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Discrimination between fission and quasi-fission from reaction time measurements

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Abstract. The formation of compound nuclei with $Z = 120$ and $Z = 124$ has been evidenced from their very long fission times measured by the blocking technique in single crystals. A possible explanation for the long measured fission times might be found in the temperature dependence of the fission barriers, as predicted by Hartree-Fock-Bogolubov calculations at finite temperature in this super-heavy nucleus domain.

1. Introduction

A compound nucleus formed by fusion of two heavy nuclei can decay either by particle emission or by fission. The latter becomes more and more probable when the mass of the compound nucleus increases. However, in the 80's, it has been shown [1, 2, 3] that the detection of fission-like fragments does not constitute an evidence for fusion: quasi-fission processes occur, characterized by the formation of composite systems that do not reach the fully equilibrated compound nucleus before splitting in two fission-like fragments. Numerous works have been devoted to quasi-fission studies. They have investigated the differences with true compound nucleus fission, usually concluding to broader mass distributions or higher most probable kinetic energies for fission-like fragments arising from quasi-fission processes. A few experiments have tried to disentangle the quasi-fission cross-section from the true fusion-fission one. However, the definition of quasi-fission in these experiments is often somewhat arbitrary, based for instance on selections justified by rough extrapolations in mass-energy plots.

The quasi-fission processes can actually arise from quite different physical origins. It can arise from vanishing fission barriers at high angular momenta, fusion being still present for the most central collisions [4]. It can also result from trajectories of the system in the potential landscapes that lead to fast separations of the partners [5]. Depending on the actual multi-dimensional potential landscape, these trajectories can be more or less favored with respect to the ones driving the system to the minimum corresponding to the compound nucleus. Finally, quasi-fission can also be due to the disappearance in the potential of the compound nucleus minimum, making thus fusion obviously impossible. Whatever the physical origin of the quasi-fission process is, the associated dynamical separation of the two fission-like fragments takes place in a very short time scale that has been indeed early measured in a quite direct way from the anisotropic associated angular distributions [3]. It was found to be less than a rotational period of the system, corresponding to typical times between 10^{-21} and 10^{-20} s.

In the present experimental work, our goal was to determine for very heavy systems, possibly leading to compound nuclei with atomic numbers larger than 110, whether the true fusion process still exists, even with rather small cross-sections as compared to quasi-fission. We have therefore searched for evidences for composite systems surviving against fission at times longer by orders of magnitude than the quasi-fission time scale, only compatible with fusion followed by fission.

2. Experimental procedure and results

For very heavy highly excited nuclei, the fission time distributions present tails arising from the competition between particle (even maybe γ -ray) emission and fission at each of the step of the nucleus decay [6, 7]. Such a tail for excited uranium nuclei can extend with sizable probabilities up to very long times, of the order of 10^{-16} s [8, 9]. The blocking technique in single crystal [10] has been applied to get direct evidence for such a tail at times longer than about 10^{-18} s for composite systems with $Z \geq 114$.

The principle of the blocking technique as applied in our experiments is to measure the fragment angular distributions around the direction of a major axis of a single crystal that is used as a target. If the composite system splits rapidly, a fission-like fragment emitted in the direction of the major axis will be deflected from its initial direction by its atomic interactions with all the atoms of the crystal row. Therefore, for short splitting times of the composite systems, the measured angular distribution presents a dip with a minimum precisely in the

