Thermodynamics of nuclei in thermal contact

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(Received 5 October 2010; revised manuscript received 23 December 2010; published 28 January 2011)

The thermodynamic properties of a dinuclear system are studied with the methods of statistical mechanics. A schematic model calculation shows that the excitation-energy transfer proceeds in energy steps of considerable amount which are subject to large fluctuations. As a consequence, thermal averaging is strong enough to assure the application of thermodynamical methods for describing the energy exchange between the two nuclei in contact. In particular, thermal averaging justifies the definition of a nuclear temperature. The division of excitation energy in thermal equilibrium is derived for several analytical descriptions of the level density.

DOI: 10.1103/PhysRevC.83.014607 PACS number(s): 24.60.−k, 25.85.−w

I. INTRODUCTION

One of the key quantities characterizing a nuclear reaction is the excitation energy found in the reaction products. The excitation energy summed over all products gives information on the dissipative character of the reaction [1,2], whereas the division of the excitation energy between different products provides information on more subtle dynamical effects [3–5]. Indications for dynamical effects were deduced from deviations of the measured excitation-energy partition from the expectation of thermodynamical equilibrium between the products at the moment of separation, see, e.g., Refs. [3–5], which was deduced from the condition that the reaction products have equal temperatures.

Very recently, the process of thermal equilibration of a dinuclear system in the regime of strong pairing correlations has been studied. Guided by recent experimental findings on nuclear level densities, it has been discussed that two moderately excited nuclei, brought into contact, form a system with very peculiar thermodynamic properties. Nuclei in this regime are characterized by a temperature, which is specific to the nucleus and varies only weakly with excitation energy. If the two nuclei have different temperatures, it has been derived that thermal equilibration induces that all excitation energy is transferred to the nucleus with the lower temperature in a process of energy sorting [6]. This process is driven by entropy, which evolves toward its maximum possible value in an isolated system according to the second law of thermodynamics.

In all these considerations, the nuclear temperature T was derived from the energy dependence of the nuclear state density Ω:

$$T = \left[\frac{\partial(\ln\Omega)}{\partial E}\right]^{-1} \tag{1}$$

A first difficulty arises from the fact that experiments most often yield the nuclear level density ρ. The state density differs from the level density by the spin degeneracy of the nuclear levels. Fortunately, the slow increase of the spin cut-off parameter with energy has practically no influence on the energy dependence of the nuclear state density. Thus, in a restricted energy range, the spin degeneracy can well be considered by a constant factor [7], and the nuclear temperature may also be deduced from the nuclear level density according to the equation

$$T \approx \left[\frac{\partial(\ln\rho)}{\partial E}\right]^{-1} \tag{2}$$

with a good approximation.

A more crucial problem is the application of thermodynamical concepts in the above-mentioned publications. The justification for assigning a temperature to microscopic or mesoscopic systems like nuclei is not evident, and the limitations of the applicability of canonical thermodynamics to small systems [8–12] are repeatedly discussed. Limitations of canonical and macrocanonical thermodynamics have also been discussed for larger nonextensive systems, e.g., for systems subject to long-range forces [13,14].

It is obvious that the system consisting of two nuclei in contact forms a microcanonical ensemble, e.g., there is no heat bath. In addition, the total energy of the system is finite and a constant of time. The same is true for the total number of particles. Due to its small size, however, the temperature of an isolated nucleus is not well defined; it is subject to fluctuations. This becomes obvious by the fluctuations of the single-particle occupation distribution from one nuclear level to another. For example, in the spirit of the exciton model [15], the excitation energy may be shared by a strongly different number of particles and holes. The analytical relations of the Fermi-Dirac statistics apply only to an average over the single-particle distributions of many neighboring nuclear levels.

At very low excitation energy, there arises still another difficulty: The nuclear levels are discrete, and they no longer overlap. Thus, the nuclear level density cannot be expressed by a continuous function of energy. This puts the definition of the nuclear temperature, Eq. (2), and its use for describing the thermodynamic behavior of the system considered in doubt even more.

The present work aims at clarifying the question how the thermodynamics of nuclei, which are brought into thermal contact, can be properly described. In particular, we are interested in the process of thermal equilibration.

