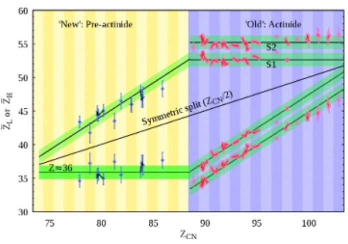
भारी-आयन प्रेरित नाभिकीय विखंडन

9

एक नए प्रकार का विखंडनः भापअकें-टीआईएफआर पेलेट्रॉन लीनॉक सुविधा द्वारा योगदान

के. माहात[1,2,*], वी. कुमार[1,2], के. रामचंद्रन[1], ए. श्रीवास्तव[1,2], एस.के. पंडित[1,2], वी.वी. पार्कर[1,2]
[1]नाभिकीय भौतिकी प्रभाग, भाभा परमाणु अनुसंधान केंद्र (भापअ केंद्र), ट्रांबे, मुंबई – 400085, भारत
[2]होमी भाभा राष्ट्रीय संस्थान, अणुशक्तिनगर, मुंबई – 400094, भारत



असममित विखंडन में हल्के एवं भारी खंडों की माध्य प्रोटॉन संख्या प्री-एक्टिनाइड्स और एक्टिनाइड्स के बीच एक विस्मयकारी संबंध दर्शाती है।

सारांश

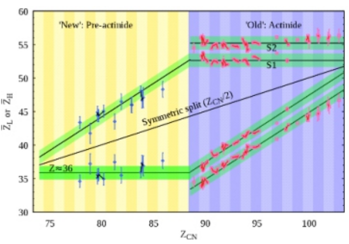
परमाणु विखंडन की व्यापक समझ, जिसमें नाभिकीय ऊर्जा से लेकर रेडियोआइसोटोप के उत्पादन तक के विस्मयकारी अनुप्रयोग हैं और जो आधुनिक भौतिकी की कई अवधारणाओं के लिए एक परीक्षण वस्तु है, इसे अब तक समझा नहीं जा सका है। क्वांटम बहु-निकाय प्रणाली की बड़े पैमाने पर सामूहिक गति इतनी जटिल है कि आठ दशकों से अधिक समय बीत जाने के बाद भी इस क्षेत्र में प्रयोगात्मक अवलोकनों द्वारा विकास संचालित होता रहा है। ^{180}Hg के बीटा-विलंबित विखंडन में असममित द्रव्यमान वितरण के अप्रत्याशित अवलोकन ने प्री-ऐक्टिनाइड नाभिक के विखंडन में बड़े पैमाने पर सैद्धांतिक एवं प्रयोगात्मक कार्यों हेतु आकर्षित किया है। यह आलेख, भापअ केंद्र-टीआईएफआर पेलेट्रॉन लीनॉक सुविधा का उपयोग करके असममित विखंडन अन्वेषण का वर्णन किया गया है।

Heavy-ion Induced Nuclear Fission

9

A New type of Fission: Contributions from the BARC-TIFR Pelletron LINAC Facility

K. Mahata[1,2,*], V. Kumar[1,2], K. Ramachandran[1], A. Shrivastava[1,2], S. K. Pandit[1,2] and V. V.Parkar[1,2]
[1]Nuclear Physics Division, Bhabha Atomic Research Centre (BARC), Trombay, Mumbai-400085, INDIA
[2]Homi Bhabha National Institute, Anushakti Nagar, Mumbai-400094, INDIA



Mean proton number of the light and heavy fragments in asymmetric fission render a striking connection between pre-actinides and actinides.

ABSTRACT

Comprehensive understanding of nuclear fission, which has tremendous applications ranging from nuclear energy to production of radioisotopes and is a testing ground for many concepts of the modern physics, remains elusive. This large-scale collective motion of the quantum many-body system is so complex that even after more than eight decades the developments in this field are steered by the experimental observation. Unexpected observation of asymmetric mass distribution in beta-delayed fission of ^{180}Hg garnered tremendous theoretical and experimental interest in fission of pre-actinde nuclei. This article describes the investigations of asymmetric fission using the BARC-TIFR Pelletron LINAC facility.