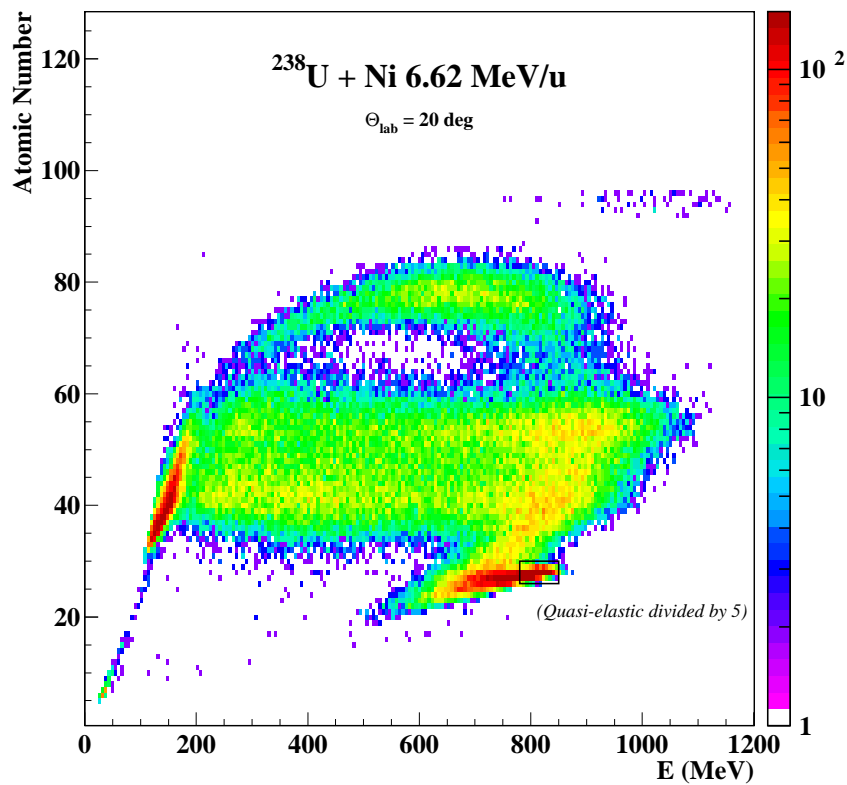


Figure 1. Atomic number versus total kinetic energy.

direction of the crystal axis. When the composite system lifetime becomes longer, the potential felt by the emitted fragments is lower resulting in a less marked dip. However, for the shortest fission times, the splitting can take place within the thermal vibrations of the crystal atoms, leading thus to a probability to detect a fragment precisely in the direction of the crystal axis that does not depend anymore on the splitting time. It has been shown that to first order, the width of the blocking dips evolves as the square root of Z/E [10], but the blocking technique has a lower time sensitivity time limit that has a pure geometrical origin, the amplitude of the thermal vibrations. Any filling of the dip in the direction of the crystal axis is a model independent signature that the composite system had a fission time longer than the time it needs to recoil out of the thermal amplitude area.

Three systems have been studied [11] at Ganil for which the possibly formed compound nuclei have excitation energies between 70 and 80 MeV. The first system was $^{208}\text{Pb} + \text{Ge}$ at 6.2 MeV/A that would lead to a compound nucleus with $Z = 114$. The second system was $^{238}\text{U} + \text{Ni}$ at 6.6 MeV/A, leading to compound nuclei with $Z = 120$, and the last one was $^{238}\text{U} + \text{Ge}$ at 6.1 MeV/A, leading to compound nuclei with $Z = 124$.

For the three systems studied, the lower sensitivity limit given by the thermal vibrations of the atoms is $\sim 10^{-18}\text{s}$, at least two orders of magnitude longer than the time scale associated to quasi-fission. The blocking effects were measured at 20° with respect to the beam axis. All coincident fragments and light charged particles were detected on a solid angle close to 4π sr by

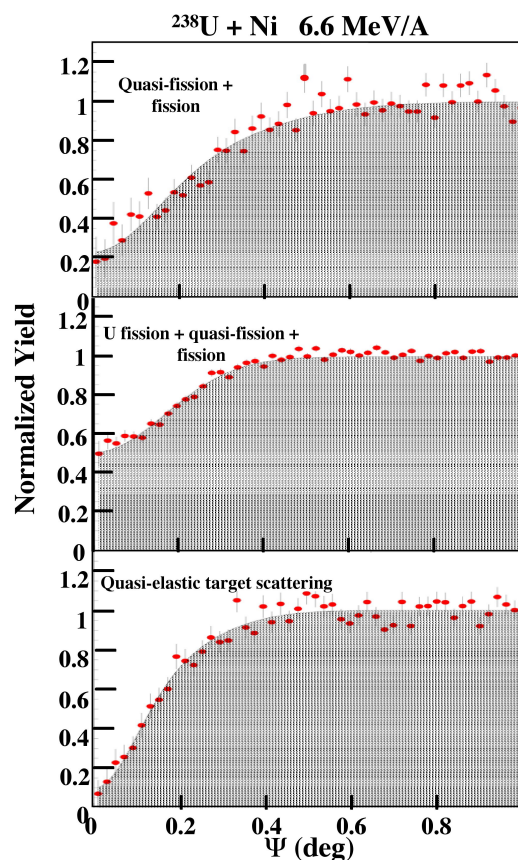


Figure 2. Blocking dips measured for three different selections of atomic numbers corresponding to different reaction mechanisms (see text).