An additional remark is in place: The condition of a fixed total excitation energy of the dinuclear system is not realized when the two nuclei brought into thermal contact move with large relative velocity. This is usually the case in heavy-ion collisions, where the system is heated by one-body dissipation according to the window formula [16]. If there is a net mass flow in addition, the nucleus that grows in size is heated more, preventing thermal equilibration. Our considerations apply to the case when the energy per nucleon, corresponding to the relative motion of the two nuclei, is small compared to the temperatures involved. This scenario is assumed to be realized in nuclear fission. The dissipation present also in this case due to the Landau-Zener mechanism is rather weak and heats the system rather homogeneously [17]. Therefore, it does not prevent thermal equilibration so much. For that reason, we will focus our considerations on nuclear fission.

II. MICROSCOPIC SCENARIO

In order to base our study on a solid and reliable foundation, we chose statistical mechanics on the microscopic level as the starting point [18]. Each one of the two nuclei is not completely isolated but in contact with the other nucleus. In this sense, the other nucleus acts as a sort of heat bath. However, there are two important differences to a heat bath in canonical thermodynamics: First, the temperature of the other nucleus, acting as a kind of heat bath, fluctuates, as mentioned above, and, second, the energy reservoir of that nucleus is finite.

On the exact microscopic level, we may formulate the problem as follows: While the complete system, consisting of the two nuclei, is isolated, the two nuclei are allowed to exchange between themselves (i) energy, e.g., by collisions between nucleons in the contact region or coupling of the two nuclei to collective modes of the whole system, and (ii) energy and nucleons, if transfer of protons and neutrons between the two nuclei is considered in addition. In fact, transfer of nucleons is assumed to be the predominant mechanism for energy transport due to the large nucleonic mean free path.

If complete knowledge is assumed of all nuclear states and their single-particle configurations,[1] it is possible to calculate the evolution of the system, allowing for energy transfer between the two nuclei, with the Monte Carlo technique: Starting from an initial configuration, the system, consisting of the two nuclei in contact, may change to any other configuration, each one with the same probability, respecting the condition of fixed total energy. Most often, the changes of the configurations proceed in the two nuclei separately according to Bohr's compound-nucleus hypothesis [19]. However, the thermal contact allows also for a coupling. In the independent-particle picture, the amount of energy transferred by nucleon transfer is limited by the energies of the available particles and holes. There are only very few configurations with holes well below the Fermi level and particles well above, because this would imply that a large fraction of energy is stored in only few degrees of freedom.

[1]In the rest of this article, the explicit formulation is done in the independent-particle picture because it is more transparent. Inclusion of residual interactions, e.g., pairing correlations, would not change the final conclusions of the specific questions studied in this chapter.

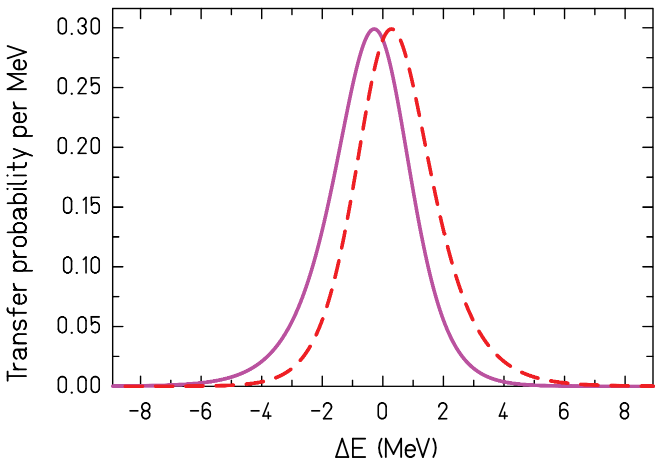

FIG. 1. (Color online) Probability function for energy transfer ΔE between two nuclei with $T_1 = 1$ MeV and $T_2 = 0.7$ MeV in thermal contact by the transfer of one nucleon. (Solid line) Change of excitation energy ΔE_1 of the first nucleus by the transfer of one nucleon from the first to the second nucleus. (Dashed line) Change of excitation energy ΔE_2 of the second nucleus by the transfer of one nucleon from the second to the first nucleus.

