Schottky-like phase transition in the fission of atomic nuclei: ^{235}U

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Background: The physics of fission and the features of the fission fragment yields—symmetric or asymmetric shapes for different atomic nuclei and their energies of excitation or temperatures T-are among the "popular puzzles" of nuclear physics. Explanation of these features requires understanding both the nature of the interaction between the nucleons and the thermodynamics of nuclear matter transformations. The new statistical method for investigation of the ordering of the postscission ensemble of fission fragments is also essential.
Purpose: The goal of this article is to demonstrate the possibility of a new type of phase transition in nuclear fission within the fixed temperature range when there is a change in the shape of fission fragment yields from asymmetric to symmetric. The temperature dependence of the thermodynamic functions indicates a Schottky-like phase transition, known from solid-state physics.
Methods: We used the proposed statistical method based on the study of thermodynamic ordering for a postscission ensemble of fission fragments. This method allows us to investigate the temperature evolution of the yields (both mass and charge) and the features of the change of the thermodynamic functions of the ensemble of the fragments of fission. The isotope ^{235}U for which the data of the fission fragment yields are well known was chosen for the numerical investigations.
Results: We have found anomalies of the thermodynamic functions of the ensemble of fragments of fission of ^{235}U in the temperature range when the shape of the yields of fragment fission changes from asymmetric to symmetric. In particular, the peaklike form of the heat capacity $C(T)$ indicates a Schottky-like phase transition. We also point out the experimental possibility of observing such a phase transition within the nuclear temperature range of 1–2 MeV.
Conclusions: This article shows that a new Schottky-like phase transition can be observed under nuclear fission. It differs from known phase-type transformations under nuclear fission, which have been intensively investigated lately and may be due to fundamental factors such as loss of statistical nonequivalence or the identity of nucleons in different fission fragments.

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I. INTRODUCTION

The investigation of the mass-charge spectra of fission fragments (MCSFF) of atomic nuclei is among the "popular puzzles," since it concerns the universal but ordinary effects for which there should be a clear explanation. Among the features of the MCSFF are the following:

(i) asymmetric (two- or three-humped) or symmetric (one-humped) shapes of the fission fragment yields;
(ii) shapes or topology of the MCSFF, which are determined by the formula/composition of the original/source nuclei (i.e., by the number of their protons and neutrons) [1];
(iii) the MCSFF dependence on the excitation energy or the nuclear temperature, T, for the nucleus under fission; for most of the atomic nuclei, the increase of T leads to the symmetric shapes of the fission fragment yields.

The last feature is related to the thermodynamics of nuclear fission, the redistribution of different fission fragment yields due to the effects of quantum, statistical, and thermal processes. Their study is essential for understanding the nature of the stability and the features of the nuclear matter transformation. We note the importance of studying the fission fragment yield for applied purposes. Thus, nuclear fission is the base of modern power engineering and military; fragments of fission of heavy nuclei are the resource that forms the isotopic and chemical composition of cosmic bodies and our planet [2]. Recent work on the study of fission fragments is related to the physics of radioactive ion beams [3] and achievements in the field of nucleosynthesis [4] as well as some practical applications in nuclear medicine and radioecology. A number of theoretical approaches treat the problem of nuclear fission. An analysis of these approaches is given in Ref. [5], which