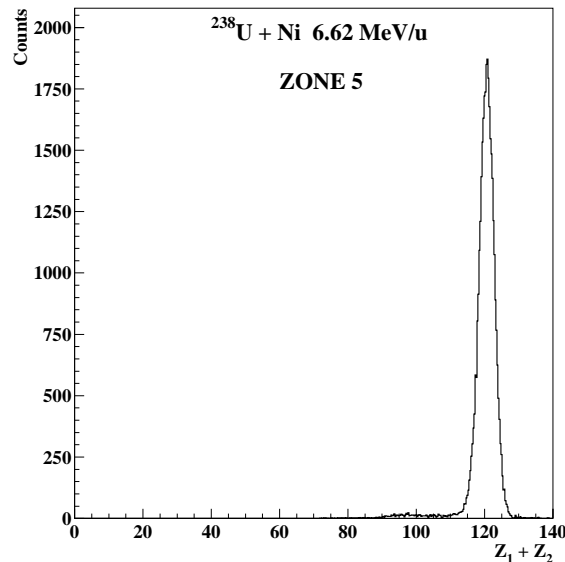


Figure 3. Sum of the atomic numbers of fission-like fragments when one of the fragments is detected at 20° with $65 < Z_1 < 85$.

the multidetector INDRA [12]. The correlation between the atomic number Z and the energy E of the fragments detected at 20° had rather similar behaviors [11], presented for example in Fig. 1 for the $^{238}\text{U} + \text{Ni}$ system. For Z close to the atomic number of the target, a quasi-elastic pic is observed, with an inelastic tail towards lower Z and E values. For $Z \approx 80$, fission-like fragments are measured with very high yields. These fission-like events arise either from quasi-fission or from true fusion-fission reactions. The fragments with Z in between the two previous regions arise either from quasi-fission processes, or from fusion-fission or from a sequential fission of the projectile-like fragment (U or Pb). Finally, for atomic number close to the projectile, a more or less pronounced deep-inelastic branch of the projectile-like fragment is observed, depending on the system considered.

The blocking dips associated to three different mechanisms selected in the Z versus E maps are presented as an example in Fig. 2 for the $^{238}\text{U} + \text{Ni}$ system. For the selection presented in the lower bin of the figure, the dip associated with quasi-elastic target scattering presents a minimum value at $\psi = 0^\circ$ (in the direction of the crystal axis) significantly lower than the one associated with the two other selections ($38 < Z < 58$ for the middle bin and $70 < Z < 85$ for the upper bin). As previously stressed, such an increase of the yield measured at $\psi = 0^\circ$ indicates composite system lifetimes longer than 10^{-18}s . It has to be noted that the blocking dips might be smeared by specific experimental conditions, for instance the size of the beam spot or defects in the crystalline structure of the targets with respect to perfect crystals. Such smearing effects might result in an artificial increase of the measured yields in the $\psi = 0^\circ$ direction, but such an increase would be the strongest for the narrowest dips, which are in our analyses the ones associated to the quasi-elastic target recoils. For the selection presented in the middle bin, the presence of sequential fission of uranium nuclei is responsible for the strong observed yield increase at $\psi = 0^\circ$. However, no sequential fission events are present in the selection done in the upper bin. For this selection, only two heavy fragments are detected in the whole space. The sum $Z_1 + Z_2$ of their atomic numbers is shown in Fig. 3 for the $^{238}\text{U} + \text{Ni}$ system, exhibiting a