The transition to every final state of the complete system which respects the conservation of total energy is equally probable. The average rate of transitions that change the partition of excitation energy between the two nuclei is proportional to the thermal coupling between the two nuclei in contact. In nuclear fission, the thermal coupling would be realized by the neck region. Let us stress that including the transfer of nucleons through the neck does not impose any additional difficulty: The configurations of the nuclei with one neutron or proton more, respectively less, should be included in the number of possible final configurations.

The results of a schematic model calculation of the energy transfer due to nucleon exchange through the neck are shown in Figs. 1 and 2. As an example, a constant single-particle

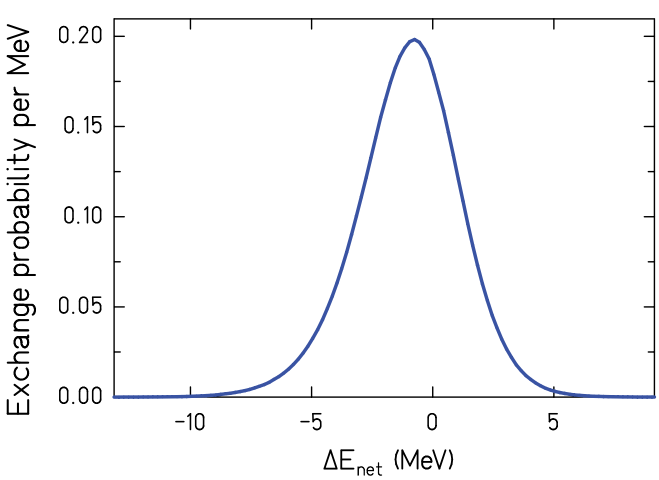

FIG. 2. (Color online) Probability function for net energy transfer between two nuclei in contact with $T_1 = 1$ MeV and $T_2 = 0.7$ MeV by the exchange of one nucleon. The curve shows the net change ΔE_{net} of the excitation energy of the first nucleus.

level density was assumed with an occupation probability as a function of single-particle energy ε given by the Fermi-Dirac distribution:

$$f(E) = \frac{1}{\exp\left(\frac{\varepsilon}{T}\right) + 1}, \tag{3}$$

where ε is counted relative to the Fermi surface, which is assumed to be identical in both nuclei. In this simple case, the result of the calculation can be formulated analytically. The probability of energy transfer by nucleon transfer from a nucleus (named 1) with T_1 to a nucleus (named 2) with T_2 is given by the product of the density f_{p1} of available nucleons in nucleus 1 and the density f_{h2} of holes in nucleus 2:

$$f_{12} = f_{p1} \times f_{h2}, \tag{4}$$

which are expressed by

$$f_{p1}(E) = \frac{1}{\exp\left(\frac{\varepsilon}{T_1}\right) + 1} \tag{5}$$

and

$$f_{h2}(E) = 1 - \frac{1}{\exp\left(\frac{\varepsilon}{T_2}\right) + 1}. \tag{6}$$

The probability of energy transfer by nucleon transfer from the nucleus 2 to the nucleus 1 is given in an analogous way. Figure 1 shows the two distributions of the energy transferred by these two processes for $T_1 = 1$ MeV and $T_2 = 0.7$ MeV. Transfer of nucleons in either direction on the average transports excitation energy from the hotter to the colder nucleus. The mean energy transported by the transfer of one nucleon was determined numerically to about 0.48 MeV in the case of the given example. The exchange of a nucleon (the transfer of one nucleon from nucleus 1 to nucleus 2 plus the transfer of one nucleon from nucleus 2 to nucleus 1), which preserves the masses of the nuclei, is connected with an energy transfer shown in Fig. 2. The mean energy transfer for one nucleon exchange amounts to 0.96 MeV and the standard deviation of the energy-transfer distribution amounts to 3.0 MeV for the given example. This model calculation can easily be run with any single-particle level density, even with any sequence of individual levels, if it is performed with the Monte Carlo technique.

