A Nonequilibrium Information Entropy Approach to Ternary Fission of Actinides

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Nuclear fission, discovered eighty years ago, remains an exciting field of research. In the last few decades, an amazing progress has been realized with respect to experimental investigations and phenomenology, as well as in theoretical treatments such as time-dependent Hartree-Fock-Bogoliubov (TDHFB) or time-dependent superfluid local density approximation, generator coordinate methods, and other techniques [1 2], but basic concepts are still open for discussion. A full microscopic description is still lacking, and it remains a challenge to present state quantum many-body theory. For a recent review on studies of thermal neutron induced (n, f), and spontaneous fission (sf) of actinides, as well as a discussion on open theoretical questions, the reader should refer to [3 4].

From a theoretical point of view, the fission process can be generally described via a picture in which the deforming nucleus, following a dynamic path, subject to fluctuations, crosses a saddle point where the nascent fission fragments are formed. The deformed dumbbell-like system, consisting of two main fragments and the connecting neck region, then evolves toward the scission point where the rupture occurs. The saddle-to-scission time is estimated as $\tau_{ss} \approx 10^{-3} - 10^{3}$ fm/c [2]. Characterization of the system’s properties and its time evolution, and the description of dissipation during this process remains nowadays a significant problem. However, dissipative dynamics has been applied to describe the non-adiabatic evolution from the saddle point to scission, see [3], though a rigorous treatment of the scission process is still unavailable.

A few signals can be used to obtain information about the scissioning nucleus, such as the mass and energy distributions of the two fission fragments, and the multiplicities of the emitted particles, which are primarily neutrons, and of $\gamma$-radiation, that can be utilized to characterize excitation energies and spins. Although the use of concepts, like the temperature, which is defined for thermodynamic equilibrium, might not be well-founded, approaches introducing concepts from statistical physics, to characterize the distribution of emitted particles, are often employed. For instance, in Refs. [5 6], the prompt fission neutron spectra of different actinides are analyzed with temperature-like parameters of the order of 1 MeV, and in Refs. [6 7], the analysis of prompt fission $\gamma$-ray spectra for actinides also suggests a temperature-like parameter of the same order. However, one has to separate prompt emission from later emission, that occurs during the de-excitation of the fission fragments, and this presents a problem, since it is not easy to identify observables which may be directly associated with the neck region at scission.

One such signature is the emission of light clusters observed in ternary fission processes, see, e.g., [14 15] and references given there. A light cluster, most often $^4$He, is emitted in a direction perpendicular to the symmetry axis defined by the two fission products, which have isospin $A/Z$ and are emitted in a direction perpendicular to the symmetry axis defined by the two fission products, which have anisotropy $A/Z$. Only a few of the observed light isotopes $\{A/Z\}$ and energies have been measured for a number of different actinide nuclei, in particular $^{233}$U(nth,f), $^{235}$U(nth,f), $^{239}$Pu(nth,f), $^{241}$Pu(nth,f), $^{248}$Cm(sf), and $^{252}$Cf(sf), see Refs. [15 16]. Data for these observed yields $Y^{ob}_{A/Z}$, normalized to $^4$He$^{ob}$ = 10000, are presented in Tab. [1]. The investigation of ternary fission has the advantage that it is directly related to the scission process, and that it can be localized in the neck region.
As known from α-decay studies, a mean-field approach like TDHFB has problems describing the formation of clusters. For ternary fission, parametrizations of the measured yields employing a statistical distribution with a temperature-like parameter $\approx 1$ MeV, see [17–19], have been explored. However, any interpretation of the detected yields in the detector contains contributions from decaying excited states and resonances so that the observed yield distribution differs from the primary distribution at the time of scission. In addition, yields of the larger light isotopes, $A \geq 10$, are clearly overestimated by the simple NSE statistical equilibrium (NSE) model, see Eq. (3) below, faces some problems. The NSO $\rho(\tau, t)$ [24] is a solution of the von Neumann equation (1) based on the nonequilibrium statistical operator (NSO),

$$ p(t) = \lim_{\epsilon \to 0} \epsilon \int_{-\infty}^{t} dt' e^{-\epsilon(t-t')} e^{-\frac{i}{\hbar}H(t-t')} \rho_{rel}(t') e^{\frac{i}{\hbar}H(t-t')} . $$