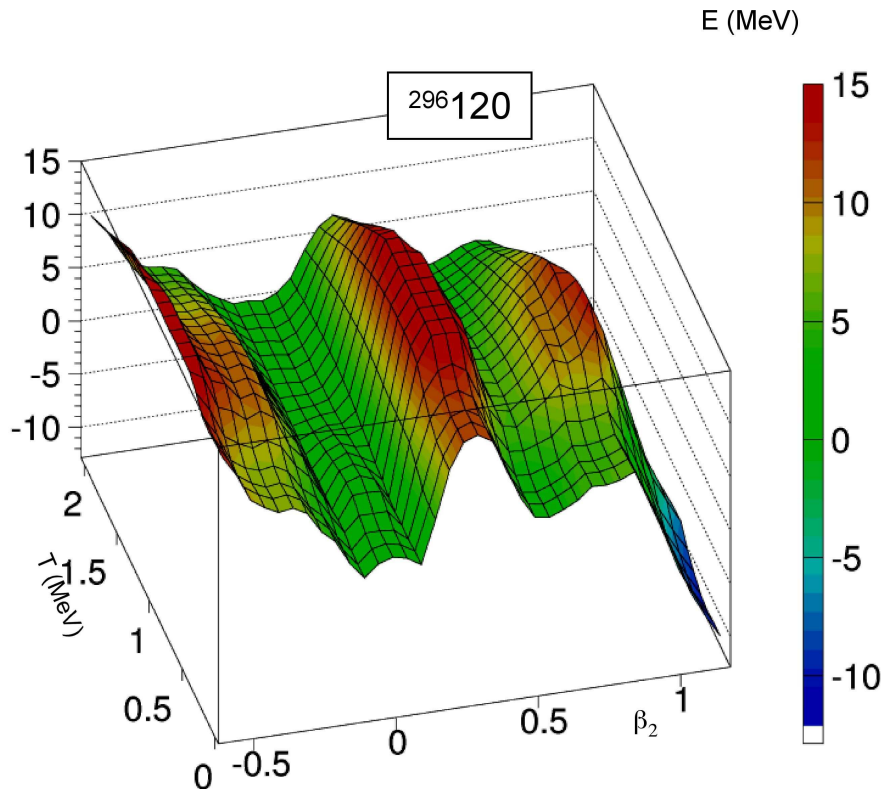


Figure 4. Hartree-Fock-Bogoliubov potential as a function of the temperature and of the deformation parameter β_2 .

narrow distribution, peaked at $Z_1 + Z_2 = 120$ with a width corresponding to the experimental Z resolution. In addition, the associated light charged particle and cluster multiplicities measured on 4π sr are very low, a few percents, providing thus an extra evidence that these fragments arise from composite systems formed by all the nucleons from the target and the projectiles (at least by all the protons; no information on possible neutron preequilibrium emission can be obtained in the present experiments). The sizable increase of the yield measured at $\psi = 0^\circ$ in Fig.2 is thus a quite direct evidence for the formation of nuclei with $Z = 120$ that survive against fission more than 10^{-18} s. Similar conclusions are obtained for the formation of nuclei with $Z = 124$ in the $^{238}\text{U} + \text{Ge}$ system, but not for the formation of $Z = 114$ in the $^{208}\text{Pb} + \text{Ge}$ system [6, 11].

3. Dependence of fission barriers on temperature: effect on fission times

From the data presented in the previous Section, three main conclusions can be drawn: i) for the $^{238}\text{U} + \text{Ni}$ and $^{238}\text{U} + \text{Ge}$ systems at 6.6 and 6.1 MeV/A, respectively, fusion occurs with sizeable cross-sections; ii) the fission barriers of all the transient nuclei formed during the decay of the $Z = 120$ and 124 nuclei formed at high excitation energies are high enough to permit these nuclei to survive against fission more than 10^{-18} s; iii) the observed fission is essentially asymmetric, with one of the fragments with $Z \approx 80$, suggesting thus a fission process at rather low residual excitation energies, compatible with the long fission times evidenced. The two last conclusions seem hardly compatible with the temperature evolution of the fission barriers usually considered in statistical models applied to super-heavy fragment decay: considering that the fission barriers of super-heavy nuclei are essentially due to shell effects, an exponential