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contains a detailed analysis of existing theoretical approaches and innovations (see also Refs. [6–8]). Conditionally, two directions for such studies can be classified as the pre- and postscission approaches. In the first of them, based on a model of the nucleus as a liquid drop, the deformation of which leads to its scission, the prescission processes are investigated. The evolution of this approach is demonstrated in different approximations (see Ref. [5]), e.g., statistical scission-point models, statistical saddle-point models, Strutinsky-type calculations of the potential-energy landscape, taking into account the dynamics of fission and stochastic effects from the saddle point to the scission point, and others. The statistical method associated with P. Fong [9] considers the statistical equilibrium between the fission fragments at the scission point, when the available phase space of the system of the two separated fragments completely determines everything. Promising are the *ab initio* MCSFF calculations that harmonize the micro- and macroscopic methods in the theory of nuclear fission. Despite the considerable progress made by using these methods, they require using data obtained from the compilation of experimental results or introducing some adjusting constants, which are generally not substantiated: from a special form of density of state for nucleons to introducing parameters characterizing a surface tension of the nucleus, a viscosity [10], and so on. As a rule, this limits the use of these theories to only selected classes of nuclei of the even-even type or the even-odd type. There are also the problems in studying the evolution of the fissioned system's parameters with the temperature T's increase or decrease within the framework of these approaches. The postscission approach has been implemented in the scission point yields (SPY) approximation [11], and investigations have been carried out when clusters with fission fragments have been realized and formed at the point of scission. In this case, one can study the results of the scission processes and may be able investigate statistical ordering of the ensemble of such clusters. This provides a method to determine the MCSFF and other observables of fission. Such an approach is essentially free from using the adjusting constants, because, for example, the nucleus temperature, T, can be determined from the experiment [5]. The binding energies of the nucleus fragments are still tabulated in numerous bases of nuclear-physical constants [12,13]. Such theories, using the thermodynamic method, have greater capacity to investigate different classes of nuclei, i.e., both light (preactinide) and super heavy nuclei [14,15]. In this work, the possibility of using the proposed approach to study the temperature behavior of thermodynamic parameters of a postscission ensemble of nuclear fragments is shown. In particular, such a calculation has been carried out on the ^{235}U isotope, demonstrating the possibility of a new type of phase transition in the fission of nuclei.

II. THEORY

The proposed method of MCSFF determination was described earlier in Ref. [16] and is based on the following assumptions.

(i) Characteristics of the MCSFF are determined from the condition of statistical ordering of the canonical ensemble that contains nuclei-fragment clusters that can be realized in the fission of the original nucleus with the atomic mass A_0 and the charge Z_0.

(ii) The ensemble of fragments of nuclear fission is a sort of constant pressure ensemble; its thermodynamic parameters pressure, P, and temperature, T, are determined by the state of the original nucleus, which can be considered as the heat bath.

(iii) The proposed theory uses a color statistics that takes into account the statistical nonequivalence of nucleons even of the same sort as protons and neutrons in the fission fragments with different binding energies. These peculiarities lead to finding the configuration of entropy, S, for the nuclear clusters containing two fission fragments.

The parameters of the equilibrium state of the two-fragment cluster ensemble can be obtained from the condition of the minimum of the Gibbs thermodynamic potential [17]:

$$G = U - TS + PV, \tag{1}$$

where PV is the work under the constant pressure. The U values are determined by the binding energy of the two-fragment cluster; its discrete spectrum $\{\varepsilon_i(V)\}$ is an additive quantity to the binding energy of the jth nucleus fragment from the ith cluster and has negative values corresponding to the bound states of the nucleons:

$$\varepsilon_i = \sum_{j=1,2} \sum_{\langle N_p \rangle_i} \sum_{\langle N_n \rangle_i} U_j(A_{j,i}, Z_{j,i}). \tag{2}$$

The symbol $\langle \cdots \rangle$ means that the summation in Eq. (2) is taken over the numbers of protons and neutrons, $N^p_{j,i}$ and $N^n_{j,i}$, satisfying the following condition:

$$\sum_{j=1,2} \left(N^p_{j,i} + N^n_{j,i} + n_{i,j} \right) = A_0, \quad \sum_{j=1,2} N^p_{j,i} = Z_0,$$

where $n_{i,j}$ is the number of fission neutrons. The configurational entropy S in Eq. (1),

$$S = \ln(\omega_\mathrm{i}), \tag{3}$$

is calculated through the degeneracy factor ω_i that takes into account the statistical nonequivalence of nucleons with different specific binding energies in the fission fragments: the degeneracy factor is

$$w_i = A_0!/\left[\prod_{j=1,2} \left(N_p^{(j)}! N_n^{(j)}! K(n_{i,j}) \right) \right], \tag{4}$$

where $K(n_{i,j}) = 1/n_{i,j}!$ and $\prod_{j=1,2} x_\mathrm{j}! = x_1!\, x_2!$. From Eq. (4), one can see that the entropy term in Eq. (3) reaches maximum if $N_p^j = N_n^j$ and is responsible for the symmetrization of the fission yields with the nuclear temperature T increase. For the matching of thermodynamic quantities and statistical averages, it is necessary to write down the probability of realization, for example, of the two-fragment cluster through the isobaric distribution function:

$$f_i(V) = \omega_i \exp\{-(\varepsilon_i + PV)/T\}/Z_p. \tag{5}$$

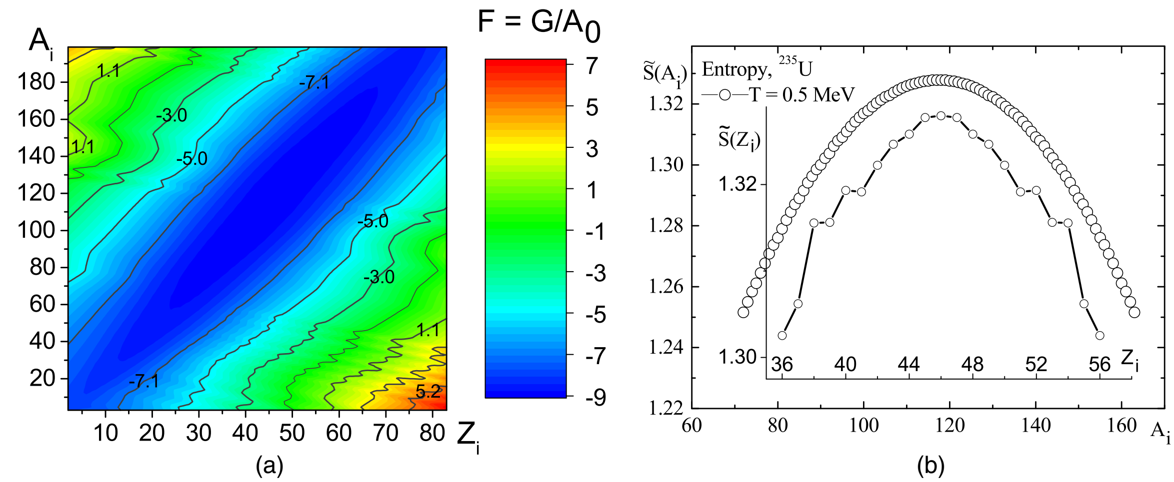

FIG. 1. (a) The concentration dependence of the Gibbs potential (1), $F = G/A_0$, and (b) the configuration entropy $\tilde{S} = S/A_0$ dependence on the nucleus mass-charge ratio for the ensemble of fission fragments. The insert in panel (b) shows the dependence $S(Z)$; the calculation is made for the ensemble of nuclear fissions of ^{235}U at $T = 0.5$ MeV.

Here Z_p is the partition function determined by the normalization rule:

$$Z_p = \sum_{k,\,V} \omega_k \exp\{-(\varepsilon_k + PV)/T\}. \tag{6}$$

The next step is going from the probability of the ith nuclear cluster formation to the distribution function $F(A_i)/F(Z_i)$ or the probability of the yield of a single fission fragment with the mass A_i or the charge Z_i. The procedure of calculating $F_i(A_i)$ or $F_i(Z_i)$ is described in Ref. [16]; the above quantities have the following normalization rules:

$$\sum_{\langle A_1\rangle} F(A_1) = \sum_{\langle Z_1\rangle} F(Z_1) = 200\%,$$

where $\langle A_1\rangle$ and $\langle Z_1\rangle$ have the same meaning as in Eq. (2). The calculated values of $F(A_1)$ make it possible to find the statistical averages as thermodynamic quantities important for studying the temperature characteristics of the scission or separation of the atomic nucleus.

III. RESULTS AND DISCUSSION

Calculations were made on the ^{235}U isotope for which there is a significant amount of data on the fission fragment yields. In this article, we investigate the equilibrium condition of the ensemble of fission fragments up to the preneutron emission; their discrete spectrum $\{\varepsilon_i(V)\}$ is calculated using the well-known mass-formula databases tabulated in recent reviews [12,13]. The nuclear temperature T appearing in Eq. (1) can be estimated by the emission spectra of the fission neutrons and protons [18], but in this article, it is used as a parameter of the theory within the 0.5–7 MeV range.

