the SF half-life are the phenomenological one [25–27] and the more complicated dynamical approach [7, 8, 28–31].

As was said above, further researches in the region of superheavy nuclei require knowledge of the decay properties (half-lives and modes of decay) of individual nuclei and of the tendencies. This assumes that the main modes, i.e. α-decay, β-decay, and SF, have to be calculated simultaneously. This paper is aimed at the analysis of the decay properties of heavy and superheavy elements with respect to all three main decay modes. In Section 1 the details of the performed calculations are given. The main output of the calculations, i.e. the nuclear map containing the decay properties and half-lives of heavy and superheavy nuclei with $Z < 133$, is discussed in Section 2.

1. MODEL

1.1. Nuclear Masses

All the theoretical approaches used for the calculation of the nuclear masses as a function of collective degrees of freedom can be divided into three groups: macro-microscopical approach (see below), the generalized Thomas–Fermi method (e.g., [32]), and microscopic approaches based on the self-consistent Hartree–Fock model (both relativistic and nonrelativistic) (e.g., [33, 34]). The macro-microscopical approach to the calculation of nuclear masses is widely used giving as a rule similar or even better agreement with the experiments than various "more fundamental" microscopic approaches.

The macro-microscopical approach based on the assumption that the mass (depending on nucleon composition Z, N and deformation) can be obtained as a sum of two terms

$$M(Z, A, \text{def.}) = M_{\text{macro}}(Z, A, \text{def.}) + \delta U(Z, N, \text{def.}), \quad (1)$$

where the first term M_{macro} describes the gross dependence of the mass. The second term δU takes into account the influence of the shell structure on the mass. The main difference between the various macro-microscopical models consists in the choice of the mean-field potential entering the Hamiltonian. Three mean-field potentials usually used are: convoluted Yukawa potential (see [21, 35] and reference therein), Woods–Saxon potential [36–40], and two-center shell-model potential [41, 42].

All the calculations obtained in this paper are based on the values of the ground-state masses and because of that all predictions made below in experimentally unknown region are model dependent. Here we use the set of ground-state masses, obtained by Möller and collaborators [35]. This model is one of the most known and tested and that makes our prediction rather reliable.

1.2. Alpha-Decay

The α-decay is characterized by the energy release Q_α and the corresponding life time T_α. The value of Q_α is given by

$$Q_\alpha = M_{\text{gs}}(Z, A) - M_{\text{gs}}(Z-2, A-4) - M_\alpha, \quad (2)$$

where $M_\alpha = 2.424911$ MeV is the mass-excess of the α-particle.

The half-life for α-decay can be estimated quite accurately using the well-known Viola–Seaborg formula [43]

$$\log_{10} T_\alpha(\text{s}) = \frac{aZ + b}{\sqrt{Q_\alpha(\text{MeV})}} + cZ + d + h_{\log}, \quad (3)$$

where a, b, c, d, and $h_{\log}$ are adjustable parameters. We use the values of these parameters obtained in [16] $a = 1.66175$, $b = -8.5166$, $c = -0.20228$, $d = -33.9069$. The quantity $h_{\log}$ takes into account hindrance of α-decay for nuclei with odd neutron and/or proton numbers [43]

$$h_{\log} = \begin{cases} 0 & Z \text{ and } N \text{ are even,} \\ 0.772 & Z \text{ is odd and } N \text{ is even,} \\ 1.066 & Z \text{ is even and } N \text{ is odd,} \\ 1.114 & Z \text{ and } N \text{ are odd.} \end{cases} \quad (4)$$

Phenomenological calculation of T_α is the most justified (among other decay-modes) and the most accurate. The errors arising from uncertainty in Q_α is much larger then the one due to the inaccuracy of phenomenological Viola–Seaborg formula.

1.3. Beta-Decay

Because the decay properties of nuclei close to the β-stability line are mostly known (except in the region of superheavy nuclei) we may restrict ourselves to the case of nuclei far from the line of β-stability. It allows us to assume that the corresponding Q-value and the density of states are large enough that we can find a level in the daughter nucleus which is close to the ground state and which fulfils the conditions of allowed β-decays. Thus, the problem simplifies to the case of the ground-to-ground allowed β transitions. This assumption may be not accurate enough for some specific nuclei close to β-stability line, but this cannot alter the general trend in the decay modes.