Note that, to be fully realistic, we should have made a calculation using the exact occupation functions, corresponding to the specific states of the two nuclei involved in each nucleon-transfer process. In other words, instead of sampling the diffusion of energy by nucleon exchange between the two nuclei from a single representative distribution depicted in Fig. 2, we should have sampled the individual amounts of energy transfer from different distributions. However, due to the central-limit theorem, the result is essentially the same after a few nucleon-exchange steps, if the first two moments of the unique representative distribution are equal to the average values of the first two moments of the individual distributions involved. By the averaging of the individual occupation functions over an energy interval in the order of the standard deviation of the representative unique distribution, nuclear-structure effects are reduced while preserving the first two moments on the average, and the effective nuclear properties approach those of the Fermi-gas model. This means that the shape of the averaged occupation function approaches the Fermi-Dirac distribution, supporting the validity of the schematic calculation behind Figs. 1 and 2. A more detailed discussion on the averaging over the properties of individual nuclear levels and its consequences for the thermodynamical behavior of the dinuclear system will be given in the next chapter.

All these considerations prove that the statistical evolution of the system is well defined, and it is even possible to predict it precisely with a realistic model. Even if the application of standard thermodynamical methods may be questionable, e.g., due to the fluctuation of the temperatures of the two nuclei involved as a function of energy partition, the application of statistical mechanics poses no problem at all. Thermal averaging or the thermodynamical limit are not required.

III. THERMAL AVERAGING

In the light of the scenario outlined in the previous chapter, the possible application of thermodynamics to the configuration of two nuclei in thermal contact may be reconsidered. The heat flow *dE/dt* between macroscopic objects, which is a continuous quantity, is governed by the temperature difference $\Delta T = T_2 - T_1$ and the thermal resistance R_T against energy exchange:

$$\frac{dE}{dt} = \frac{\Delta T}{R_T}. \tag{7}$$

However, Eq. (7) only describes the average behavior of the dinuclear system because the energy exchange between the two nuclei in contact in the scenario considered above proceeds in steps of considerable magnitude, as shown in the previous chapter. One nucleon exchange may increase or decrease the excitation energy of one and the other nucleus by an amount, which is in the order of the energy range where single-particle levels are partly filled. This range is at least in the order of one MeV, even at low excitation energies due to pairing correlations. The transferred energy is a considerable fraction of the total excitation energy of the system. Therefore, the process of energy exchange cannot be considered as a continuous process, but it proceeds in rather large and fluctuating steps.

At the first glance, this seems to complicate the application of thermodynamics to the dinuclear system even more, but the contrary is true: The transfer of energy between the two nuclei in steps causes an averaging of the thermodynamic properties of the two nuclei. After some steps of energy transfer, the resulting thermal driving force corresponding to the entropy gradient averages over a considerable energy region, which is given by the magnitude of the energy steps. This thermal averaging smoothes out the fluctuations of the microcanonical temperature defined by Eq. (2) in the region where the nuclear level density is continuous but subject to fluctuations. Even more, in the region of discrete, not overlapping, levels, this effect averages over the levels in a finite interval. Note that any of the discrete levels of the nucleus with the lower excitation

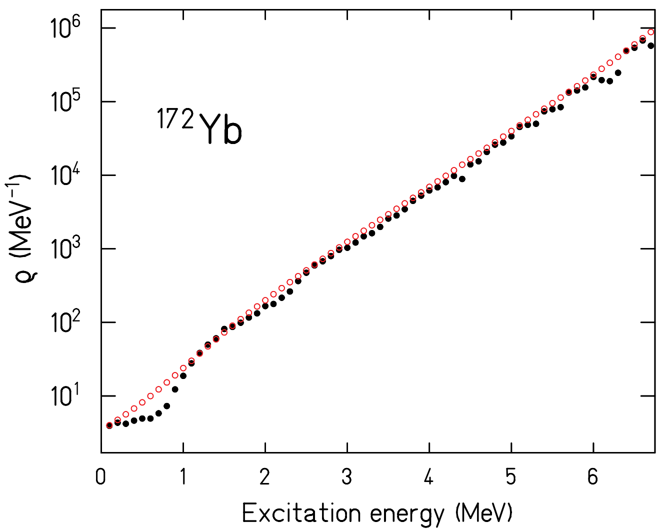

FIG. 3. (Color online) The measured level density [29] of ^{172}Yb (full symbols) is compared with the result of a smoothing procedure (open symbols). The logarithm of the level density was convoluted with a Gaussian function with a standard deviation corresponding to the temperature T of this nucleus. The value of $T = 0.57$ MeV was deduced from the level density around $E = 5$ MeV according to Eq. (2). Border effects were avoided by extrapolating the logarithm of the level density to both sides, below 0.1 MeV and above 6.7 MeV, by a linear fit to the measured data before convolution.