Figure 1 shows the results of calculation of the thermodynamic parameters of the ensemble of nuclear clusters containing fission fragments with the atomic mass A_i or the charge Z_i. The above calculations were carried out for $T =$ 0.5 MeV or under conditions of "cold" fission of ^{235}U. The last requirement condition can be realized, for example, in the case of spontaneous fission of ^{235}U. This calculation shows the existence of sets $\{A_i, Z_i\}$ at which the minimum of the thermodynamic potential of Gibbs (1) [Fig. 1(a)] is realized. These sets have a high probability of formation with the fission of ^{235}U and dominate in its MCSFF. Figure 1(b) demonstrates the concentration dependence of the configuration entropy $S(A)/S(Z)$, which reaches a maximum at $A_i \sim 117-118$ and $Z_i = 42$ and is responsible for the symmetrization of the MCSFF of ^{235}U [Fig. 1(b)]. As is seen from the $S(Z)$ dependence, there is a fine structure in the vicinity of the magic number $Z = 50$ [see insert in Fig. 1(b)]. Such features are not observed for the $S(A)$ dependencies, where the magic numbers 50 and 82 correspond to the atomic masses of nuclei 104 and 131, respectively. Such a situation may be due to the higher sensitivity of the entropy S to the Z values because of the inequality $Z < A$ that holds for each nucleus. Figure 2 shows the temperature dependence of MCSFF for the 0.5–7 MeV temperature range in the case of ^{235}U fission obtained from the most probable nuclei sets with the mass A_i [Fig. 2(a)] or the charge Z_i [Fig. 2(b)]. It was found that taking into account the cumulative effects does not significantly affect the MCSFF dependence. It can be seen that, in the case of a "cold" ^{235}U fission within the 0.5–1 MeV temperature interval, the calculation shows the anisotropy and too sharp peaks for the mass spectra centered on 104 and 132. For the charge spectra [Fig. 2(b)], these peaks are centered on 42 and 50 and are formed by the nuclear clusters containing $\{^{104-107}$Mo, $^{131-128}$Sn$\}$, $\{^{102,103}$Nb, ^{133,132}Sb$\}$, and other isotopes. Note that similar results on the significant anisotropy of the mass spectra were obtained within the framework of the SPY model [11]. There are several explanations for such behavior, from the ability of the apparatus to consider short and ultrashort fission fragments [19] to the need to study the dynamics of the ^{235}U separation and its two fragments' creation [7,8]. As seen in Fig. 2, when the temperature of the original nucleus increases, there is a transition from the asymmetric or two-humped shape to the symmetric one-humped MCSFF one, which is formed

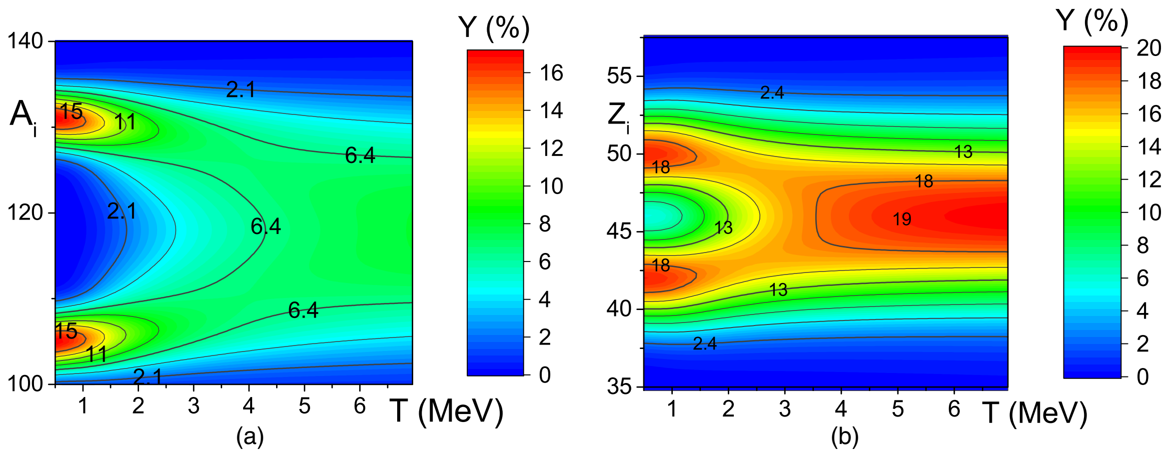

FIG. 2. Temperature dependence of mass (a) and charge (b) yields of the fission fragments of the isotope ^{235}U.

by the clusters of nuclear fragments containing short-lived $\{^{117,118}\mathrm{Pa}\}$, $\{^{114,113}\mathrm{Ru}, ^{121,122}\mathrm{Cd}\}$, and other isotopes.