In the description we will follow formulas and notations according to [23, 44, 45]. The life time with respect to all kinds of β processes T_β is given by $1/T_\beta = 1/T_{\beta^+} + 1/T_{\beta^-} + 1/T_{EC}$. In particular for the allowed β-decays one may use [46] with a small correction to the constant:

$$\log_{10}\left[f_0^b T_b(\text{s})\right] = 4.7. \quad (5)$$

Thus, the estimation of β-decay half-lives is reduced to the calculation of the Fermi function f_0^b.

The energy release in the case of β^- ground-state to ground-state decay is given by the mass difference

$$Q_{\beta^-} = M(A,Z) - M(A,Z+1), \qquad (6)$$

while for the β^+-decay it is:

$$Q_{\beta^+} = M(A,Z) - M(A,Z-1) - 2m_ec^2. \qquad (7)$$

In our calculations we agree with the experiment within two orders of magnitude for $T_\beta^{\exp} < 1000$ s. This is sufficient to estimate β-decay lifetime in competition with α-decay and spontaneous fission almost for every experimentally unknown nucleus.

1.4. Spontaneous Fission

The most realistic calculations of the SF half-life are based on the search for the least action path in a multidimensional deformation space. But this kind of analysis can be performed only in a rather restricted area of the nuclear map due to large calculation times. Thus, the preliminary estimation of the region of the nuclear map, where SF plays an important role is necessary. One of the first systematics of the SF half-life based on the fission barrier height and the value of the fissility parameter Z^2/A was proposed by Swiatecki [25]. After then many attempts were done to improve the SF half-life systematics (see, e.g., [26, 27]). Parameters of all of them were fitted to existing experimental data, which are very few, especially in the region of superheavy nuclei. Moreover, because of low statistics the experimental errors are often quite large in this region. It means that the prediction power of these systematics is under question for the superheavy nuclei. In order to predict the general trend of the SF half-life and to fix the regions where as in SF plays a significant role we propose to employ the same idea [25] that the half-life is mainly determined by the barrier height in potential energy surface. To determine the coefficients of the systematics we include in the fitting procedure not only the experimental data [47–49] but also the realistic theoretical predictions [28, 29] for the region $100 \le Z \le 120$ and $140 \le N \le 190$. Thus we get:

$$\begin{aligned}\log_{10} T_{SF}(\mathrm{s}) &= 1146.44 - 75.3153Z^2/A + 1.63792 \\ &\times \left(Z^2/A\right)^2 - 0.0119827\left(Z^2/A\right)^3 \qquad (8a)\\ &+ B_f\left(7.23613 - 0.0947022Z^2/A\right) + \delta;\end{aligned}$$

$$\delta = \begin{cases} 0 & Z \text{ and } N \text{ are even,} \\ 1.53897 & A \text{ is odd,} \\ 0.80822 & Z \text{ and } N \text{ are odd.}\end{cases} \qquad (8b)$$

In (8) B_f is the fission barrier, which according to the topographical theorem [50, 51] is calculated as a difference between the liquid-drop barrier $B_f(LDM)$ [52] and the ground-state shell correction $\delta U(g.s.)$, i.e. $B_f = B_f(LDM) - \delta U(g.s.)$. Figure 1 shows the dependence of the SF half-life on the neutron numbers for the nuclei with even atomic numbers from uranium to flerovium. Obviously Eq. (8) qualitatively reproduces the behavior of the half-lives in the experimentally known region. However the proposed relation substantially underestimates the abrupt decrease of the half-life for californium, fermium, and nobelium around $N = 160$. In the region of superheavy nuclei we get reasonable agreement with the data. In Fig. 1 we also show the calculations of Ref. [28] for the isotopes of $Z = 104$–114. It is seen that in this region both models give similar results for those nuclei, for which experimental data exist. Note that the model of Ref. [28] predicts for some nuclei a too abrupt decrease of the half-lives around $N \simeq 170$ and much larger ones around the closed shell $N = 184$.