energy is accessible if the partner nucleus with the higher excitation energy is in the regime of overlapping levels. Thus, the effective temperatures of the nuclei, which drive the process of energy exchange, can well be evaluated by Eq. (2). However, instead of the true level density ρ, a smoothed level density should be inserted into Eq. (2), which results from averaging the measured nuclear level density ρ over a finite energy interval.

The magnitude of thermal averaging is illustrated in Fig. 3, where the measured level density of ^{172}Yb is compared with the result of a smoothing procedure. The smoothing procedure simulates the thermal averaging due to the finite magnitude and the fluctuations of the energy transfer between two nuclei in thermal contact. As a conservative estimate, the smoothing was performed using a Gaussian function with a standard deviation equal to the average nuclear temperature $T = 0.57$ MeV as given by the inverse of the global logarithmic slope of the level density. Thus, the standard deviation of the smoothing function is a factor of 4 narrower than the distribution of the individual energy-exchange values. Even with this rather weak smoothing, the structures in the measured level density around 1.2 and 2.4 MeV, corresponding to the first quasiparticle excitations, which are also typical for other nuclei, are completely washed out. This result reveals that the averaging wipes out nuclear-structure effects to a large extent and, thus, the application of the Fermi-Dirac occupation function in our schematic calculation described in the preceding chapter seems to be fully justified.

Finally, we conclude that the process of energy transfer is governed by a differential equation similar to Eq. (7), however, with an additional fluctuating term F:

$$\frac{dE}{dt} = \frac{\Delta T}{R_T} + F. \tag{8}$$

This fluctuating term introduces a random behavior of the energy transfer and, thus, induces a thermal averaging as described above. One may obtain an estimate of the relative importance of the two terms in Eq. (8) by considering the exchange of two nucleons, either protons or neutrons, between the two nuclei in contact as the main mechanism of energy transfer. From a set of numerical calculations as the one described in Sec. II using different temperature values, we deduced that the mean energy transfer in one nucleon-exchange process is generally in the order of 3 times the temperature difference $(T_1 - T_2)$:

$$\frac{dE}{dt} \approx 3\frac{dn}{dt}\Delta T + F, \tag{9}$$

where dn/dt is the nucleon-exchange rate. Equation (9) defines the thermal resistance R_T, introduced in Eq. (7): We find that the thermal resistance is roughly equal to 1/3 of the inverse of the nucleon exchange rate. Extending the transport-theoretical considerations of Randrup [20], who derived the nucleon-transfer rate in a di-nuclear system in thermal equilibrium, to the present case of different temperatures T_1 and T_2, we arrive at the following expression for the nucleon-exchange rate dn/dt:

$$\frac{dn}{dt} = \frac{1}{2}\left(\sum_{i=n,p}\int_{-\infty}^{\infty}\omega_i(\varepsilon)K_i(\varepsilon)f_{12}(\varepsilon)v_{\mathrm{bar}}(\varepsilon)F_{\mathrm{geo}}d\varepsilon + \sum_{i=n,p}\int_{-\infty}^{\infty}\omega_i(\varepsilon)K_i(\varepsilon)f_{21}(\varepsilon)v_{\mathrm{bar}}(\varepsilon)F_{\mathrm{geo}}d\varepsilon\right), \tag{10}$$

where $\omega_i(\varepsilon)$ is the neutron n, respectively proton p, single-particle level density. $K_i(\varepsilon)$ is the transmission coefficient that considers the effect of a potential barrier between the two nuclei (see Fig. 16 of Ref. [21]). f_{12} and f_{21} are given by Eq. (4) and its implicitly described counterpart, respectively. $v_{\mathrm{bar}}(\varepsilon)$ is the normal component of the mean nucleon velocity, and F_{geo} is an energy-independent geometrical factor, which is directly proportional to the cross section of the neck and inversely proportional to the volume of the initial nucleus. Since the factors f_{12} and f_{21} strongly favour contributions in the vicinity of the Fermi energy E_F and $T_1;T_2 \ll E_F$, the nucleon velocity $v_{\mathrm{bar}}(\varepsilon)$ may be approximated by the normal component of the Fermi velocity v_F, and $\omega_i(\varepsilon)$ and $K_i(\varepsilon)$ can be replaced by their values $\omega_i(E_F)$ and $K_i(E_F)$ at the Fermi energy. Thus, Eq. (10) reduces to:

$$\frac{dn}{dt} \approx \frac{1}{2}v_F F_{\mathrm{geo}}\left(\sum_{i=n,p}\omega_i(E_F)K_i(E_F)\int_{-\infty}^{\infty}f_{12}(\varepsilon)\,d\varepsilon + \sum_{i=n,p}\omega_i(E_F)K_i(E_F)\int_{-\infty}^{\infty}f_{21}(\varepsilon)\,d\varepsilon\right). \tag{11}$$

Note that the values of the two integrals are identical, in agreement with the fact that the net average mass transfer is zero. Numerical calculations revealed that the nucleon exchange rate according to Eq. (11) is closely proportional to $T_{\mathrm{mean}} = \frac{1}{2}(T_1 + T_2)$, if T_1 and T_2 do not differ by more than a factor of 2. This result is consistent with Randrup's result [20], who found that the nucleon transfer rate in a dinuclear system is exactly proportional to the nuclear temperature under the conditions of thermal equilibrium and zero relative velocity between the two nuclei in contact.

Experimental information on the thermal resistance can be deduced from the even-odd effect in fission-fragment element distributions [22], which shows that only the most asymmetric mass splits where thermal pressure is very strong present a strong even-odd effect. This indicates that the time for complete energy sorting is comparable to the saddle-to-scission time.

The fluctuation term F in Eq. (8) is directly related to the width of the distribution of net energy transfer, an example of which is shown in Fig. 2. The numerical calculations presented in Sec. II showed that the standard deviation of this distribution is in the order of $2(T_1 + T_2)$ in one nucleon-exchange step. This result can easily be understood, as the latter standard deviation is connected to the energy range of partly filled single-particle states near the Fermi level, which is defined by the nucleus temperature. Since the temperature of a heavy nucleus at low excitation energy is in the order of $T = 1$ MeV, the magnitude of the fluctuating term is in the order of $4T = 4$ MeV for one nucleon exchange. This makes clear that the fluctuating term in Eq. (8) is very important. If the masses of the two nuclei in contact do not differ too much, the fluctuating term is even appreciably larger than the mean energy transfer per nucleon exchange. The shape of the amplitude distribution of the fluctuating term is complex, but, in addition to its second moment, its exact shape is not important, since the net transfer after a few nucleon exchange steps will quickly approach a Gaussian function due to the central limit theorem.

IV. EXCITATION-ENERGY SHARING AND LEVEL DENSITY

After having established the validity of usually applied thermodynamic concepts for modeling the process of thermal equilibration in a dinuclear system, we would like to demonstrate that the excitation-energy sharing between the two nuclei in contact in thermal equilibrium strongly depends on the characteristics of the nuclear level density ρ. In most cases, the so-called Fermi-gas level density [23] has been assumed, which is described by

$$\rho(E) \propto \exp(2\sqrt{aE}), \tag{12}$$

where a is the level-density parameter. We have omitted the preexponential factor, which has only little influence on the energy sharing. Since the level-density parameter a is roughly proportional to the nuclear mass A, the excitation energy is divided in proportion to the masses of the reaction products [3,24].

