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THE ROLE OF THE DINUCLEAR SYSTEM
IN THE PROCESSES OF NUCLEAR FUSION,
QUASI-FISSION, FISSION
AND CLUSTER FORMATION*

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Роль двойной ядерной системы в процессах слияния ядер, квазиделения, деления и формирования кластеров

Слияние ядер, квазиделение, деление, формирование кластеров в возбужденных ядрах рассматриваются как процессы формирования и эволюции двойной ядерной системы. Этот подход позволяет выявить новые аспекты полного слияния ядер, показать, что квазиделение играет важную роль в ядерных реакциях, используемых для синтеза сверхтяжелых элементов. Дана качественная картина деления возбужденного ядра и показана важная роль формирования кластеров в этом ядерном процессе.

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The Role of the Dinuclear System in the Processes of Nuclear Fusion, Quasi-Fission, Fission and Cluster Formation

The nuclear fusion, quasi-fission, fission and cluster formation in an excited nucleus are considered as the processes of the formation and evolution of the dinuclear system. This approach allowed one to reveal new aspects of nuclear fusion, to show that quasi-fission plays an important role in nuclear reactions used to synthesise superheavy elements. A qualitative picture is given of the fission process of an excited nucleus and an important role of cluster formation in this process is shown.

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

1 Introduction

This report presents a non-traditional approach to describing nuclear processes accompanied by a deep rearrangement of the structure of nuclei such as nuclear fusion, quasi-fission, fission, emission of clusters. The main content of these nuclear processes is the formation and evolution of the dinuclear system.

Researches on reactions with low-energy heavy ions revealed two new objects in the nuclear microworld: nuclear molecules and dinuclear systems (DNS). The idea of DNS was originally proposed while deep inelastic transfer reactions (DITR) were studied [1]. The properties of those reactions were successfully described by assuming that a nuclear complex, a dinuclear system, is formed after the collision kinetic energy being completely dissipated. DNS differs from nuclear molecules in that its states are unequilibrium. DNS evolves in time by transferring nucleons from one nucleus to the other. The evolution of DNS is of a statistic nature and proceeds along a set of trajectories in the Z-A space of the nuclei which DNS is composed of. The evolution is governed by the system's potential energy, which is a function of its charge (mass) asymmetry and angular momentum of collision. An essential feature of the DNS's evolution is the fact that the nuclei that compose DNS retain their individuality, which is due to their having shell structure.

DITR allow us to obtain unique information on the interaction of two nuclei that happened to be in close contact after the collision kinetic energy being completely dissipated. In DITR two simultaneous processes occur: one is the evolution of DNS along the system's charge (mass) degree of freedom, the other one is the system's decay from intermediate states. The study of the charge, mass, energy and angular distributions of DITR products gives us quite a good idea of DNS's evolution. It can be said that DITR are open nuclear reactions. On the contrary, nuclear fusion, quasi-fission, fission, emission of clusters are closed reactions. In those reactions, experimenters only observe the end products, the nuclear process itself having already been completed by that moment. Those may be, for example, the decay products of an excited compound nucleus, which, as it is known, does not remember the history of its formation.

The basic idea of the new approach is the assumption that the formation and evolution of DNS is the universal way for Nature to carry out a deep rearrangement of a nucleus or nuclei in the processes of nuclear fusion, quasi-fission, fission, emission of clusters [2]. That is the reason why this approach was called the Dinuclear System Concept (DNSC).

2 Nuclear fusion and quasi-fission within the framework of DNSC

2.1 Nuclear fusion scenario

According to DNSC, nuclear fusion occurs in the following way [3]. At the capture stage, after the collision kinetic energy has completely been dissipated, a dinuclear system is formed. The fusion of nuclei is an evolutionary process in which nucleons of one nucleus are gradually, shell-by-shell transferred to the other nucleus. The nuclei of DNS retain their individuality all the way to the compound nucleus. Figure 1 shows what the fundamental differences are between the picture of complete nuclear fusion that is provided by DNSC and the popular macroscopic dynamic model (MDM) of Swiatecki [4, 5]. The latter model treats real nuclei, which are built from nucleons and have shell structure, as drops of viscous nuclear liquid. After the surfaces of the drops have come into contact the individuality of the nuclei disappears rapidly as a result of a neck being formed, and a strongly deformed mononucleus comes into existence. Overcoming the nuclear friction, the mononucleus evolves in deformation space towards a compound nucleus, which is of a more compact form. Within the framework of DNSC, the nuclei of DNS retain their individuality all the way the system evolves toward the compound nucleus.

