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Constraining the equation of state of nuclear matter from fusion hindrance in reactions leading to the production of superheavy elements

M. Veselsky* and J. Klimo
Institute of Physics, Slovak Academy of Sciences, Bratislava, Slovakia

Yu-Gang Ma
Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China

G. A. Souliotis
Laboratory of Physical Chemistry, Department of Chemistry, National and Kapodistrian University of Athens, Athens 15771, Greece
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The mechanism of fusion hindrance, an effect preventing the synthesis of superheavy elements in the reactions of cold and hot fusion, is investigated using the Boltzmann-Uehling-Uhlenbeck equation, where Coulomb interaction is introduced. A strong sensitivity is observed both to the modulus of incompressibility of symmetric nuclear matter, controlling the competition of surface tension and Coulomb repulsion, and to the stiffness of the density-dependence of symmetry energy, influencing the formation of the neck prior to scission. The experimental fusion probabilities were for the first time used to derive constraints on the nuclear equation of state. A strict constraint on the modulus of incompressibility of nuclear matter $K_0 = 240$–260 MeV is obtained while the stiff density-dependences of the symmetry energy ($\gamma > 1$) are rejected.

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I. INTRODUCTION

In the last two decades of the past century, the heavy elements up to $Z = 112$ were synthesized using cold fusion reactions with Pb, Bi targets in the evaporation channel with an emission of one neutron [1]. The experimentalists had to face a rapid decrease of cross sections down to the picobarn level due to increasing fusion hindrance whose origin was unclear. Since the turn of the millennium, still heavier elements with $Z = 113$–118 were produced in the hot fusion reactions with emission of 3–4 neutrons using ^{48}Ca beams with heavy actinide targets between uranium and californium [2–8]. Again the increase of fusion hindrance was observed, caused by a competition of the fusion process with an alternative process called quasifission. Quasifission occurs when instead of fusion the system forms an elongated shape evolving towards the scission point. The systematics in the reactions with lead and uranium targets, published in [9,10], shows that quasifission sets on for beams ^{48}Ca and heavier. In terms of reaction mechanism, quasifission is similar to the nucleon exchange between colliding nuclei, however it proceeds while the shape of the system also changes dramatically. Compared to the fusion-fission, proceeding via the formation of the compound nucleus, the angular distribution of fission fragments is forward-peaked in the center-of-mass frame, total kinetic energy (TKE) is lower and the mass asymmetry ranges from the mass asymmetry of the projectile-target configuration towards the symmetric mass split, with the yield decreasing monotonously. A large systematics of high quality data on quasifission in the reactions, leading to the production of superheavy elements, was obtained in recent years in Dubna [11], Tokai [12], and Canberra [13]. It is usually considered that the process of quasifission is governed by a complex dynamics of the projectile-target system, which is often described using theoretical tools such as the model of a dinuclear system [14–17] or the Langevin equation [18–21]. Besides the above theoretical tools, the competition of fusion and quasifission was also addressed using the implementations of the Boltzmann equation known as ImQMD [22–25] and using the time-dependent Hartree-Fock theory [26–29]. However, the success of a simple statistical model of fusion hindrance, introduced in [30,31] suggests that the competition of fusion and quasifission could be dominantly driven by the available phase-space and hindrances originating in diabatic dynamics are not decisive. In the present work we employ the Boltzmann-Uehling-Uhlenbeck (BUU) equation with the Pauli principle implemented separately for neutrons and protons and with the Coulomb interaction. We demonstrate how various equations of state of nuclear matter implemented into such a transport simulation influence the competition of fusion and quasifission. Based on available data on reactions, leading to the production of superheavy nuclei, we extract the most stringent constraints on the stiffness of the nuclear equation of state and on the density-dependence of the symmetry energy.

II. SIMULATIONS

In order to describe theoretically the competition of fusion and quasifission at energies close to the Coulomb barrier, the goal is to describe the evolution of the nuclear mean field of the two reaction partners. However, besides nuclear mean field, it is necessary to take into account the electrostatic interaction among protons and also it is necessary to guarantee the preservation of the Pauli principle in a strict way. The evolution of nuclear mean field can be described by solving