The NSO $p(t)$ [24] is a solution of the von Neumann equation with boundary conditions characterizing the state of the system in the past, as expressed by the relevant statistical operator. This relevant statistical operator $\rho_{rel}(t)$ is constructed from known averages using information theory. As it is well known from statistical physics, the

| isotope | $R^{\nu,\lambda}_{A,Z}$ (1.3) | $^{233}$U(n_{th},f) | $^{235}$U(n_{th},f) | $^{239}$Pu(n_{th},f) | $^{241}$Pu(n_{th},f) | $^{248}$Cm(sf) | $^{252}$Cf(sf) |
|--------|------------------|-----------------|-----------------|-----------------|-----------------|----------------|----------------|
| $^1$H  | -                | 1.24177         | 1.21899         | 1.3097          | 1.1900          | 1.23234        | 1.25052        |
| $^3$H  | -                | -3.52615        | -3.2627         | -3.46688        | -3.02055        | -2.92719       | -3.1107        |
| $^5$He | 0.973            | 40.986          | 49.76           | 68.632          | 41.563          | 49.533         | 61.579         |
| $^8$He | 0.998            | 457.27          | 715.29          | 714.79          | 780.39          | 913.76         | 943.12         |
| $^{10}$He | 0.0876      | 2.7772          | 4.97            | 5.627           | 6.057           | 8.742          | 8.219          |
| $^{12}$He | 0.097         | 0.0124          | 0.0076          | 0.0253          | 0.00431         | 0.00645        | 0.00933        |
| $^{14}$He | -              | 10000           | 10000          | 10000           | 10000           | 10000          | 10000          |
| $^{16}$He | 1               | 8858.46         | 8706.1          | 8615.7          | 8556.9          | 8331.98        | 8454.0         |
| $^{20}$He | 0.689          | 1130.75         | 1289.04         | 1374.7          | 1439.0          | 1680.75        | 1540.9         |
| $^{24}$He | -              | 137             | 191             | 192             | 260             | 354            | 270            |
| $^{28}$He | 0.933          | 115.89          | 158.98          | 159.01          | 211.68          | 276.96         | 222.4          |
| $^{32}$He | 0.876          | 21.262          | 33.997          | 35.983          | 51.742          | 80.634         | 58.16          |
| $^{36}$He | -              | 0.9989          | 0.9897          | 0.9846          | 0.9860          | 0.9899         | 0.9622         |
| $^{40}$He | 0.971          | 3.4725          | 6.764           | 6.4095          | 12.481          | 21.280         | 13.32          |
| $^{44}$He | 0.255          | 0.047077        | 0.105           | 0.111           | 0.219           | 0.455          | 0.258          |
| $^{48}$He | -              | 1.0229          | 1.1936          | 1.3496          | 1.1811          | 1.1042         | 1.8409         |
| $^{52}$He | 1.07           | 5.7727          | 2.594           | 5.147           | 2.188           | 2.819          | 2.544          |

TABLE I. Lagrange parameters $\lambda_i$, observed yields $Y^{\nu,\lambda}_{A,Z}$ [14, 15, 17] and primary yields $Y^{rel, vir}_{A,Z}$ (virial approximation) from ternary fission $^{233}$U(n_{th},f), $^{235}$U(n_{th},f), $^{239}$Pu(n_{th},f), $^{241}$Pu(n_{th},f), $^{248}$Cm(sf), and $^{252}$Cf(sf) (lines denoted by the superscript 'obs') as well as relevant (primary) yields $Y^{rel, vir}_{A,Z}$ for H, He nuclei. The prefactor $R^{\nu,\lambda}_{A,Z}(\lambda_T)$ at $\lambda_T = 1.3$ MeV which represents the intrinsic partition function is also given. The final yields $Y^{rel, vir}_{He} = Y^{rel, vir}_{He} + Y^{rel, vir}_{He}$ and $Y^{rel, vir}_{He} = Y^{rel, vir}_{He} + Y^{rel, vir}_{He}$ are compared to the observed yields. For more details see the Supplementary material [21].