As mentioned above, the symmetrization of MCSFF occurs due to the increase of the influence of the entropy term in Eq. (1) with temperature increase, which results in the symmetric or one-humped fission mode shape. Note that the configuration entropy represents the statistical peculiarities of the restructuring of the nuclear ensemble for a given temperature T and is sensitive to the changes of their mass-charge ratio and redistribution of nucleons in the fission fragments. These clusters form the MCSFF shown in Fig. 2, and, apparently, the probability of their realization is also determined by an entropy term in Eq. (1), especially at high temperatures.

Some experimental results for a given range of temperatures indicate that the peak is much broader and the heavy-fragment peak ranges from $A = 132$ and $A = 145$.

Note that the abovementioned features $S(A_i)$ and $S(Z_i)$ [Fig. 1(b)] are responsible for the formation of a different topology of the mass and charge spectra of fission fragments (Fig. 2). Of particular interest is the study of the redistribution of the mass-charge spectrum of the fission fragments when the temperature T increases. The changes in the proton-neutron ratio in the MCSFF that occur in this case modify the value of the initial or total energy term U in Eq. (1) and lead to the new equilibrium state of an ensemble in general. Moreover, the temperature growth reduces the probability of the realization of fission fragments containing magic numbers of protons and neutrons and stimulates the yield of the nuclei with less specific binding energy than in the case of "cold" fission.

The result of such a temperature arrangement within an ensemble of fission fragments is the change in their initial energy, as well as the energy released in the fission of ^{235}U. To study the features of the thermodynamic ordering, one has to investigate the temperature dependence of the heat capacity $C(T)$, which can be determined as the derivative at a constant pressure of the initial energy of the two-fragment nuclear cluster, $C(T) = \frac{\partial \overline{U}}{\partial T}$, where

$$\overline{U} = -\frac{d\ln(Z_p)}{d\beta} = \sum_{\langle A_i\rangle} \varepsilon_i F(A_i). \tag{7}$$

Here Z_p is the partition function (6), $\beta = 1/\mathrm{T}$, ε_i is taken from the $\{\varepsilon_i(V)\}$ set, and the method of summation by $\langle A_i\rangle$ has the same meaning as in Eq. (2). In this case, $C(T)$ is a dimensionless quantity.

The results of calculation of $U = \overline{U}/A_0$ and, thus, for $C(T)$ in the 0.5–7 MeV temperature range are shown in Fig. 3. As seen, $U(T)$ decreases with T. The fine structure in Fig. 3(a) is nonexpressive; only the bending of the $U(T)$ curve takes place in the 1–2 MeV range. The temperature dependence of the heat capacity $C(T)$ [Fig. 3(b)] in this temperature region has a clear peak, as well as some less expressed features at higher temperatures. The peaklike dependence of $C(T)$ is a well-known phenomenon in solid-state theory, indicating the presence of some temperature anomaly for materials having metastable states with a discrete set of energy levels for structural configurations such as polymers, glasses, or defective semiconductors, and was called the Schottky anomaly [20,21]. Schottky anomalies occur in systems with discrete energy levels, and when the temperature approaches the difference between the energy levels, there is a significant change in entropy for the small temperature variation.