*martin.veselsky@savba.sk

the Boltzmann equation. One of the approximations for the solution of the Boltzmann equation, the Boltzmann-Uehling-Uhlenbeck model, is extensively used [32,33], which takes both the nuclear mean field and the Fermionic Pauli blocking into consideration. The BUU equation reads

$$\begin{aligned}\frac{\partial f}{\partial t} &+ v \cdot \nabla_r f - \nabla_r U \cdot \nabla_p f \\ &= \frac{4}{(2\pi)^3} \int d^3 p_2 d^3 p_3 d\Omega \frac{d\sigma_{NN}}{d\Omega} v_{12} \\ &\quad \times [f_3 f_4 (1-f)(1-f_2) - f f_2 (1-f_3)(1-f_4)] \\ &\quad \times \delta^3 (p + p_2 - p_3 - p_4), \end{aligned} \tag{1}$$

where $f = f(r,p,t)$ is the phase-space distribution function. It is solved with the test particle method of Wong [34], with the collision term as introduced by Cugnon *et al.* [35]. In Eq. (1), $\frac{d\sigma_{NN}}{d\Omega}$ and v_{12} are the in-medium nucleon-nucleon cross section and relative velocity for the colliding nucleons, respectively, and U is the sum of the simple single-particle mean field potential with the isospin-dependent symmetry energy term

$$U = a\rho + b\rho^{\kappa} + 2a_s \left(\frac{\rho}{\rho_0}\right)^{\gamma} \tau_z I, \tag{2}$$

where $I = (\rho_n - \rho_p)/\rho$, ρ_0 is the normal nuclear matter density; ρ, ρ_n, and ρ_p are the nucleon, neutron, and proton densities, respectively; τ_z assumes the value 1 for neutron and -1 for proton, the coefficients a, b and exponent κ represent the properties of symmetric nuclear matter, while the last term describes the influence of the symmetry energy, where a_s represents the symmetry energy at saturation density and the exponent γ describes the density dependence.

The in-medium nucleon-nucleon cross sections are typically approximated using the experimental cross sections of free nucleons (e.g., using the parametrization from Cugnon [35]). Alternatively, as shown in the work [36], in-medium nucleon-nucleon cross sections can be estimated directly using the equation of state and used successfully, e.g., to describe the evolution of transverse flow in a wide range of relativistic energies [37]. However, while at low energies close to the Coulomb barrier the collision term still plays some role [28], the choice of the in-medium nucleon-nucleon cross sections does not influence the results of simulations, since at such low relative momenta both choices of cross sections exceed the cutoff value applied in the BUU code.

In order to describe the nuclear collisions close to the Coulomb barrier, it is crucial to implement properly the electrostatic interaction among protons. It is however impossible to introduce a density-dependent term into the single-particle potential in Eq. (1), since electrostatic interaction has a long range and for the infinite nuclear matter it would diverge. In the present work, instead of modification of the single-particle potential, we complement the corresponding density-dependent force $\nabla_r U$ acting at a given cell of the cubic grid (with a mesh of 1 fm) with the summary force generated by the all remaining protons outside of a given cell. This approach thus avoids a fluctuation of the Coulomb force due to interaction of protons at small distances inside the cell, and also avoids double counting since the interaction of protons within the same cell is considered in the collision term. Specifically we consider only the interaction with the protons of the same set of test particles, which allows to perform simulations practically with the increasing the CPU time consumption. This circumstance allowed to perform this study in principle. The effect of the Coulomb force was tested using peripheral and midperipheral collisions of ^{64}Ni + ^{208}Pb and for peripheral collisions at 12 and 16 fm we observe Coulomb scattering while for midperipheral collisions at 8 fm a binary collision similar to deep-inelastic scattering is observed. Besides the introduction of tthe Coulomb interaction, at low beam energies close to the Coulomb barrier it is necessary to guarantee a strict preservation of the Pauli principle. We assure this by implementing the Pauli principle separately for protons and neutrons.

The simulations were performed using various assumptions on the stiffness of the equation of state of symmetric nuclear matter, as represented by the single-particle potential in Eq. (2). The exponent κ was varied between values of 7/6 and 2, corresponding to the range of incompressibilities between 200 and 380 MeV (the value of incompressibility depends linearly on κ). Besides the stiffness of the equation of state of symmetric nuclear matter, we implemented several assumptions on the stiffness of the density dependence of symmetry energy by varying the exponent γ in Eq. (2) between 0.5 and 1.5. For each calculated reaction, the simulation was performed using 600 test particles, with 20 different sequences of the pseudorandom numbers. The simulations were performed using a computing workstation with four Xeon Phi coprocessor cards with 61 cores, allowing to perform hundreds of simulations (up to 1000) in parallel.