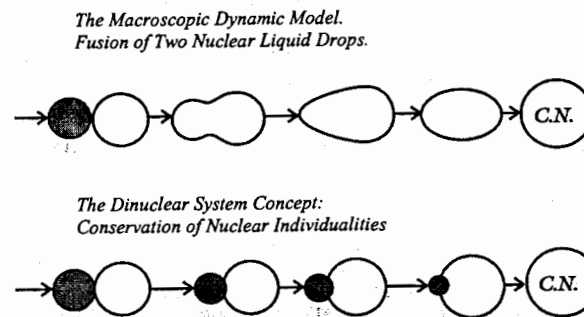


Fig. 1. Nuclear fusion from the viewpoint of the macroscopic dynamic model and the dinuclear system concept.

2.2 Peculiarities of the fusion of massive nuclei that were revealed by DNSC

DNSC allowed one to reveal two peculiarities of the complete fusion of massive nuclei: a) there is a specific inner fusion barrier B_{fus}^* and b) there is competition between the complete fusion channel and quasi-fission channel in DNS, formed at the capture stage.

As is known from the study of DITR, the evolution of DNS is directed by the system's potential energy, considered as a function of its charge (mass) asymmetry and angular momentum of collision $V(Z, L)$. Fig. 2 shows the potential energy of DNS that is formed in the reaction $^{110}\text{Pd}+^{110}\text{Pd}$. $V(Z, L)$ includes Coulomb, nuclear and centrifugal potentials [6]. On formation, a DNS is similar to a gigantic nuclear molecule. Its potential energy is at a minimum. For complete fusion to occur, the evolving DNS has to overcome a potential barrier. The barrier was called "inner fusion barrier" and denoted by B_{fus}^* [6]. The asterisk means that the energy to overcome the barrier is taken from the DNS's excitation energy E^* . Two massive nuclei can fuse to form a compound nucleus if the condition $E^* > B_{fus}^*$ is met.

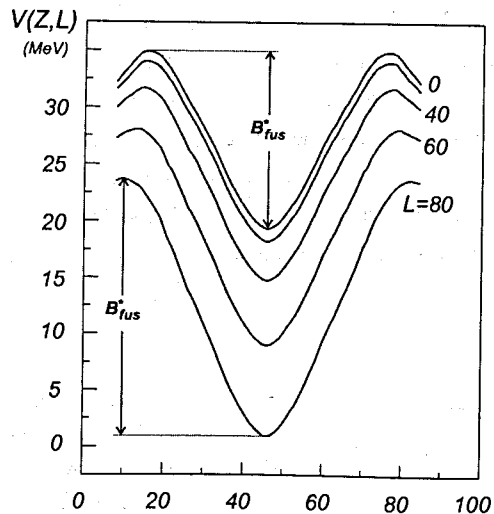


Fig. 2. Potential energy of DNS formed in the reaction $^{110}\text{Pd} + ^{110}\text{Pd}$. Z is the atomic number of one of DNS's nuclei, L the collision angular momentum. The inner fusion barriers B_{fus}^* are indicated. The compound nucleus's potential energy is taken to be 0.

In asymmetric nuclear reactions, there are two ways for DNS to evolve (Fig. 3). One of them leads to a compound nucleus. To follow this way, DNS has to overcome the barrier B_{fus}^* . The other way leads to the symmetric form of the system. Following this way, DNS meets with no obstacles. After DNS has reached the symmetric form, its potential energy is at a minimum. Once DNS is symmetric, the Coulomb repulsion between the nuclei of DNS reaches its maximum, and the system breaks up into two fragment nuclei of approximate by equal masses: quasi-fission results. For quasi-fission to occur, DNS has to overcome a barrier in the nucleus-nucleus potential: the quasi-fission barrier B_{qf} . The evolution of DNS being of a statistic nature results in competition between the complete fission channel and quasi-fission channel.