In this work, we use ternary fission data to investigate the scission process. In particular, we extend the nonequilibrium approach, already presented in Ref. [22], but with $Z \leq 2$, considering partial intrinsic partition functions including continuum contributions on the level of quantum virial expansions. We present results for all the actinides given in Table I for which the necessary set of yields is available, and we derive critical values of parameters that are relevant for nucleation kinetics.

We describe fission as a nonequilibrium process, using the method of the nonequilibrium statistical operator (NO),

$$ p(t) = \lim_{\epsilon \to 0} \epsilon \int_{-\infty}^{t} dt' e^{-\epsilon(t-t')} e^{-\frac{i}{\hbar}H(t-t')} \rho_{rel}(t') e^{\frac{i}{\hbar}H(t-t')} . $$

(1)
relevant distribution is determined from the maximum of information entropy under given constraints, which are represented by Lagrange multipliers \( \lambda_i \). A minimum set of relevant observables consists of the conserved observables, energy \( H \), and the numbers \( N_r \) of neutrons and protons \( (\tau = n, p) \). The solution is the generalized Gibbs distribution

\[
\rho_{\text{rel}} \propto \exp\left[-\left(H - \lambda_n N_n - \lambda_p N_p\right)/\lambda_T\right].
\]

(2)

Note that these Lagrange multipliers \( \lambda_i \), which are in general time-dependent, are not identical with the equilibrium parameters \( T \) and \( \mu_r \), but may be considered as nonequilibrium generalizations of the temperature and chemical potentials. Only when the system is in thermodynamic equilibrium, can the information entropy be unambiguously identified with the thermodynamic entropy, and from this we define the quantities \( T \) and \( \mu_r \) with the known properties. Note that the NSO allows the possibility of including other relevant observables, such as the pair amplitude in the superfluid state, or the occupation numbers of the quasiparticle states to derive kinetic equations and calculating reaction rates [24].

Typically, in a variational problem, we have to replace the Lagrange multipliers \( \lambda_i \) by given mean values [22]. In contrast to the noninteracting system (ideal quantum gases), where the equilibrium solutions are the well-known equations of state, the interaction in the Hamiltonian leads to a many-particle problem which can be treated with the methods of quantum statistics. (Note that the mathematical concepts developed in equilibrium quantum statistics can also be used for the generalized Gibbs state \( \rho_{\text{rel}} \).) We perform a cluster expansion for the interacting system [25], and partial densities of different clusters \( \{A, Z\} \) are introduced. The relevant yields \( Y_{\text{rel, vir}}^{\text{vir}} \) in the virial approximation are calculated as

\[
Y_{A,Z}^{\text{rel, vir}}(\lambda_T) \propto R_{A,Z}^{\text{vir}}(\lambda_T) g_{A,Z} \left( \frac{2\pi\hbar^2}{Am\lambda_T} \right)^{-3/2} e^{(B_{A,Z} + (A-Z)\lambda_T)/\lambda_T}
\]

(3)

(nondegenerate limit), where \( B_{A,Z} \) denotes the (ground state) binding energy and \( g_{A,Z} \) the degeneracy [26]. The prefactor

\[
R_{A,Z}^{\text{vir}}(\lambda_T) = 1 + \sum_{\text{exc}} \left[ g_{A,Z,i} / g_{A,Z} \right] e^{-E_{A,Z,i}/\lambda_T}
\]

(4)

is related to the intrinsic partition function of the cluster \( \{A, Z\} \). The summation is performed over all excited states of excitation energy \( E_{A,Z,i} \) and degeneracy \( g_{A,Z,i} \) [26], which decay to the ground state. Also, the continuum contributions are included in the virial expression. For instance, the Beth-Uhlenbeck formula expresses the contribution of the continuum to the intrinsic partition function via the scattering phase shifts, see [27, 28]. For \( R_{A,Z}^{\text{vir}}(\lambda_T) = 1 \), the simple NSE is obtained, i.e., neglecting the contribution of all excited states inclusive continuum correlations.