III. RESULTS AND DISCUSSION

In order to investigate the role of the equation of state of nuclear matter in the competition of fusion and quasifission in reactions leading to heavy and superheavy nuclei, we selected a representative set of reactions, where experimental data exists. As one of the heaviest systems, where fusion is still dominant, we use the reaction ^{48}Ca + ^{208}Pb. This reaction was measured [9,38], and a typical dominant peak at symmetric fission was observed in the mass vs TKE spectra of fission fragments, with TKE consistent to fusion-fission proceeding through the formation of the compound nucleus ^{256}Nb. Onset of quasifission was observed [39] in the reaction ^{64}Ni + ^{186}W, leading to compound system ^{250}No, where a prominent fusion-like peak is not observed anymore, however, symmetric fission, which can be attributed to fusion-fission, is still observed relatively frequently. Quasifission becomes even more dominant in the reaction ^{48}Ca + ^{238}U, nominally leading to compound nucleus ^{286}Cn. Nevertheless, the symmetric fission events still amount to about 10% of fission events [40]. Comparison with the reaction ^{64}Ni + ^{186}W shows that the relative amount of symmetric events in reaction ^{48}Ca + ^{238}U is twice lower than in the reaction ^{64}Ni + ^{186}W, thus implying the relative amount of 20% of symmetric fission for the latter reaction. In reactions ^{64}Ni + ^{208}Pb [9], ^{48}Ca + ^{249}Cf [7], and ^{64}Ni + ^{238}U [41] the quasifission already dominates and fusion hindrance amounts to several orders of magnitude (10^{-3}–10^{-5} [30,31]).

Simulations were performed at beam energy 5 MeV/nucleon, which is above the Coulomb barrier and in all cases corresponds to the nearest experimental point within few MeV. Since the angular momentum range where quasifission events are produced is not known precisely and also to assure that we will not observe deep-inelastic transfer, which occurs at peripheral collisions, we simulate the most central events, with impact parameter set to 0.5 fm (exactly central events practically do not occur in experiment). Simulations were performed up to the time 3000 fm/c, sufficient for formation of the final configuration in all investigated cases. To carry out a comparison with experiment, we need to determine from available experimental information the probability of fusion in central collisions. For the reaction ^{48}Ca + ^{208}Pb the fusion probability is close to 100%, while for reactions ^{64}Ni + ^{208}Pb, ^{48}Ca + ^{249}Cf, and ^{64}Ni + ^{238}U it is close to zero (10^{-3}–10^{-5}). Of the two remaining reactions, the total fusion probability of 10% and the fact that fusion probability peaks at central collisions infer the constraint on fusion probability in the reaction ^{48}Ca + ^{238}U at central events between 20–50% (upper limit is based on the assumption that quasifission is dominant even in central collisions). Since a comparison of shapes of experimental mass distribution in reactions ^{48}Ca + ^{238}U and ^{64}Ni + ^{186}W shows that there is approximately twice higher relative abundance of fusion in reaction ^{64}Ni + ^{186}W, we constrain the fusion probability in this reaction at central collisions between 40–80%. These constraints remain still relatively loose, but they reflect the dominant scenarios and thus the dynamics of competition of fusion and quasifission. As a consequence, this representative set of reactions allows to constrain the parameters of equation of state of nuclear matter.

From the investigated reactions, the collisions of ^{64}Ni + ^{186}W exhibit the highest sensitivity to the parameters of the equation of state. Figure 1 shows the typical evolution of the nucleonic density for the collision ^{64}Ni + ^{186}W, simulated using the soft equation of state with incompressibility $K_0 =$ 202 MeV and the soft density-dependence of symmetry energy $\gamma = 0.5$. One can see that the impinging projectile nucleus establishes contact with the target nucleus, however the weak surface tension, caused by the soft equation of state, is not sufficient to overcome Coulomb repulsion of the projectile and target which re-separate after approximately 1200 fm/c (scission time is comparable with other approaches [14,18,21,22,26]). Similar evolution was observed in all 20 simulated test particle sets. Figure 2 shows a simulation of the same reaction with $K_0 = 300$ MeV and $\gamma = 0.5$. In all simulations of this case the stronger surface tension generated by the stiff equation of state allows to overcome the Coulomb repulsion and system undergoes fusion. Strong sensitivity to the stiffness of the equation of state is thus demonstrated. Figure 3 shows the simulation with $K_0 = 202$ MeV and $\gamma = 1.5$. One can observe that increased stiffness of the density-dependence of symmetry energy can also prevent the system from separating into two fragments. In this case the weak surface tension allows to form elongated configuration (similar to Fig. 1), however, the stiffer symmetry energy prevents the formation of a low-density