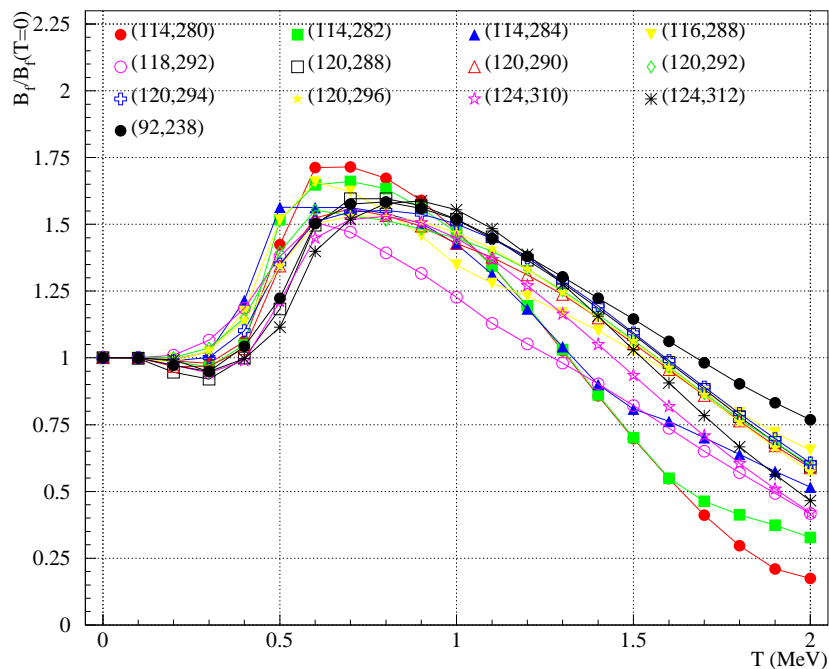


Figure 5. Evolution of the maxima of the fission barriers with temperature.

decrease of the fission barriers is usually assumed in order to take into account the shell effect damping with temperature [13]. However, the shell damping coefficient must often be considered as a free parameter in order to reproduce the data.

In order to have a better insight into the evolution of the super-heavy nucleus fission barriers with temperature, Hartree-Fock-Bogoliubov (HFB) calculations at finite temperature have been recently undertaken [14, 15], using the effective interaction D1S from D. Gogny [16, 17]. The HFB potentials have been calculated for a series of nuclei (super-heavy nuclei and ^{238}U) as a function of their temperature and deformation. A typical two-dimensional plot of these energies is presented in Fig. 4 for the nucleus ($Z=120$, $A=296$). From such plots, the fission barriers can be inferred at a given temperature from the difference between the maximum potential obtained at a given deformation β_2 and the HFB ground state energies considered at $T = 0$ MeV. Surprisingly, the fission barrier maximum is not found at $T = 0$ MeV, but at a much higher temperature that can be better determined in Fig. 5 where the fission barrier maxima at temperature T have been normalized with respect to the one at $T = 0$ MeV for all the studied nuclei: between $T \approx 0.6$ and $T \approx 0.8$ MeV the maxima of the fission barriers can be larger than the one at $T = 0$ MeV by more than 50%.

The maximum observed in Fig 5 arises from the evolution of the pairing energies with deformation and temperature. Indeed, to determine the height of a fission barrier, the pairing effects have to be taken into account both at the deformation corresponding to the ground state and at the deformation corresponding to the top of the barrier. Fig. 6 presents for ($Z = 120$, $A=296$), as a function of the deformation, the total HFB energy at $T = 0$ MeV in the top panel and, in the lower panels, the pairing energies at various temperatures. For this nucleus, the ground state corresponds to two minima, symmetric with respect to $\beta_2 = 0$, at $\beta_2 \approx -0.1$ and $\beta_2 \approx +0.1$, whereas the maximum of the fission barrier is at $\beta_2 \approx 0.3$. Fig. 6 shows that the contribution of the pairing energies to the potential energy at $T = 0$ is much larger for