At low excitation energies, the slope of the logarithm of the nuclear level density is found to vary much less strongly than predicted by Eq. (12). Two descriptions, which have been introduced by Gilbert and Cameron [25], are still widely used (e.g., [26]): the back-shifted Fermi-gas formula

$$\rho(E) \propto \exp[2\sqrt{a(E - E_{bF})}] \tag{13}$$

and the constant-temperature formula

$$\rho(E) \propto \exp\left(\frac{E - E_{bC}}{T}\right). \tag{14}$$

Recent experiments revealed that the nuclear level density ρ up to excitation energies E of about 10 [27] or even 20 MeV [28] are well represented by the constant-temperature formula (14) with a parameter T that essentially does not depend on energy; see for example the full dots in Fig. 3. As illustrated in Fig. 3, after thermal averaging the constant-temperature formula even better describes the effective averaged level density. According to Eq. (2) the parameter T of Eq. (14) coincides with the nucleus temperature. The constant-temperature behavior has been explained by the gradual melting of Cooper pairs in the energy domain governed by strong pairing correlations [29]. It was already mentioned above that for a system of two nuclei in contact, the constant-temperature formula leads to the surprising result that essentially all excitation energy is transported to the heavy nucleus by a process of energy sorting [6]. The excitation-energy sorting is clearly reflected by several fission observables such as the number of prompt neutrons emitted by the fission fragments [6].

When some transfer of nucleons is considered, also the even-odd mass differences have a sizeable influence on the last steps of the energy-sorting mechanism in the regime of a constant-temperature level density. The eventual exchange of one neutron and one proton may transform an odd-odd nucleus in its ground state to an even-even nucleus in its ground state, which is more strongly bound by about two times the pairing gap parameter Δ, assuming the same value for protons and neutrons. This leads to an increase in entropy due to the higher excitation energy available in the other nucleus. A schematic scenario, which explains the complex features of the even-odd effect in fission-fragment nuclear-charge distributions along these lines, has been described in Ref. [22].

The description of the energy division in the case of the back-shifted Fermi-gas formula is more complex. In this case, Eq. (2) leads to the following energy dependence of the nuclear temperature:

$$T \propto \sqrt{\frac{(E - E_{bF})}{A}}. \tag{15}$$

The condition of thermal equilibrium leads to the following implicit equation for the energy division between the two nuclei:

$$\frac{E_1 - E_{bF1}}{E_2 - E_{bF2}} = \frac{A_1}{A_2}. \tag{16}$$

The indeces 1 and 2 stand for the first and the second nucleus, respectively. Since the backshift energy E_{bF} is negative for most nuclei, this leads to a division of excitation energy, which is most often in between the division according to the mass ratio and complete energy sorting. Below a certain threshold of the total energy, which depends on the values of the back-shift energies and nuclear masses in Eq. (16), there is no solution for Eq. (16) and the result is identical with the one of the constant-temperature formula, corresponding to complete energy sorting.

Finally, we would like to stress that the pure Fermi-gas level-density formula [Eq. (12)] is generally not a realistic description of the nuclear level density. Even at higher excitation energies, where the level density is expected to be well represented by the independent-particle model, which is behind the Fermi-gas formula, a considerable back-shift energy is expected according to Eq. (13). It is understood as the consequence of pairing correlations and shell effects. Thus, also in this regime the excitation-energy division according to thermal equilibrium in a dinuclear system follows Eq. (16). The assumption that the excitation energy in thermal equilibrium is shared according to the ratio of the masses is never valid in a strict sense.

V. CONCLUSIONS

The process of thermal equilibration in a di-nuclear system is conventionally described using thermodynamical concepts. In particular, these concepts were applied in the recent discovery of the energy-sorting process in nuclear fission [6]. In the present work, considerations obtained with schematic calculations based on statistical mechanics show that this description is justified in spite of the small size of nuclei. Indeed, the energy transfer between the two nuclei in contact proceeds in steps, mainly by nucleon exchange through the contact zone. The nucleons that take part in the exchange are those with energies close to the Fermi energy. Therefore, the energy transfer leads to a fluctuation of the excitation energy of one and the other nucleus by a considerable amount of the order of $2(T_1 + T_2)$. After a number of steps, the system averages over a considerable energy region, and thus the entropy which determines the dynamical evolution of the system is an average one. Consequently, the irregularities of the level density are smoothed out by the very nature of the energy-exchange process, allowing for the definition of a nuclear temperature. Finally, the dependence of the excitation energy division on the expression used for the level density was discussed.

ACKNOWLEDGMENTS

This work was supported by the EURATOM 6. Framework Program "European Facilities for Nuclear Data Measurements" (EFNUDAT), Contract No. FP6-036434. K.-H.S. thanks the CENBG for warm hospitality. The anonymous reviewer is thanked for his constructive comments.

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