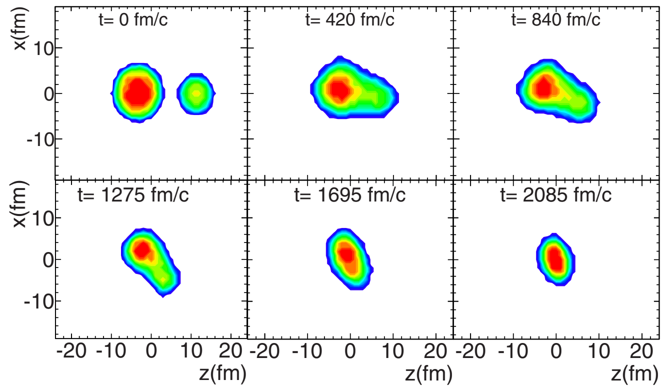

FIG. 2. Typical evolution of nucleonic density for the central collision ^{64}Ni + ^{186}W at beam energy 5 MeV/nucleon, simulated using the stiff equation of state ($K_0 = 300$ MeV) and the soft density-dependence of symmetry energy ($\gamma = 0.5$). Stronger surface tension overcomes Coulomb interaction and quasifission is prevented.

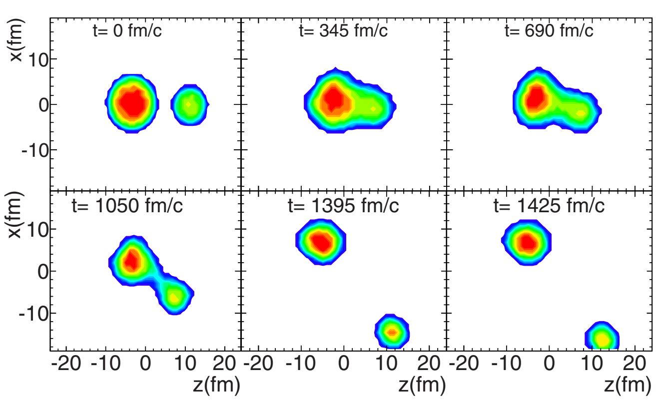

FIG. 1. Typical evolution of nucleonic density for the central collision ^{64}Ni + ^{186}W at beam energy 5 MeV/nucleon, simulated using the soft equation of state with incompressibility $K_0 = 202$ MeV and the soft density dependence symmetry energy with $\gamma = 0.5$. Weak surface tension is overcome by Coulomb interaction and quasifission occurs.

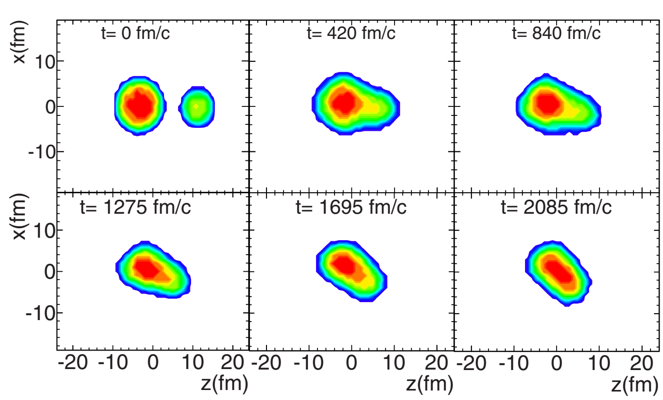

FIG. 3. Typical evolution of nucleonic density for the central collision ^{64}Ni + ^{186}W at beam energy 5 MeV/nucleon, simulated using the soft equation of state ($K_0 = 202$ MeV) and the stiff density-dependence of symmetry energy ($\gamma = 1.5$). Despite weak surface tension the stiff density-dependence of symmetry energy prevents formation of a neck and quasifission is prevented.

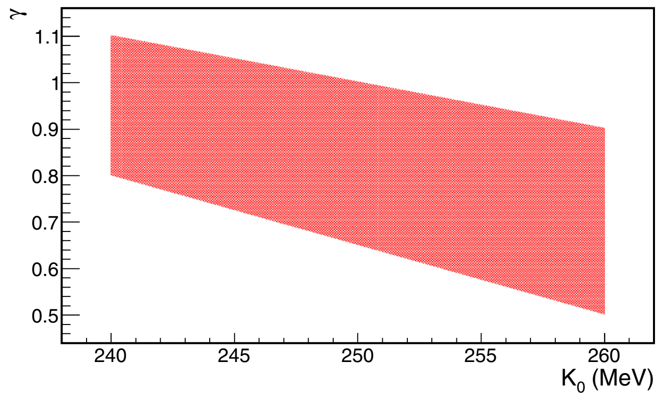

FIG. 4. Constraint on stiffness of symmetric nuclear matter (modulus of incompressibility) and on density-dependence of the symmetry energy [exponent γ from Eq. (2)] derived from the simulations of competition between fusion and quasifission.

neutron-rich neck and thus the contact between the two reaction partners is preserved until the surface tension finally overcomes the Coulomb repulsion. Figures 1–3 demonstrate a strong sensitivity of the system ^{64}Ni + ^{186}W to the parameters of the equation of state.