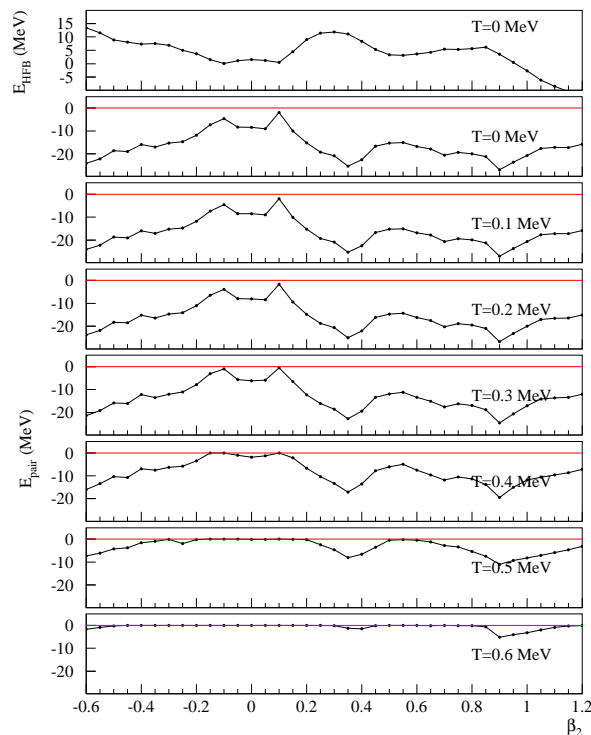


Figure 6. Hatree-Fock-Bogoliubov calculations for $Z = 120$, $A = 296$. Upper panel: Hartree-Fock-Bogoliubov energy at $T = 0$ MeV; lower panels: pairing energies at various temperatures.

deformations corresponding to the maximum of the barrier ($\beta_2 \approx 0.3$) than for deformations corresponding to the ground state configurations ($\beta_2 = \pm 0.1$). For increasing temperatures, the pairing energy at $\beta_2 \approx 0.3$ decreases in absolute value, but remains of sizable importance up to rather high temperatures, whereas the pairing energy for the ground state configurations becomes rapidly negligible. Such strong evolutions of the differences of the pairing energies at the top of the fission barriers and close to the ground states are responsible for the increases of the fission barriers between 0 and about 800 keV. Let us mention that the Gogny effective interaction is known to provide an accurate description of nuclear pairing and is usually considered as a reference in this domain in the theoretical nuclear community.

This unexpected evolution of the fission barriers with temperature should imply quite dramatic changes in the statistical competition between particle (essentially neutrons) emission and fission for super-heavy nuclei. In the following, we describe a schematic scenario that can take into account long fission times together with tiny survival probabilities. It considers at each step of the decay the non-monotonic evolution of the fission barriers with temperature from Fig.5, coupled with the increasing neutron deficiency of the surviving nuclei. For instance, the nucleus with $Z = 120$ formed in $^{238}\text{U} + \text{Ni}$ reactions at $T \approx 1.6$ MeV (see previous chapter) presents a fission barrier height similar to the one at $T = 0$ MeV. Due to nuclear matter viscosity, particle emission is strongly favored with respect to fission during a transient time [18] and the initial compound nucleus will cool down very fast towards a region of higher fission barriers by evaporation of a few neutrons. Once the statistical equilibrium for the fission width is reached [19, 20], a regular competition between evaporation and fission takes place, but involving nuclei with higher fission barriers and neutron bindings, implying thus longer lifetimes. The

initially formed nucleus has thus, thanks to neutron emission, a sizable probability to survive against fission and to reach the region between 0.6 and 0.9 MeV corresponding to very high fission barriers. There, it will have very long lifetimes and a high probability to decay through neutron emission, leading thus to overall long fission times. However, after a few more neutron emissions these nuclei enter a domain of temperature ($T \lesssim 0.4$ MeV) where the fission barriers becomes lower due both to the temperature decrease and to higher and higher neutron deficiency. Considering the lower fission barriers and the higher involved neutron bindings, the nucleus will finally undergo fission with a very high probability, but at a very long overall fission time.

4. Conclusion

Direct evidence for the formation by fusion of nuclei with $Z = 120$ and 124 has been reached thanks to their very long measured fission times. A quite unexpected evolution of the fission barriers with the temperature is obtained by Hartree-Fock-Bogoliubov calculations at finite temperature. Considering such an evolution, a schematic scenario for the decay of these nuclei can be suggested that takes into account qualitatively the apparent discrepancy between the long measured fission times and the extremely difficult synthesis of super-heavy elements. It stresses the need for realistic parameters when statistical models are used in the super-heavy nucleus domain and the possible misleading conclusions that might be drawn from rough parameter extrapolation. In order to reach quantitative reproductions of the super-heavy nucleus decay through statistical models, more efforts are now clearly needed to fix the model parameters.

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