Proc. 13th Int. Conf. on Nucleus-Nucleus Collisions
JPS Conf. Proc. **32**, 010002 (2020)
https://doi.org/10.7566/JPSCP.32.010002

Experimental measures of fission time scales

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(Received July 19, 2019)

The nuclear fission process involves a drastic rearrangement of the nuclear matter of the fissioning system as it undergoes dramatic shape changes from a single, compact nucleus to two separated fission fragments. Much research, both theoretical and experimental, has focused on understanding the dynamics of this process using a number of different approaches. Soon after the discovery of fission in 1938-39, Bohr and Wheeler proposed the transition state model in which the fission decay width / lifetime was obtained by the statistical counting of transition states at the fission barrier. Subsequently, Kramers suggested that the friction or viscosity of the nuclear matter can play a substantial role in slowing down the decay rate by large factors. Since then, many theoretical works have addressed this issue, most intensely during recent decades, but consensus on the dynamical description of fission has not yet been reached.

In light of this situation, it is important to consider whether experiments can shed some light on the issue. Several different methods have been used to obtain experimental information on the dynamics of fission and the associated time scales for the process. In general, the approach is to compare the fission time to some other process that is believed to be better understood. Although the divergence of results obtained by the different methods has been known for years, these discrepancies have not yet been resolved and they have recently received renewed attention.

In this talk, I will review some of the experimental measurements of fission time scales and discuss the discrepancies between the different methods.

KEYWORDS: Nuclear fission, nuclear dynamics, timescales

1. Introduction

Despite the fact that it is energetically favorable for heavy nuclei to divide into two roughly equal-size fragments via the fission process, this decay mode is strongly suppressed by the potential barrier (the fission barrier) that separates the initial and final states. Only when the nucleus is excited to an energy comparable to the height of the fission barrier can this process compete with other decay channels. Slow neutron capture is one method for imparting excitation energy to the nucleus, and this was indeed the route used by Hahn and Strassmann [1], building on earlier work by Hahn, Meitner and Strassmann *e.g.* [2], that led to the clear chemical identification of Ba nuclei among the final products. Soon afterwards, Meitner and Frisch [3] gave the theoretical explanation of this observation, in terms of nuclear fission, and this interpretation was verified by Frisch [4] via physical measurements.

Despite the fact that nuclear fission is a complicated, dynamic process in which a heavy, atomic nucleus transforms into two large nuclear fragments, many aspects of the reaction were initially well described by a simple transition-state model. This approach was initially introduced in the trail-blazing paper by Bohr and Wheeler [5], in which the fission decay width of a compound system is computed by enumerating the number of open channels at the bottle-neck for the process, namely the 'transition state' or saddle point. This approach is analogous to the counting of the final states in the daughter nucleus for simpler decay channels such as neutron or γ-ray emission. The influence of the dynamics of the fission process was soon afterwards pointed out by Kramers [6], who considered the process in analogy to the Brownian motion and derived, under certain assumptions, an expression for the fission decay width, which incorporated the effects of dissipation.

Although the effects of nuclear dissipation on the fission decay width and time scale was considered in theoretical works [7], this topic was not studied experimentally for several decades until the measurements of the neutron multiplicity from highly excited nuclei formed in heavy-ion reactions [8,9] showed a substantial deficit compared to statistical model expectations. Subsequently, several studies and experimental techniques have been developed and applied in order to study the dynamics of the fission process. These techniques rely typically on a comparison of the fission time-scale relative to another time-scale, or "clock", in the process that is believed to be better understood. Each method is sensitive to fission time scales over a limited interval as shown in Fig. 1.

Sikdar *et al.* [10] have recently pointed out that there is a substantial discrepancy between the fission time scales measured with different "clocks" for the Z=120 system formed in the ^{64}Ni + ^{238}U reaction. The discrepancy is most pronounced between fission times obtained with so-called "atomic clocks", notably the crystal blocking technique and compound-nucleus X-ray emission, in relation to those obtained by "nuclear clocks" such as the rotation rate of the di-nuclear system, and the emission rate of evaporation neutrons and Giant Dipole Resonance (GDR) γ-rays.

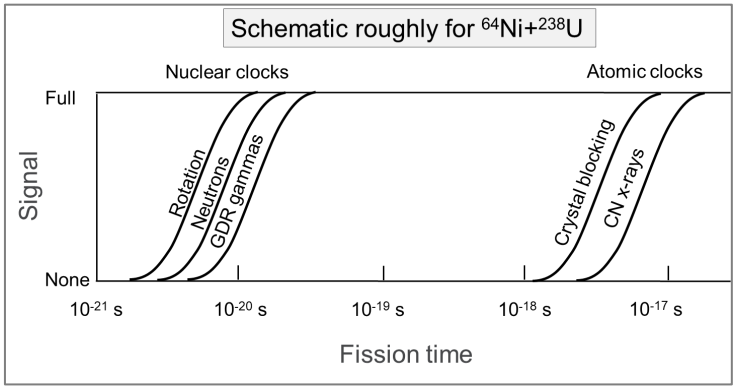

Fig. 1: Relation between the fission time and the experimental signal for five different methods.