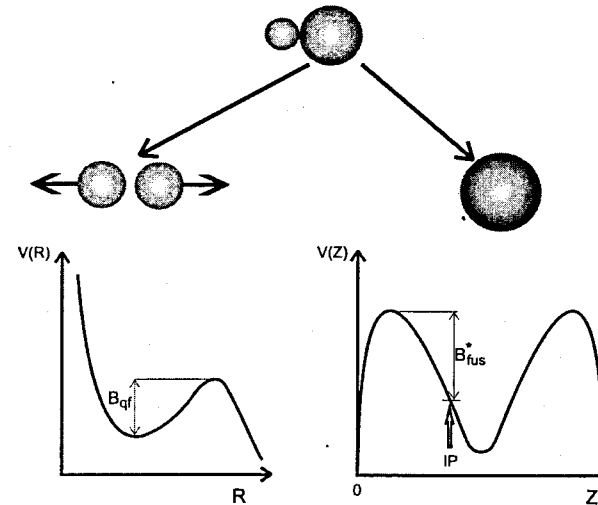


Fig. 3. Two ways of the evolution of a massive asymmetric DNS, $V(R)$ is nucleus-nucleus potential, $V(Z)$ – the potential energy of DNS, Z – the atomic number of one of DNS's nuclei, B_{qf} – the quasi-fission barrier, B_{fus}^* – the inner fusion barrier, I.P. – the reaction injection point.

The model of complete fusion-quasi-fission competition was first created for symmetric nuclear reactions between massive nuclei [6]. In those reactions, both the potential energy $V(Z, L)$ and nucleus-nucleus potential $V(R)$ of DNS just formed are at a minimum ("potential pocket"), that is, DNS is in a quasi-equilibrium state. The relationship between the complete fusion channel and quasi-fission channel was assumed to be determined by the densities of DNS's states on the tops of the barriers B_{fus}^* and B_{qf} : $\rho_{B_{fus}^*}$ and ρ_{qf} . The probability of a compound nucleus being formed after capture has occurred P_{cn} was described by the relationship:

$$P_{cn} = \frac{\rho_{B_{fus}^*}}{\rho_{B_{fus}^*} + \rho_{qf}} \quad (1)$$

The density of DNS's states was calculated according to the model proposed in [7].

Fig. 4 shows the production cross sections σ_{ER} for the evaporation residues in the reaction $^{110}\text{Pd}+^{110}\text{Pd} \rightarrow ^{220}\text{U}$ calculated using the model [6]. As is seen, for the collision energies above the Coulomb barrier, the calculated results and experimental data are in quite a good agreement. MDM-based calculations give much larger values for σ_{ER} . The discrepancy of four orders between the calculations done according to the two models is due to the fact that MDM does not account for competition between complete fusion and quasi-fission, which is the dominant channel in this reaction.

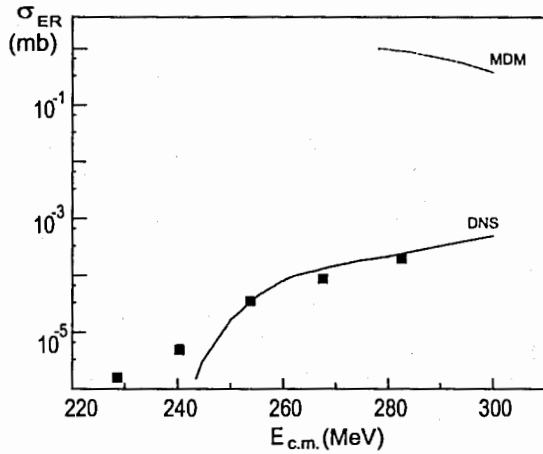


Fig. 4. Cross sections for the evaporation residues σ_{ER} in the reaction $^{110}\text{Pd} + ^{110}\text{Pd}$ as a function of bombarding energy. The closed squares represent the experimental data from [8], the solid curve – the σ_{ER} calculations using DNSC, the dashed curve – the σ_{ER} calculations using MDM.