In the other system of comparable mass, collisions of ^{48}Ca + ^{208}Pb typically result in fusion, with exception of the simulations with $K_0 = 202$–230 MeV and $\gamma = 0.5$–1.0. For such a soft equation of state, collisions usually result in quasifission and thus such equations of state can be considered in conflict with experiment. Heavier systems ^{64}Ni + ^{208}Pb, ^{48}Ca + ^{249}Cf, and ^{64}Ni + ^{238}U usually undergo quasifission for $K_0 = 202$–255 MeV, for stiffer equations of state fusion appears and eventually becomes dominant. Thus a stiff equation of state with $K_0 = 272$–300 MeV can be rejected. Also a stiff symmetry energy with $\gamma = 1.5$ combined with soft equations of state with $K_0 = 202$–255 MeV lead to fusion and thus can be rejected. The remaining system ^{48}Ca + ^{238}U behaves similarly to ^{64}Ni + ^{186}W, consistently with constraints derived from other systems. For collision movies and other information on results of simulations see also the Supplemental Material [42].

IV. CONCLUSIONS

As a result of the analysis of competition between fusion and quasifission, it was possible to set a rather strict constraint on the incompressibility of the equation of state of nuclear matter $K_0 = 240$–260 MeV with softer density dependence of the symmetry energy with $\gamma = 0.5$–1.0 (see Fig. 4). This constraint is based on simulations of collisions, where maximum density reaches 1.4–1.5 times the saturation density. The shape of the constrained area reflects a trend of softening the density-dependence of the symmetry energy, necessary to balance the increase of incompressibility. Such trend stems from competition of the surface tension, related to the stiffness of the equation of state of symmetric nuclear matter, with the Coulomb repulsion. This corresponds to the traditional picture of nuclear fission, where fissility of the system is defined as a ratio of the Coulomb repulsion to twice the surface energy. However, the present analysis goes beyond this simple macroscopic picture and elucidates the crucial role of the density-dependence of the symmetry energy in the dynamics of the system close to the scission point. In comparison with other methods, such as constraining the equation of state using the nuclear giant resonances [43–45] or the flow observables in relativistic nucleus-nucleus collisions [46], in the present analysis the effect of the nuclear equation of state is manifested directly, and thus it is not affected by uncertainty related, e.g., to the description of the underlying nuclear structure in the former or disentangling the effect of the two-body dissipation via nucleon-nucleon collisions in the latter case. While the present work focuses on the fusion probability, the properties of the fragments in the case of quasifission were also inspected. We typically observe a close to symmetric mass split, which is natural to expect since properties of final fragments such as shell structure are not considered. The energy of Coulomb repulsion at the moment of split is comparable to experimental values of total kinetic energy (e.g., in the case of reaction ^{64}Ni + ^{208}Pb [9] and ^{48}Ca + ^{238}U [10]), which implies a reasonably realistic shape of the system at the moment of scission. Observed scission times of about 1300 fm/c are comparable to other simulations of the quasifission process, TDHF [29] or ImQMD [25]. The role of the shell structure in nuclear fission remains an open question, as demonstrated by the recent observation of the asymmetric fission of ^{180}Hg [47], contrary to expectations, based on the shell structure of fission fragments. The effect of closed nuclear shells can be manifested differently in the fusion and quasifission channel, specifically for each system, and thus no simple trend must necessarily exist. The deformation, known to be present in uranium and transuranium targets, was also not considered. We expect deformation to enhance the fusion cross section mostly at sub-barrier energies, while in the present work we consider energies above the barrier. On the other hand, recent experimental data [27] show that deformation may in fact enhance the probability of quasifission. Still, since the present work investigates reactions with both spherical and deformed targets and a relatively strict constraint is obtained, the role of deformation of the target does not appear as crucial. Thus, by using a representative set of investigated reactions we provide a solid base for the assumption that the extracted constraints do not depend critically on the effect of shell structure. In order to obtain even more strict constraints, in particular on the density-dependence of symmetry energy, more experimental data are necessary to close the onset of quasifission, where the sensitivity of the neck dynamics to the density dependence of the symmetry energy is highest.

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