In the low-density limit, virial expansions of the intrinsic partition functions of the channel \( \{A, Z\} \) have been obtained [23, 27, 28] for \( ^6\text{H}, ^4\text{H}, ^4\text{He}, ^8\text{Be} \) using the measured phase shifts in the corresponding channels. The values are given in the second column of Tab. I for \( \lambda_T = 1.3 \text{ MeV} \). Well-bound states with energy of the continuum edge large compared to \( \lambda_T \) have only a weak contribution of the continuum states so that \( R_{A,Z}^{\text{vir}}(\lambda_T) \approx 1 \), if no further excited states are present. An interpolation formula, which relates the prefactor \( R_{A,Z}^{\text{vir}}(\lambda_T) \) to the energy of the edge of continuum, is given in [24], and the corresponding estimates of the prefactor for the He isotopes with \( 6 \leq A \leq 9 \) are also shown in Tab. I.

Within the NSO approach [1], the relevant distribution \( \rho_{\text{rel}} \) serves as initial condition to solve the von Neumann equation for \( \rho(t) \) describing the evolution of the system according to the system Hamiltonian. The relevant (primary) distribution \( Y_{A,Z}^{\text{rel}}(\lambda_T) \) contains stable and unstable states of nuclei, as well as correlations in the continuum (e.g., resonances).

The concept of introducing the relevant primary yield distribution according to the NSO is supported by several experimental observations, among these are the observation of \(^5\text{He}\) and \(^7\text{He}\) emission [13]. For \( Z > 2 \), as it is discussed below, 3.368 MeV \( \gamma \) rays from the first excited state of \(^{10}\text{Be}\) have been observed [29], and excited states of \(^8\text{Li}\) at 2.26 MeV excitation energy have been reported in Ref. [15]. Also of interest are the inferred data for \(^8\text{Be}\) and \(^7\text{Li}_{7/2}^-\) observed in [29], which cannot be described with the NSE but demand a treatment with continuum states.

The relevant distribution evolves dynamically to the final, observed yields according to the von Neumann equation. This process is described by reaction kinetics, and the NSO allows calculation of the reaction rates [24]. Here, we approximate this process by the feeding of the observed states from the primary states occurring in the relevant distribution. For example, for \( Z \leq 2 \), the final yields are related to the primary yields \( Y_{\text{rel}} \) as \( Y_{\text{final}} = Y_{\text{rel}} + Y_{\text{rel}} \), \( Y_{\text{final}} = Y_{\text{rel}} + Y_{\text{rel}} + Y_{\text{rel}} \), \( Y_{\text{final}} = Y_{\text{rel}} + Y_{\text{rel}} + Y_{\text{rel}} \), and \( Y_{\text{final}} = Y_{\text{rel}} + Y_{\text{rel}} + Y_{\text{rel}} + Y_{\text{rel}} \).

In this work, to construct the relevant distribution \( \rho_{\text{rel}} \) from an information theoretical approach, we use the least squares method, see [21], to reproduce the observed yields. We calculate the primary distribution \( Y_{A,Z}^{\text{rel, vir}} \) using the intrinsic partition function in the virial form, i.e., using the excited states and scattering phase shifts neglecting in-medium corrections. The optimum values of the Lagrange parameters \( \lambda_i \) are given in Tab. I for the different ternary fissioning actinides. Of interest is the dependence of \( \lambda_i \) from \( \{A, Z\} \) of the parent actinide nucleus [18, 19]. The current accuracy of the experimental data is not sufficient to determine significant trends.