2. STABILITY OF HEAVY NUCLEI

Figure 2 shows the nuclear map for the total half-life and decay mode for nuclei with $Z \le 132$. The known nuclei are situated along the β-stability line with a shift to the proton-rich region especially for heavy and superheavy nuclei. Almost all proton-rich nuclei with $Z \le 118$ having reasonable for experimental identification half-lives are already synthesized. The circles in Fig. 2 correspond to the nuclei with $Z = 119$–124, which may be obtained in 3n channel of the fusion reactions: ^{50}Ti + ^{249}Bk, ^{50}Ti + ^{249}Cf, ^{54}Cr + ^{248}Cm, ^{54}Cr + ^{249}Bk, ^{54}Cr + ^{249}Cf, ^{58}Fe + ^{248}Cm, ^{58}Fe + ^{249}Bk, and ^{58}Fe + ^{249}Cf. The synthesis cross section of these new superheavy nuclei with $Z > 118$ in fusion reactions is predicted to decrease substantially due to the change of the projectile from ^{48}Ca to a heavier one [6]. Moreover, it is seen (see Fig. 2) that the nuclei being synthesized in these reactions have the half-lives on the order of 1 μs. It means that the nuclei heavier than the element 118 even being synthesized could be hardly detected because of their very short half-lives. This conclusion is model dependent. As was stated above, these calculations are based on the ground-state masses.

Figure 3 shows how the 1 μs contour depends on the model used for the nuclear masses calculations. The contour in Fig. 3a is obtained with the nuclear masses calculated by Möller and collaborators [35], while Fig. 3b was calculated here within the two-center shell model. Both models give a quite similar prediction of the half-lives for the nuclei which can be synthesized in the above written projectile-target combinations. At the same time the border of 1 μs on the neutron-rich side differs substantially within these two models for nuclear masses. Such a discrepancy appears due to the extrapolation of the models parameters to unknown regions, while the results for experimentally studied nuclei are quite similar. Discovery of other "accessible" proton-rich nuclei is certainly of interest. However, in our opinion, the most challenging region for future studies is the region of more heavy and more neutron-rich nuclei. One of such interesting