In this paper, I will discuss the results from data for the ^{64}Ni+^{238}U system, which, fortunately, has been measured using these different methods at similar bombarding

energies. In this system, it is expected that the observed fission products arise chiefly from the quasi-fission process and that the observed reaction time reflects the time from the formation of a rotating, di-nuclear system, which undergoes a dynamic process mass transfer and elongation until the final fragments separate at the scission point

2. Di-nuclear rotation

The di-nuclear rotation "clock" relies simply on observing the angle of rotation of the system while the two nuclei are in contact during a quasi-fission reaction as illustrated in Fig. 2.

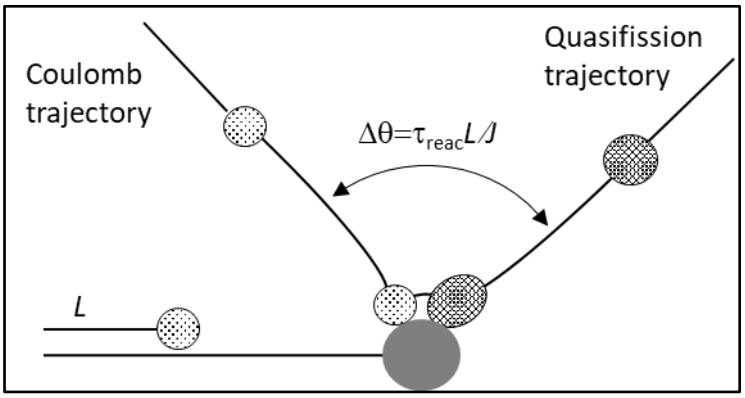

Fig. 2: Illustration of the rotation of the di-nuclear system during contact.

Tōke *et al.* [11] studied the ^{238}U+^{64}Ni system at a beam energy of 6.0 MeV/u and correlated the observed scattering angle of the quasi-fission products with average L-value by dividing the measured cross section into three bins. After correcting for the deflection angle assuming Coulomb trajectories, the rotation angle, Δθ, was determined. Applying simple expressions for the relevant moments of inertia, J, it was thus possible to derive the reaction time, τ_{reac}, using the expressions

$$\omega = {}^{L}/_{J}\,,\ \Delta\theta = \tau_{reac}\,{}^{L}/_{J}\,, \text{and } \tau_{reac} = \Delta\theta\ {}^{J}/_{L.} \quad (1)$$

The analysis yields fission times in the range of (2.5-7.5)×10^{-21}s for this system. A recent analysis of quasi-fission of 6.01 MeV/u ^{40}Ca+^{238}U give similar time scales [12] in agreement with time-dependent Hartree-Fock calculations presented in the same publication.

3. Pre-scission neutron emission

This method is somewhat complicated because one needs to be able to separate different sources of neutrons, namely those emitted from the composite system prior to scission and those emitted by the fragments post scission. The measurements take advantage of the different kinematics for these two sources by measuring neutron spectra

at different angles relative to the measured direction of the fission fragments. Hinde and collaborators [13-15] have carried out a series of measurements using this method including the ^{64}Ni+^{238}U reaction at 6.5 MeV/u [15] considered here. The analysis of the data results in fission times of τ_{reac} = (9-19) ×10^{-21} s for fragments in the mass range 120<A<200 u, which is in good agreement with the results from di-nuclear rotation.

4. Giant Dipole Resonance γ-ray emission

In this approach, the high-energy γ-ray spectra measured in coincidence with (quasi)-fission fragments are analyzed and compared to predictions of statistical model calculations to determine the fission time-scale in relation to the well-known rate for γ-ray emission. The method has been used to study a range of heavy-ion induced reactions [16,17], but unfortunately, there are no data available for the ^{64}Ni+^{238}U system being considered here. However, we may consider the results obtained by Nestler *et al.* [18], who studied the ^{58}Ni+^{165}Ho reaction at a beam energy of 6.35 MeV/u. These authors found no evidence for the GDR γ-ray component in the spectrum, which gave an upper limit for the fission time scale of $\tau_{reac} < 11\times10^{-21}$ s in good agreement with those obtained for the ^{64}Ni+^{238}U system using the di-nuclear rotation and neutron emission "clocks".

5. Crystal Blocking

The crystal blocking "clock" [19] relies on the fact that the prompt emission of fission fragments from lattice sites are substantially suppressed, whereas a delay in the fission process allows the system to travel to locations in between these where they can freely emerge along "channels" in the crystal. Morjean *et al.* [19] have recently studied the reaction of ^{238}U ions impinging onto a Ni crystal at 6.62 MeV/u and observing the yield in the direction of the $\langle\mathbf{110}\rangle$ crystal axis. The dip in the yield along this axis is found to correspond to fission time of $\sim\tau_{reac} = 1\times10^{-18}$ s, which is substantially longer than those found with the "nuclear" techniques discussed above. The sensitivity of this method is extended into the 10^{-18} s range by using an inverse kinematics reaction that provides a high recoil velocity of the fissioning system.