The measured total yields of H and He isotopes are nearly perfectly reproduced by the corresponding sums of primary yields. The yield of \(^6\text{He}\) is slightly overestimated by \( Y_{\text{rel, vir}} \). In contrast, the yield of \(^8\text{He}\) is
underestimated by $Y_{\alpha,He}^{\text{final,vir}}$. Both ratios $Y_{\alpha,He}^{\text{obs}}/Y_{\alpha,He}^{\text{final,vir}}$ and $Y_{\alpha,He}^{\text{obs}}/Y_{\alpha,He}^{\text{rel}}$ are presented in Tab. 1. Presently, the relevant distribution $Y_{\alpha,Z}^{\text{rel,vir}}$ does not take in-medium effects, in particular Pauli blocking, into account. Medium modiﬁcations are more effective for weakly bound clusters. As proposed in [22] for $^{252}$Cf(sf), a stronger reduction of the yield of $^8$He obs compared to $^8$He rel may be related to the very low binding energy (0.975 MeV) of the $^8$He nucleus below the $\alpha + 2n$ threshold. The suppression of $^8$He obs appears for all considered systems and may be considered as a signature of the Pauli blocking. Pauli blocking is determined by the medium surrounding the cluster, and an estimate of the corresponding neutron density was given in [22]. However, to address the problem, precise experimental data are needed. Experimental studies are still scarce, and the data are often not consistent [31,32]. Unbound nuclei such as $^5$He should be very sensitive to medium modiﬁcations. The virial expression for the intrinsic partition function is known [28], and the corresponding primary yields are given in Tab. 1. Fortunately, in the case of $^{252}$Cf, the primary yields of $^6$He and $^7$He have been measured [15], and the value $Y_{^5He}^{\text{obs}} = 1736(274)$ has been obtained. In principle, because of the medium modiﬁcations, the different cluster states may serve as a probe to determine the neutron density in the neck region at scission, but the uncertainties are still rather large.

Nuclei with $Z > 2$ are also observed in ternary fission. A detailed measurement of the yields of isotopes up to $^{30}$Mg has been made for $^{241}$Pu(nth,f) [14,16]. Extended sets of data for $Z > 2$ are also measured for $^{235}$U(nth,f) and $^{245}$Cm(nth,f) [16]. We extend our analysis of the measured data up to $Z = 6$ using the relevant distribution, see [20]. In general, the neutron separation energy $S_n$, for each isotope, is adopted as the threshold energy for the continuum, but cluster decay is also possible, e.g., $^6$Li $\rightarrow \alpha + d$, $^7$Li $\rightarrow \alpha + t$, $^7$Be $\rightarrow \alpha + h$, $^8$Be $\rightarrow 2\alpha$, $^{10}$B $\rightarrow \alpha + ^5$Li, etc. In some cases, such as $^6$He, $^8$He, $^{11}$Li, two-neutron separation determines the threshold. To estimate the continuum correlation, the interpolation $P_{\alpha,Z}(\lambda_T)$ [22] was used at the corresponding binding energy $E_{\alpha,Z}^{\text{thresh}} - E_{\alpha,Z,i}$ of the (ground state or excited) cluster. The ﬁnal yields $Y_{\alpha,Z}^{\text{final,vir}}$ are calculated as sums of the feeding contributions, see [20].

The question arises whether global Lagrange parameters $\lambda_T, \lambda_n, \lambda_p$ exist, which are valid for all $Z$, as expected for matter in thermodynamic equilibrium. Before we discuss this question, we present a calculation with the relevant distribution given above, employing only three Lagrange parameters $\lambda_i$, but taking also Li isotopes into account. A least squares ﬁt of ﬁnal yields $Y_{\alpha,Z}^{\text{final,vir}}$ to $Y_{\alpha,Z}^{\text{obs}}$ for $^2$H, $^3$H, $^4$He, $^8$He, $^7$Li, $^8$Li, $^9$Li has been performed. The accuracy of the ﬁt increases since $^4$He and $^{11}$Li are not included. Both are weakly bound systems for which medium effects and dissolution may become of relevance, as discussed above. Again we emphasize that in-medium corrections are not included in the present calculation. The Lagrange parameter values $\lambda_T = 1.2023$ MeV, $\lambda_n = -2.9981$ MeV, $\lambda_p = -16.6285$ MeV are obtained. There are only minimal changes compared to those derived from the ﬁt of Tab. 1 for $^{241}$Pu(nth,f), and we conclude that our approach can also reproduce the yields of isotopes with $Z > 2$.