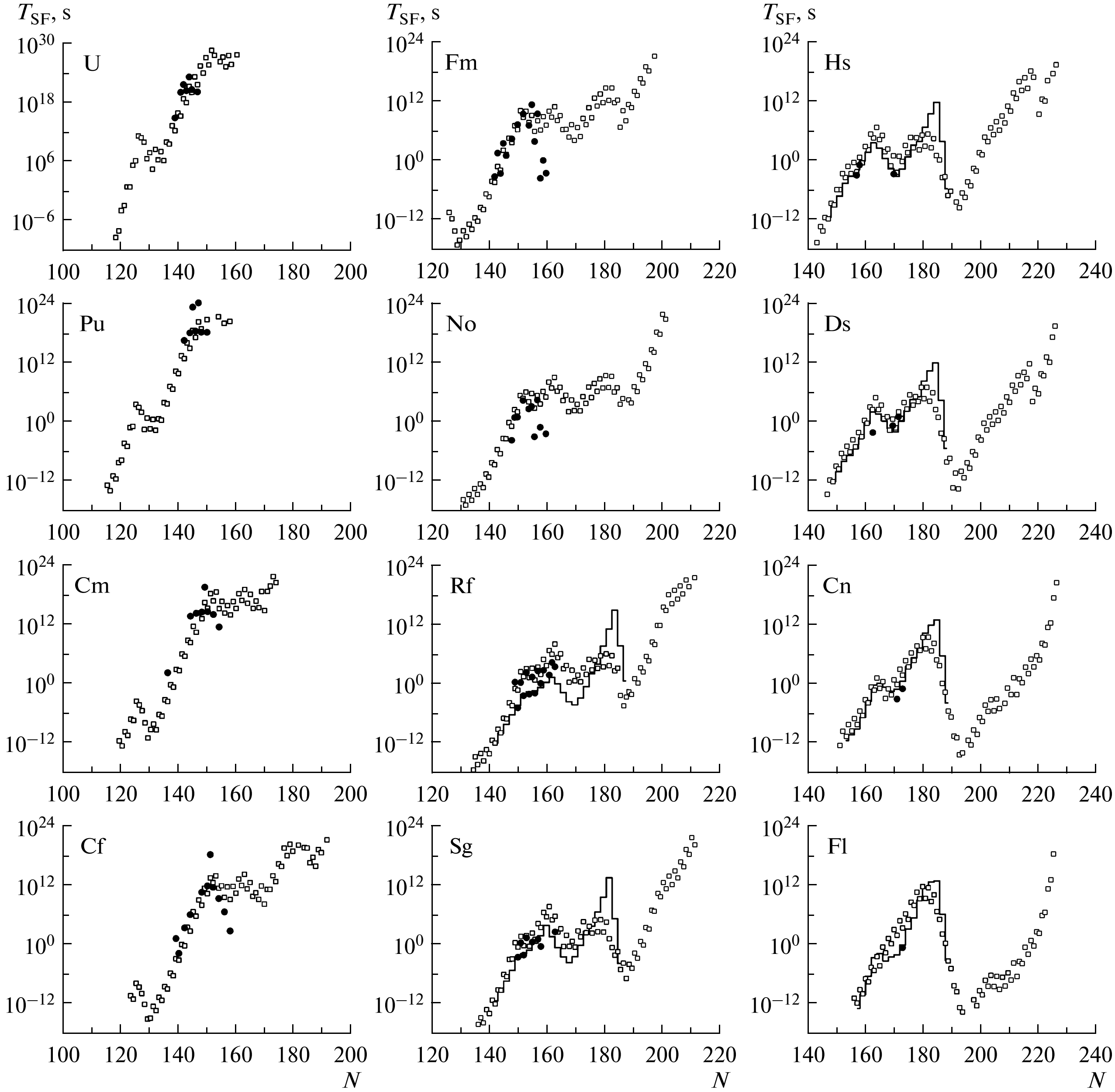

Fig. 1. Dependence of the SF half-life on the neutron number for the isotopes from U to Fl. The white squares are the estimation by the phenomenological formula (8), the black dots are the experimental data [47–49], and the full lines are the calculations of Ref. [28].

regions is the island of stability of superheavy nuclei centered at $Z \sim 114$ and $N \sim 184$. According to our predictions the most long-living nuclei in this area are the β-stable isotopes of copernicium ^{291}Cn and ^{293}Cn with the half-lives of about 100 years. The main decay mode of ^{291}Cn is predicted to be SF and ^{293}Cn decays by α-decay and SF with nearly equal probabilities. These isotopes—if synthesized—could be accumulated because of the relatively large lifetime. Unfortunately these two isotopes are presently unreachable directly by fusion reaction. However, one hypothetical way to produce ^{291}Cn is the triple β^+ (or EC) decay of ^{291}115 which in turn could be, for example, synthesized after α-decay of ^{295}117 in the reaction $^{48}\text{Ca} + {}^{249}\text{Bk} \rightarrow {}^{295}117 + 2\text{n}$. Recently [4] the element 117 has been discovered in 3n and 4n evaporation channels of the fusion reaction ^{48}Ca + ^{249}Bk with the cross section ~1 pb. In total 6 events were registered. This means that such a method hopefully may be realized in future with the progress in experimental technologies.

It is known that some of the dubnium isotopes (i.e. ^{270}Db) are expected to be β^+-decaying, while only