The presence of the peaklike $C(T)$ behavior with temperature for nuclear matter that is a postscission ensemble of the nuclei-fragment clusters reflects the features of the yields for the two-fission nuclear clusters with the discrete energy spectrum $\{\varepsilon_i(V)\}$. The anomaly of the $C(T)$ [Fig. 3(b)] also shows the loss of the statistical nonequivalence of nucleons and the drastic change in the entropy term S [Fig. 1(b)] when ^{235}U is heated. As well as in the case of solids, the temperature-dependent conflict between the energy and the entropy effect is a necessary condition of its observation and may be considered the phase transition [22]. This sort of the phase transition of nuclear matter differs from the reactions of multifragmentation and spallation studied recently [23–25]. It should be noted that experimental observation of the anomaly in the $C(T)$ at the nuclear fission reactions is possible. As well as in the case of solid-state matter, one can detect such effects in the experiments with the fissioning nuclei "heating" in the form of a smooth increase in their excitation energy with the temperature increase. This can be done in the synthesis-type reactions and then in the fission-type reactions of heavy nuclei

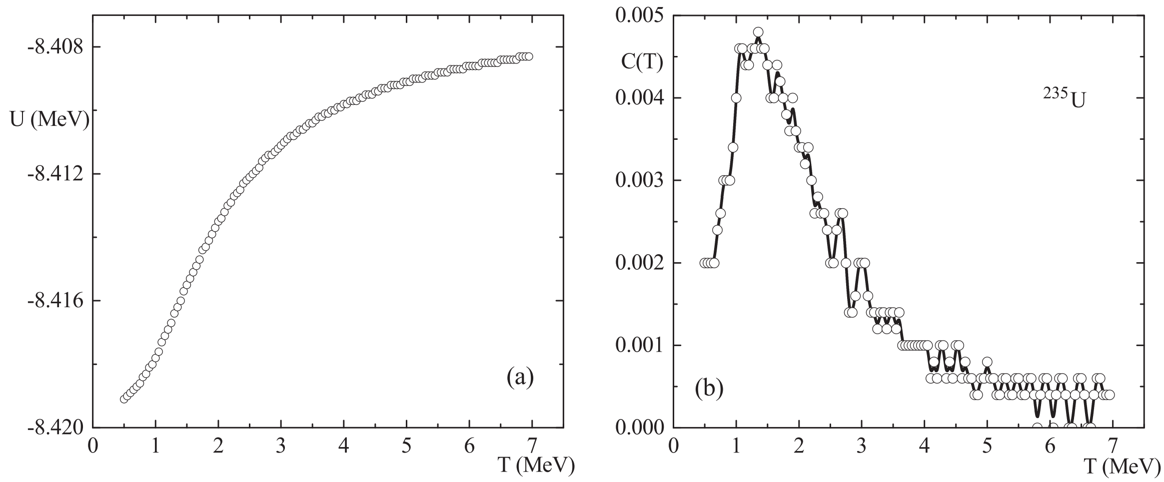

FIG. 3. (a) Temperature dependence of the initial energy and (b) Heat capacity $C(T)$ for the ensemble of fission fragments of ^{235}U.

in nuclear-nuclear collisions according to the "projectile and target" schemes. Another way is the direct nuclei fission reactions stimulated by incident particles such as protons, gamma quanta, etc., with increasing excitation energy of the fissioning nucleus. Like in Fig. 3, the experiment must provide a smooth kinetic energy transfer to the fissioning nucleus in the range of 0.5–5 MeV/nucleon. The subject of such study can be observables such as the characteristics of different secondary projectiles in the spallation and fragmentation reactions: the cross sections and the velocity spectra. These parameters, being dependent on the energy released in the fission of the initial nucleus, should have a nonlinear behavior in the above nuclear temperature range [see Fig. 3(a)]. The derivative of such dependencies can demonstrate the peaklike behavior that reproduces the same trend of $C(T)$. A preliminary list of nuclear centers capable of realizing these experiments is given in Refs. [3,26–28].

IV. CONCLUSIONS

In summary, the study of the temperature dependence of thermodynamic parameters of the ensemble of ^{235}U fission fragments has demonstrated the possibility of their abnormal behavior in the range of nuclear temperatures of 1–2 MeV. This effect may be due to such fundamental factors as the loss of statistical nonequivalence or the identity of nucleons (protons and neutrons) in the fission fragments. The above anomaly of $C(T)$ indicates the realization of a new type of phase transition for nuclear matter due to symmetric or second-type phase transitions that are well known in solid-state physics. Their study may complement the pattern of other phase transitions in nuclear matter studied earlier. Note the experimental possibility of observing such a phase transition according to the scheme realized in Refs. [23,24].