6. K X-ray emission

This method takes advantage of the generation of inner shell vacancies during the heavy ion reaction. The subsequent filling and the associated emission of characteristic K X-rays occurs on a short time scale that offers another method of assessing the life-time of the combined system. Standard methods are used to calculate the K X-ray spectrum and the life time of the K-shell vacancies. Frégeau *et al.* have used this approach to study of the ^{238}U+^{64}Ni reaction at 6.6 MeV/u [20]. The authors observe a peak in the X-ray spectrum in coincidence with quasi-fission fragments in the range 50 < Z < 91 that they interpret as K X-ray emission from the combined system during its lifetime. The analysis shows that the composite Z=120 system has a lifetime of $\sim\tau_{120} = 1\times10^{-18}$ s. This time scale is in agreement with the result from crystal channeling, but substantially longer than those measured with "nuclear clocks".

7. Comparison and Discussion

The time scales of the composite system formed in the ^{238}U+Ni collision system are listed in Table I. In one case, the bombarding energy is about 10% lower than for the other three experiments and in another, a non-separated Ni target material was used. However, these differences in experimental conditions are unlikely to affect the main conclusions from this comparison.

There is reasonable, but not perfect, agreement between the methods using the di-nuclear rotation and the emission of pre-scission neutrons to estimate the lifetimes of the fissioning system. The remaining discrepancy of a factor of 2-3 may very well stem from the different methods used in the analysis of the data or the fact that the di-nuclear rotation experiment was performed at a 10% lower beam energy. This discrepancy may possibly be reduced in a consistent re-analysis of the two data sets, but it is not considered essential in this context.

Table. I: Comparison of the characteristics and quasi-fission times obtained by four different methods

System	E_{beam} (MeV/u)	Method	Fragment range	Lifetime (10^{-21} s)	Reference
^{238}U+^{64}Ni	6.0	Di-nuclear rotation	180<A<222	2.5-7.5	Tōke *et al.* [11]
^{64}Ni+^{238}U	6.5	Neutron emission	120<A<220	9-19	Hinde *et al.* [15]
^{238}U+^{nat}Ni	6.62	Crystal blocking	67<Z<85	~1000	Morjean *et al.* [19]
^{238}U+^{64}Ni	6.6	K X-ray emission	35<Z<90	>2500	Frégeau *et al.* [20]

Likewise, one finds that the results from the experiments using “atomic clocks”, namely crystal blocking and K X-ray emission, are in reasonable agreement with a time scale of the order $\tau_{reac} = (1\text{-}2.5)\times10^{-18}$ s, almost three orders of magnitude longer than what was found by the first two methods. Frégeau *et al.* [20] have considered the possibility of a bi-furcation of the reaction strength into a short-lived and a long-lived component as an explanation for the disparate results. This possibility is, however, considered implausible by Sikdar *et al.* [10], in part because it would require a high fission barrier, relative to the neutron binding energy, in the last step of the decay cascade where small decay widths, and long life times, may be present. If a substantial fraction of the capture cross section would reach this last stage of the decay chain, it seems inconceivable that the all of this flux would lead to fission and not even a tiny fraction would end up in the Z=120 ground state. In an attempt to synthesize element Z=120 using the ^{64}Ni+^{238}U reaction, Hofmann *et al.* [21] found an upper limit for the cross section of $\sigma_{Z=120}$ <90 fb. Assuming that the mid-target bombarding energy was about 6 MeV/u, which corresponds to a capture cross section of ~130 mb [11], this result indicates less than ~10^{-12} probability of populating the Z=120, A=299 nucleus in a 3n evaporation cascade. Therefore, it does not seem plausible that a substantial fraction of the capture cross section, ~10% and 57%, as speculated in Refs. [19, 20], respectively, could have reached this last stage of the evaporation chain without populating the ground state at a cross section above the experimental upper limit.

8. Conclusions

In this paper, I have discussed the discrepancy that exists between published results on the fission time scale for the ^{64}Ni+^{238}U system obtained by four different experimental methods. This discussion builds on work presented in Refs. [10,11,15,18-20]. It does not resolve the issue, but points to the unlikelihood that a large fraction of the capture cross section leads to a slow branch via compound nucleus formation as discussed in Refs. [19,20]. On the other hand, a slow, dynamical path through the potential energy landscape, see e.g. Ref. [22], to scission may not be excluded as a source for the slow fission branch.

One may be tempted to call into question the validity of some of the results presented here based on the fact that they are at variance with those of previous measurements and various accepted theoretical models of the fission dynamics. However, a more profitable path may be to critically re-analyze all of the data, while also repeating the experiments. For example, using modern techniques, it seems possible to perform both the di-nuclear and neutron-emission measurements in a single experiment. Additional effort should be devoted to resolve the discrepancy as it severely challenges our present understanding of the reaction mechanism in heavy systems.

Acknowledgment

This work was supported by the U.S. Department of Energy, Office of Nuclear Physics, under contract No. DE-AC02-06CH11357.

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