2.3 Analysis of fusion reactions used to synthesise transuranium and superheavy elements

DNSC allows one to carry out a realistic analysis of nuclear reactions used to synthesise novel transuranium (TUE) and superheavy elements (SHE). Over many years, the fission of the excited compound nucleus has been the main problem in the synthesis of novel TUE. The production cross section σ_{ER} for TUE was defined by the relationship:

$$\sigma_{ER} = \sigma_c W_{sur} \quad (2)$$

where σ_c is the capture cross section, W_{sur} – the probability for a compound nucleus to survive during deexcitation. σ_c was usually calculated by the optical model. To calculate W_{sur} , the statistical model was used. As the Z of a compound nucleus increases, W_{sur} decreases, the decrease being drastic enough. However as the SHE region is approached, fission barriers show an increase in value, which gave hope that SHE production cross sections would not turn out to be vanishingly small. Cross sections were calculated for elements 104, 108 and 110 in cold fusion reactions accompanied by emission of one and two neutrons from a compound nucleus [9]. The calculated cross sections were found to increase as the atomic number of a compound nucleus increases, which is accounted for by the influence of the closed proton shell at $Z=114$. For element 104, it was found that the results of calculations agreed satisfactorily with experimental data, whereas for 108 and especially for element 110, there was a dramatic discrepancy. Experimental data proved to be 3-5 orders smaller in value than calculated data. This discrepancy is not accountable for within the

framework of traditional models of complete fusion. It may have been assumed that the decrease in the cross section is due to quasi-fission. However none of the traditional models of nuclear fusion was capable of assessing its influence.

Within the framework of DNSC the production cross section for TUE and SHE is defined by the relationship:

$$\sigma_{ER} = \sigma_c P_{cn} W_{sur} \quad (3)$$

which, in addition to the factors σ_c (DNS production cross section) and W_{sur} , includes the factor P_{cn} , the probability of a compound nucleus being formed after capture. The factor P_{cn} just accounts for competition between the channels of complete fusion and quasi-fission in DNS formed at the capture stage. The advantage of DNSC over the traditional models of complete fusion lies in the fact that it allows one to create models of competition between complete fusion and quasi-fission that enable the factor P_{cn} to be calculated.

2.4 Models of competition between complete fusion and quasi-fission for asymmetric nuclear reactions

Created were two DNSC-based models of competition between complete fusion and quasi-fission in asymmetric nuclear reactions. The first model used the Monte Carlo technique to describe the evolution of DNS [10]. Some simplifying assumptions were made concerning the evolution of DNS. DNS was assumed to be capable of going only to the system's configurations neighbouring by Z and N . That is, the system evolves by transferring a proton and one or two neutrons from nucleus to nucleus, no cluster transfer occurs. The transfer probability was believed to be proportional to the density of DNS's states in the neighbouring configurations. The set of the trajectories along which DNS evolves in the Z - A space was reduced to one trajectory running across the valley of the system's potential energy. After getting over the barrier B_{fus}^* , DNS was believed to finish its evolution by a compound nucleus being formed. If evolving DNS had happened to achieve a symmetric form, it went into the quasi-fission channel.

The model was used to analyse four nuclear reactions of different initial charge and mass asymmetry that result in producing the same compound nucleus of ^{246}Fm [11]. Fig. 5a presents the potential energy of DNS formed [10]. As is seen, the inner fusion barrier is only several MeV for the reaction $^{40}\text{Ar} + ^{206}\text{Pb}$, whereas it increases up to 20 MeV for the reaction $^{136}\text{Xe} + ^{110}\text{Pd}$. Fig. 5b shows the values of P_{cn} as a function of the collision angular momentum that were calculated using this model. $P_{cn} \sim 5 \times 10^{-1}$ for the

reaction with ^{40}Ar , whereas P_{cn} falls down to 5×10^{-4} for the reaction with ions of ^{136}Xe . The calculated values of P_{cn} allowed to describe the experimental data for the ^{244}Fm production cross sections in reactions with ions of ^{40}Ar and ^{76}Ge [10] as well as to understand the reason why ^{244}Fm had not been found in reactions with ions of ^{86}Kr and ^{136}Xe [11].