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[1] F. A. Khan, D. Bhowmick, D. N. Basu, M. Farooq, and A. Chakrabarti, Phys. Rev. C **94**, 054605 (2016).

[2] D. Porcelli and M. Baskaran, in *Handbook of Environmental Isotope Geochemistry*, edited by M. Baskaran (Springer, Berlin, 2012), pp. 11–32.

[3] K.-H. Schmidt, S. Steinhäuser, C. Böckstiegel, A. Grewe, A. Heinz, A. Junghans, J. Benlliure, H.-G. Clerc, M. de Jong, J. Müller, M. Pfützner, and B. Voss, Nucl. Phys. A **665**, 221 (2000).

[4] R. H. Cyburt, B. D. Fields, K. A. Olive, and T. H. Yeh, Rev. Mod. Phys. **88**, 015004 (2016).

[5] K.-H. Schmidt and B. Jurado, Review on the Progress in Nuclear Fission, www.khs-erzhausen.de/Preprints/Review-on-Progress-in-Nuclear-Fission.pdf, 2018, Nuclear Studies Center of Bordeaux Gradignan.

[6] A. Andreyev, K. Nishio, and K.-H. Schmidt, Rep. Prog. Phys. **81**, 016301 (2018).

[7] J. Randrup and P. Möller, Phys. Rev. Lett. **106**, 132503 (2011).

[8] J. Sadhukhan, C. Zhang, W. Nazarewicz, and N. Schunck, Phys. Rev. C **96**, 061301 (2017).

[9] P. Fong, Phys. Rev. **102**, 434 (1956).

[10] K. T. R. Davies, A. J. Sierk, and J. R. Nix, Phys. Rev. C **13**, 2385 (1976).

[11] J.-F. Lemaitre, S. Panebianco, J.-L. Sida, S. Hilaire, and S. Heinrich, Phys. Rev. C **92**, 034617 (2015).

[12] G. Audi, F. Kondev, M. Wang, W. Huang, and S. Naimi, Chin. Phys. C **41**, 030001 (2017).

[13] G. Audi, M. Wang, A. Wapstra, F. Kondev, M. MacCormick, X. Xu, and B. Pfeiffer, Chin. Phys. C **36**, 1603 (2012).

[14] V. Maslyuk, O. Parlag, and T. Marinets, Phys. Part. Nucl. Lett. **4**, 46 (2007).

[15] V. T. Maslyuk, O. Parlag, M. I. Romanyuk, O. I. Lendyel, and T. I. Marinets, Europhys. Lett. **119**, 12001 (2017).

[16] V. T. Maslyuk, O. A. Parlag, O. I. Lendyel, T. I. Marynets, M. I. Romanyuk, O. S. Shevchenko, J. J. Ranyuk, and A. M. Dovbnya, Nucl. Phys. A **955**, 79 (2016).

[17] L. G. Girifalco, *Statistical Physics of Materials* (Wiley, New York, 1973).

[18] A. Kelić, J. B. Natowitz, and K.-H. Schmidt, Eur. Phys. J. A **30**, 203 (2006).

[19] J. Wilson *et al.*, Phys. Rev. Lett. **118**, 222501 (2017).

[20] A. Tari, *The Specific Heat of Matter at Low Temperatures* (Imperial College, London, 2003).

[21] V. T. Maslyuk, J. Non-Cryst. Solids **212**, 80 (1997).

[22] J. Lee, J. Korean Phys. Soc. **67**, 1133 (2015).

[23] M. Hempel, V. Dexheimer, S. Schramm, and I. Iosilevskiy, Phys. Rev. C **88**, 014906 (2013).

[24] B. K. Sharma and A. Bhagwat, Acta Phys. Pol., B **48**, 195 (2017).

[25] S. Mallik, G. Chaudhuri, P. Das, and S. Das Gupta, Phys. Rev. C **95**, 061601 (2017).

[26] R. Raabe, Eur. Phys. J. Plus **131**, 362 (2016).

[27] Y. Liu, D. W. Stracener, and T. Stora, New J. Phys. **19**, 085005 (2017).

[28] T. Motobayashi, EPJ Web Conf. **66**, 01013 (2014).