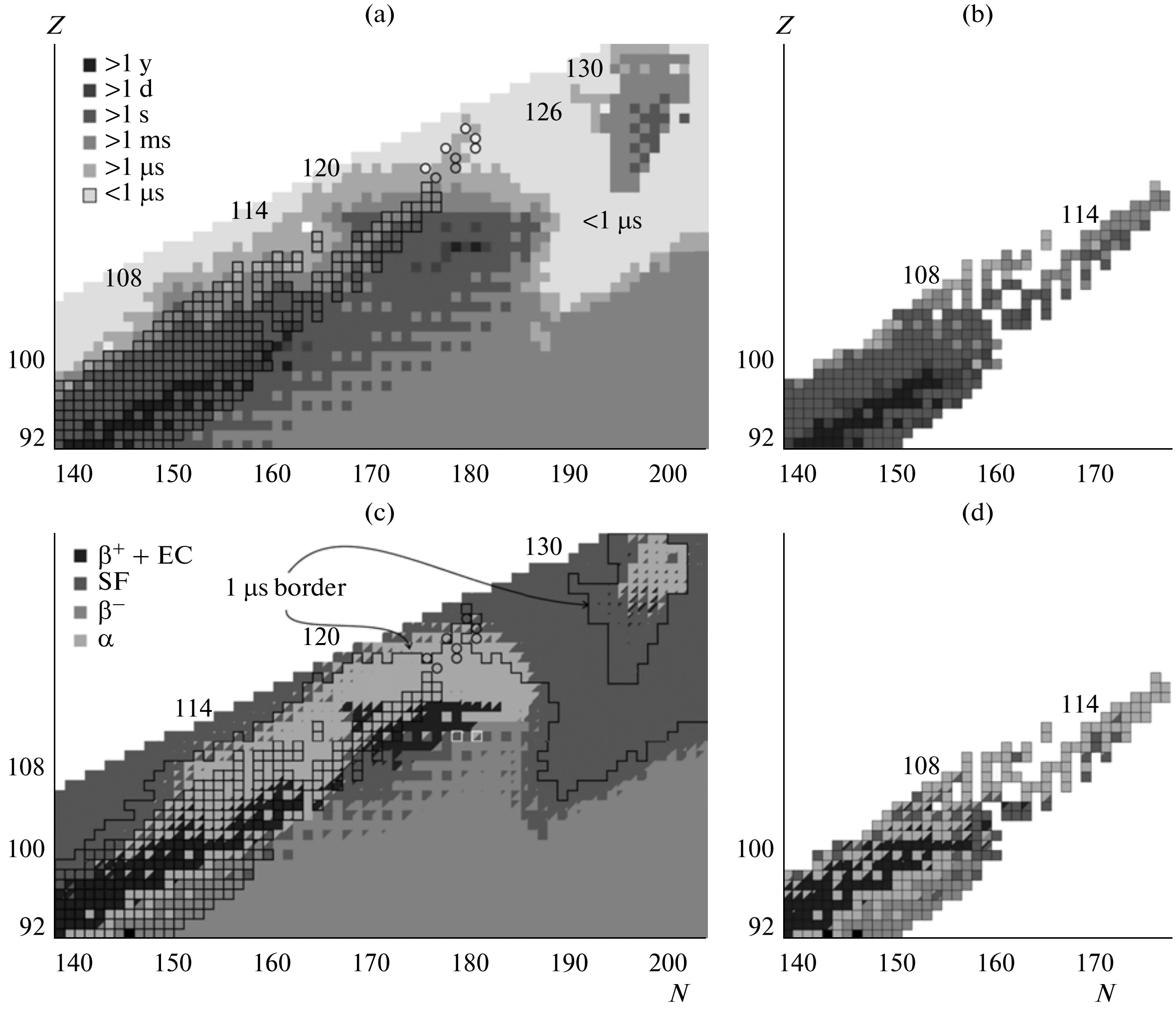

Fig. 2. Upper part of the nuclear map reflecting the total half-lives (a), (b) and the decay modes (c), (d). The left panels (a), (c) show the calculations and the right panels (b), (d)—the experimental data taken from [47–49]. The contour lines on the left bottom panel correspond to the border of 1 μs half-life. The circles show the nuclei with $Z = 119$–124, which may be synthesized in *3n* channel of fusion reactions ^{50}Ti + ^{249}Bk, ^{249}Cf and ^{54}Cr, ^{58}Fe + ^{248}Cm, ^{249}Bk, ^{249}Cf (see the text). The bounded cells correspond to the experimentally known nuclei. The bounded nuclei with the white colour border (c) are the most stable copernicium isotopes ^{291}Cn and ^{293}Cn (see the text).