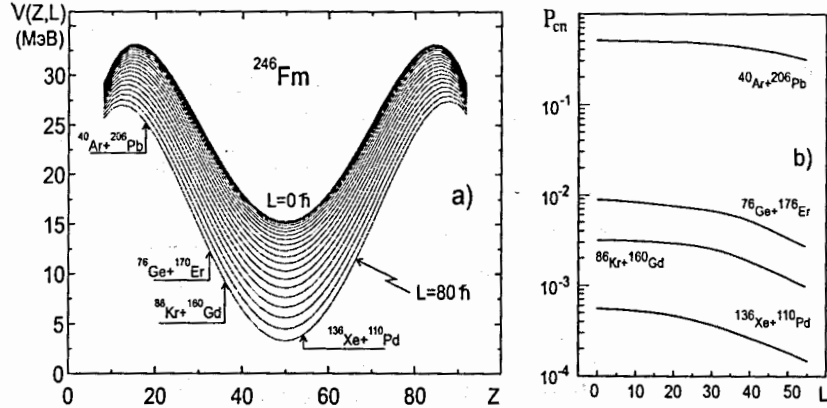


Fig. 5. a) Potential energy of DNS formed in four reactions, the same compound nucleus of ^{246}Fm being formed. b) Production probability for the compound nucleus of ^{246}Fm as a function of collision angular momentum.

Within the second model [12] the evolution and decay of DNS is considered as a process of diffusion that is described by two collective variables η and R . η characterises the system's mass asymmetry: $\eta = (A_1 - A_2)/(A_1 + A_2)$, where A_1 and A_2 are the mass numbers of the colliding nuclei. R is the distance between the centres of the system's nuclei. Diffusion along η leads to a compound nucleus being formed, diffusion along R to quasi-fission. Within the framework of this model, DNS may go into the quasi-fission channel not only from the symmetric configuration but also from the asymmetric configurations that precede it. To describe diffusion, the quasi-stationary two-dimensional solution to the Fokker-Planck equation and Kramers's approach were used. It is the quasi-stationary probability fluxes through the potential barriers B_{fus}^* and B_{qf} that define the probability of a compound nucleus being formed after capture occurs:

$$P_{\text{cn}} = \frac{\lambda_{\text{fus}}^{\text{Kr}}}{\lambda_{\text{fus}}^{\text{Kr}} + \lambda_{\text{qf}}^{\text{Kr}}} \quad (4)$$

The model was tested by applying to nuclear reactions for which the cross sections of evaporation residues σ_{ER} were measured experimentally and the values of W_{sur} were known. Then the model was used to calculate the values of P_{cn} for reactions used to synthesise TUE and SHE. The values of P_{cn} were calculated for the reactions of the cold fusion in the synthesis of elements 102-114 (Fig. 6) [13]. P_{cn} is seen to drop rather rapidly as the Z of the nucleus produced increases. P_{cn} is 5×10^{-2} for element 104, whereas it drops down to 1×10^{-6} for element 112. This drop is in agreement with the experimental data obtained in the cold fusion reactions used to produce heaviest nuclei. For example, the production cross section for 110 element was 12 pb and that for 112 was 1 pb [14]. P_{cn} drops by a factor of 10 from element 110 to element 112. The calculations of the values of P_{cn} made within the framework of DNSC testify that in cold fusion reactions, quasi-fission is a decisive factor that makes the production cross section of SHE decrease as their atomic number increases.