Using these Lagrange parameter values $\lambda_i$ and considering all observed data for isotopes with $Z \leq 6$, the ratio $Y_{\alpha,Z}^{\text{obs}}/Y_{\alpha,Z}^{\text{final,vir}}$ is shown as a function of the mass number $A$ in Fig. 1. Surprisingly, yields of $^9$Be, $^{10}$Be, $^{11}$B are also well reproduced. For $A \geq 11$, the calculations overestimate the observed yields, and the ratios decrease strongly, starting around $A = 10$.

An explanation of the decrease has been given in [21] using nucleation theory. Whereas small clusters are already in the quasi-equilibrium distribution $Y_{\alpha,Z}^{\text{rel}}$, larger clusters need more formation time so that the observed yields are smaller than those predicted by the relevant distribution. From reaction kinetics, the expression

$$Y_{\alpha,Z}^{\text{obs}}/Y_{\alpha,Z}^{\text{final,vir}} = \frac{1}{2} \text{erfc} \left[ b(\tau)(A^{1/3} - a(A,c,\tau)) \right]$$

is obtained, see [21], where $b(\tau) = (27.59 \text{ MeV}/\lambda_T)^{1/2} \times (1 - e^{-2\tau})^{-1/2}$ and $a(A,c,\tau) = A^{1/3}(1 - e^{-c\tau}) + e^{-\tau}$. With $\lambda_T = 1.2$ MeV, the least squares ﬁt to the data of $^{241}$Pu(nth,f) (black line in Fig. 1) gives $\tau = 1.5406$, $A_c = 16.143$. Then, $\sigma_{\tau,s-s} = \tau A_c^{2/3}/(3.967 \rho)$, and with $\rho = 4 \times 10^{-4} \text{ fm}^{-3}$ [21] follows $\sigma_{\tau,s-s} = 6793 \text{ fm}/c$. This time scale supports the slow evolution from saddle to scission proposed recently as a dissipative process [2,3].

The strong reduction of isotopes $A > 10$ compared to estimates of a statistical model is also seen in [17]. In
addition, the overestimate of $^3$He is shown. The correct treatment of continuum states proposed in this letter removes this discrepancy. In addition, the yields of weakly bound clusters $^{11}$Li, $^{10}$C are strongly overestimated, see [33]. A reason may be the shift of the binding energy due to in-medium effects. If the density is larger than the Mott density, the bound states are dissolved. Bond due to in-medium effects. If the density is larger than

$^{6}$Be, $^{11}$Be, $^{14}$Be, $^{14}$B, $^{15}$C. The yields of all these isotopes are overestimated. This may be considered as indication of in-medium effects (Pauli blocking) leading to a shift and possibly the dissolution of the cluster. This possibility should be considered when more accurate data are available.

Yields of ternary fission of $^{241}$Pu$n_{th}, f$ were also calculated within the evaporation-based surface-plus-window dissipation model [19] [34], where the yields of light clusters are obtained considering the height of the Coulomb barrier as a dynamical variable. The Coulomb interaction has been treated as mean field. To improve the liquid drop model used in that phenomenological approach, an adequate theory of ternary fission should be based on the TDHF solutions. It should be pointed out that the neutrons and protons have extended wave functions and are not correctly described by the distribution [3], see [22]. The Coulomb interaction contributes to the Hamiltonian also in our approach which will be extended to the treatment of inhomogeneous systems. As pointed out in Ref. [2], the dynamics near scission is strongly dissipative and existing versions of TDDFT are not adequate since they are lacking fluctuations in collective coordinates.

In conclusion, ternary fission is a dissipative process, and understanding fission of actinide nuclei is of fundamental interest to work out nonequilibrium statistical physics. Emitted light isotopes are described by a nonequilibrium distribution, excited states and continuum correlations are taken into account on the level of virial expansions. In-medium effects and nucleation kinetics are interesting aspects to reconstruct the nonequilibrium distribution. An improved description of the influence of mean-field effects may be obtained from TDHF calculations and similar approaches, but cluster formation remains problematic in any mean-field theory. Ternary fission is an outstanding signal to explore the fission process, and more consistent and accurate data are necessary to work out a complete description of the ternary fission process within non-equilibrium quantum statistics.

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