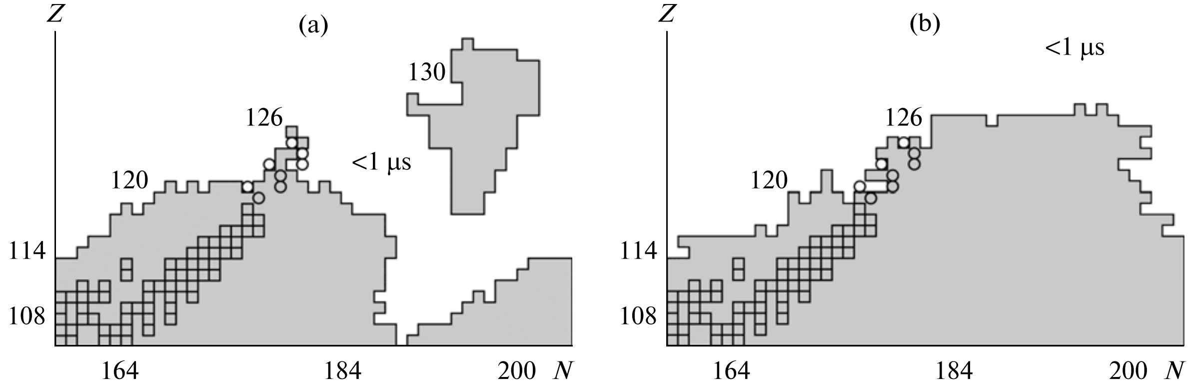

Fig. 3. The contour of 1 μs half-life of superheavy neclei obtained with the ground-state masses from Ref. [35] (a) and the two-center shell model (b). The squares show the known nuclei and the circles correspond to the nuclei with $Z = 119$–124 which may be synthesized in 3*n* channel of fusion reactions ^{50}Ti + ^{249}Bk; ^{249}Cf and ^{54}Cr; ^{58}Fe+ ^{248}Cm; ^{249}Bk; ^{249}Cf (see the text).

the SF mode was yet observed. The corresponding experiments are planned for the near future in order to verify (or not) this prediction. One can see that not only the nuclei around dubnium isotopes, but also the neutron-rich isotopes of superheavy elements with $111 \leq Z \leq 115$ nearest to the synthesized recently in Dubna in the ^{48}Ca-induced fusion reactions undergo β^+-decay. It means that their experimental identification (if they will be synthesized) may be significantly complicated. On the other hand, our calculations of β-decay half-lives are based on the assumption of allowed β-transitions. As was said above, β-decay can be substantially suppressed, especially for nuclei close to the β-stability line (i.e. having small values of Q-decays). It means that some of the nuclei found here to have β^+-decay as the main mode, may have a much longer β-decay time, and the main decay mode could be α-decay. However, the gross tendency (i.e. existence of the region of β^+-decaying nuclei) will remain. To perform a more reliable calculation of T_β for a specific nucleus the structure of the mother and daughter nuclei are required.

Another interesting region of nuclei (see Fig. 2) is situated in between $120 \leq Z \leq 127$ and $197 \leq N \leq 200$. It forms the next "island of stability" for superheavy nuclei. It is well-separated from the known island of stability around $Z = 114$, $N = 184$ by the "gulf" caused by SF and α-decay, where the half-lives do not exceed 1 μs. The nuclei within the new predicted island of stability have half-lives of about 1 s. Their main decay modes are α-decay and SF. Of course, the prediction of this and other islands of stability is strongly model-dependent. The main reason of existence of the found island is the increase in SF half-life. This result is based on a rather simple phenomenological model for the SF process. In order to verify these predictions more fundamental calculations should be made taking into account dynamical effects and the multidimensional nature of the fission process. The possibility of experimental study of this region of the nuclear map is rather unclear. At least these nuclei are not accessible by fusion reactions.

CONCLUSIONS

We have calculated the nuclear decay properties (the decay modes and half-lives) of nuclei with $Z \leq 132$. The calculations are based on phenomenological relations for the decay times as well as on the ground-state properties according to the macro-microscopic model [35] and those calculated within the two-center shell model. The main decay modes were taken into account, namely α-decay, β^--decay, β^+-decay, electron capture, and SF. For SF we proposed a phenomenological formula with the parameters defined from the fit to experimental data and the results of previous realistic calculations in the superheavy mass region [28, 29]. We found that the island of stability of superheavy nuclei is centered at β-stable copernicium isotopes ^{291}Cn and ^{293}Cn having a half-life of about 100 years. Because of such a short half-life, the search in nature of superheavy nuclei may be performed in cosmic rays. At existing experimental facilities the synthesis and detection of nuclei with $Z > 120$ produced in fusion reactions may be difficult (or impossible) due to their short half-lives (shorter than 1 μs). The found area of β^+-decaying nuclei with $111 \leq Z \leq 115$ to the right of those synthesized in the fusion reactions with the ^{48}Ca beam may significantly complicate their experimental discovery. Hopefully in this region the α-decay dominates for the nuclei already discovered in hot fusion reactions. We found a second island of stability of superheavy elements at $Z \sim 124$, $N \sim 198$ with the maximum half-lives ~ 1 s. It is separated from the "continent" by the "gulf formed by nuclei with half-lives shorter than 1 μs. However, the existence of the second island of stability is model dependent. A more detailed analysis of decay properties of nuclei in this region is required especially for the SF process.

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