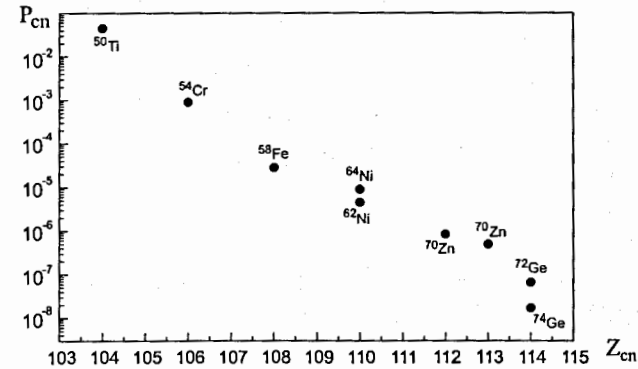


Fig. 6. Production probability for a compound nucleus in cold fusion reactions (H.I., 1n) used to synthesise TUE and SHE. Z_{cn} is the atomic number of the compound nucleus, the bombarding ions are indicated.

In warm fusion reactions using ions of ^{48}Ca , quasi-fission also rather strongly affects the production cross section of heavy elements, which results in its decrease. Fig. 7 presents the values of P_{cn} for the reactions ^{232}Th , ^{238}U , $^{244}\text{Pu}+^{48}\text{Ca}$ as a function of the excitation energy of the compound nucleus [15]. DNSC revealed a danger previously unknown that is waiting for experimenters on the way to the synthesis of spherical SHE. It is the danger of quasi-fission [16].

Concluding this part, I would like to draw attention to the fact that DNSC puts quite a clear physical interpretation of complete fusion and quasi-fission processes. Indeed, it is impossible for two colliding nuclei to penetrate deeply into one another at low energies. Such a collision results in an overlap between only some part of their surface

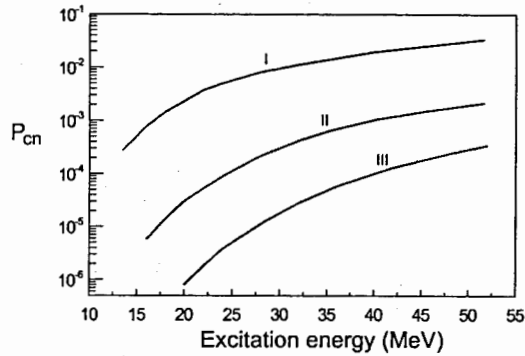


Fig. 7. Production probability for the compound nuclei of elements 110, 112 and 114 as a function of the excitation energy of the compound nucleus for targets of ^{232}Th (I), ^{238}U (II), ^{244}Pu (III) and ions of ^{48}Ca .

layers. That overlap was assessed to be only (5-7)% of each nucleus's volume, which allows the interacting nuclei to keep their individuality. At the same time, the overlap zone serves as a window through which to transfer weakly binded nucleons of one nucleus's upper shell to the other nucleus's free levels. If nucleons are transferred from the lighter nucleus to the heavier one, the process results in an excited compound nucleus being formed. If nucleons are transferred in the opposite direction, a symmetric DNS is formed that breaks up into two fragment nuclei under Coulomb forces, that is, quasi-fission occurs.

3 A qualitative picture of the fission of an excited nucleus within the framework of DNSC and the role of clusters

The complete fusion of two nuclei and the fission of a nucleus may be considered as forward and reverse nuclear processes, and this implies that they are to go through similar intermediate states. If it is true, then fission has to involve the formation and evolution of DNS [17].

The potential energy of a massive DNS looks as shown in Fig. 2. It has two minima: one corresponds to the compound nucleus and the other to the symmetric shape of the system. The inner fusion barrier B_{fus}^* divides the potential energy into two regions, which we will denote by I and II, region I being on the left side. Let us assume DNS has been able to form in region I and get over the barrier B_{fus}^* , into region II. In this region DNS evolves according to the above-stated quasi-fission scenario: nucleons are transferred from the heavier nucleus to lighter one, the system becomes symmetric and in the end breaks up in two fragment nuclei of approximately equal masses.

However, how is DNS produced in an excited nucleus, and how does it succeed in reaching the top of B_{fus}^* and getting over it?

In an attempt to answer this question we should take advantage of Nature's hint and consider the phenomenon of emitting clusters, the nuclei of light elements, from excited nuclei. It was found that, at excitation energy of several tens of MeV, intermediate and heavy nuclei emit the nuclei of light elements from ^4He to the nuclei of magnesium, silicon, sulphur and even heavier elements [18]. But before escaping from the mother nucleus, a cluster has to be formed as an independent nucleus. Based on the Pauli exclusion principle, the only place at which this event may occur is the surface of the mother nucleus. But two nuclei that are in close contact and intensively interact with one another just constitute DNS.

Experiments show that various clusters of different A and Z escape from excited nuclei. But this means that an excited nucleus constantly goes from one cluster state to another, from one DNS to another. An excited nucleus does not prove to be a monotonical system something like a bag of nucleons. An excited nucleus has a variety of cluster states. On its surface, the nuclei of light elements are formed, grow and disappear again.

An excited compound nucleus is in a statistic equilibrium state. The time it is in one or another cluster configuration is proportional to the system's state density for this configuration. Using the statistic model, work [19] succeeded in describing an experimentally observed yield of various clusters. Thus, there is no problem for DNS to be formed in an excited nucleus. It can be stated that DNS is the form in which an excited nucleus exists.

For the top of the B_{fus}^* to be reached, quite a massive cluster has to be formed in the excited nucleus, which is done at the sacrifice of the compound nucleus's excitation energy and results in its being cooled. This in turn makes the density of the system's states drop, which means that it may take a time considerable on nuclear scale to form a critical cluster, i.e. a critical DNS after the production of which the systems goes from region I to region II. It is known that the fission time for reactions with heavy ions is one order greater than the quasi-fission time: several unities times 10^{-19} s as against several unities times 10^{-20} s [20]. This difference appears to testify that the production of a critical cluster configuration, a critical DNS, takes most of the fission time.

Substitution of real nuclei with drops of a uniform viscous nuclear liquid allows one to consider nuclear fission only as a process occurring in deformation space. Based on

the fact that nuclei consist of neutrons and protons, and have shell structure, it should be admitted that the process of the formation and evolution of DNS is the most natural way for an atomic nucleus to break up into two fragment nuclei.

So, leaning on DNSC, we have drawn a picture of nuclear fission different to the traditional one. To develop this approach further, there is the need to create theoretical models capable of quantitatively describing the various aspects of this fundamental nuclear process.

4 Summary

Proposed is a new approach to interpretation and description of nuclear processes, which are characterised by a deep rearrangement of the nucleonic structure of a nucleus or nuclei, such as the fusion of two nuclei, quasi-fission, fission, emission of clusters. The new approach rests on data on the interaction between two nuclei that have happened to be in close contact after the kinetic collision energy being completely dissipated. These data were obtained while studying deep inelastic transfer reactions. According to the proposed approach, the essence of the above-listed nuclear processes is the formation of a dinuclear system and its evolution by transferring nucleons from one nucleus to the other. The new approach allowed us to reveal the unknown peculiarities of the fusion of massive nuclei: the inner fusion barrier B_{fus}^* and competition between the complete fusion channel and quasi-fission channel in a dinuclear system formed at the capture stage. On the basis of this approach, models of competition between complete fusion and quasi-fission in symmetric and asymmetric nuclear reactions were created. With the help of these models, we succeeded in describing experimental data on the production cross sections for the evaporation residues in the fusion reactions of massive nuclei. It was shown that, in the cold fusion reactions used to synthesise superheavy elements, quasi-fission is the main factor that makes the production cross section of a new element decrease as its atomic number increases. A new interpretation is proposed of the fission of an excited compound nucleus. It was revealed how important the formation of clusters is for nuclear fission. On the whole, the proposed approach allows one to draw a clearer and more natural picture of the processes associated with a deep rearrangement of the structure of a nucleus or a pair of nuclei in reactions of nuclear fusion, quasi-fission, fission and emission of clusters.

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