KEYWORDS: *Asymmetric fission, Shell corrections, Heavy-ion induced fusion-fission, Entrance channel dependence*

*Author for Correspondence: K. Mahata
E-mail: kmahata@barc.gov.in

Introduction

Nuclear fission is an example of large-scale collective motion of nuclei, where a single nearly spherical nuclei undergoes deformation and finally splits into two or more fragments. Drastic rearrangements of nucleons take place during this process. The macroscopic Liquid Drop Model (LDM), which explains the gross properties of fission, predicts symmetric fission to be most probable. However, the low energy fission of actinides was observed to be dominantly asymmetric. This was attributed to the preferential fragment formation near magic nuclei that have large shell (microscopic) corrections. The incorporation of oscillating quantum shell corrections, which moulded many hills and valleys in the smooth LDM potential energy surface, also explained the observations of fission isomers and near constancy of fission barrier of the actinides. This also led to the predictions of existence of island of stability beyond actinides, which gave a huge impetus for heavy-ion induced nuclear reaction studies. The elements in this island are called super heavy elements (SHE) and are completely stabilized by the shell corrections as there is no liquid drop barrier for them.

Attempts to form SHE in fusion of two heavy nuclei by overcoming their Coulomb repulsion opened a new dimension to the nuclear physics research. The composite system, produced when two heavy-ion come in contact, possess high excitation energy and angular momentum. Evolution of such systems needed detailed investigations. As shell corrections weakens with the increase in excitation energy, it is paramount to consider the gradual damping of shell correction in order to have reliable prediction of the fission survivability of such hot and rotating compound nucleus. At high excitation energies, the effect of dissipation is also observed. The presence of dissipation slows down the dynamical evolution, enhancing the neutron emission probability and fission survivability. It was also realized that when the charge product (Z_1Z_2) of the colliding partners is larger than 1000, the composite system may reseparate before forming fully equilibrated compound nucleus. This phenomenon is called quasi-fission and has drawn tremendous attention as it severely impedes formation of the SHE. Apart from the charge product, deformation, difference in N/Z ratio and magicity of the colliding partners are found to influence the dynamical evolution in heavy-ion induced reactions.

Reliable predictions of fission properties are very crucial not only for harnessing nuclear energy and production of radioisotopes in a more safe and efficient way, but also for understanding synthesis of heavy elements and neutrino physics research. Even though, lots of progress have taken place, a comprehensive understanding of the heavy-ion induced fission process has not been achieved so far and experimental surprises still steer the progress in this field. Particularly, fission in the pre-actinde region remained less explored as the fission probability is low. Fission studies in this region, having large ground state shell corrections and a balanced fission-neutron evaporation competition, might help to understand the two key factors, namely the damping of shell correction and dissipative dynamics of nuclear medium.

Asymmetric fission in pre-actinides

Mass asymmetric split, which dominates in low-energy fission of actinides, was found to recede while moving towards lighter nuclei. Though the experimental mass distributions around A = 200 displayed slight dip around symmetric split, mass distributions for A<200 were characterized by single peak as expected form the LDM behaviour. A recent experimental observation at ISOLDE, CERN in beta-delayed fission of ^{180}Hg [1] has generated lots of excitements in this area as it has challenged the understanding of fission developed through the study in the actinide region. In case of ^{180}Hg, the liquid drop model favoured symmetric fragment, ^{90}Zr, has magic numbers of neutrons (N = 50) and semi-magic numbers (Z = 40) protons. Thus, from liquid drop model as well as fragment magicity point of view, symmetric fission was expected to be preferred. However, the experimental mass distribution of ^{180}Hg in beta-delayed fission was found to be dominantly asymmetric. This observation garnered tremendous theoretical and experimental interest in this field. Though most of the theoretical model studies were limited to a few cases, the predictions of Brownian Shape Motion (BSM) model are available for a large number of nuclei [2].

Beta-delayed fission studies are limited to a few nuclei and have limited statistics. Experimentally, heavy-ion fusion-fission route was also explored to study the same phenomena systematically. Several studies were carried out using the BARC-TIFR Pelletron LINAC facility. Due to the Coulomb barrier, it is not possible to keep the excitation energy of the fissioning system very low. In case of beta-delayed fission, the excitation energy is just above the fission barrier. While the light particle (p, α) induced fission studied have been carried out down to excitation energy 10 MeV above the barrier, heavy-ion induced studies are limited to even higher excitation energies. At higher excitation energies shell effects diminish considerably, reducing the preference for the shell driven fission mode(s). However, it allows the study of damping of shell correction with excitation energy, which has implication in fission survivability of heavy and super-heavy compound nuclei. Observation of asymmetric fission in ^{35}Cl+^{144,154}Sm [3] was one of the first few studies, which showed the sensitivity of heavy-ion fusion-fission to the newly observed asymmetric fission mode in the

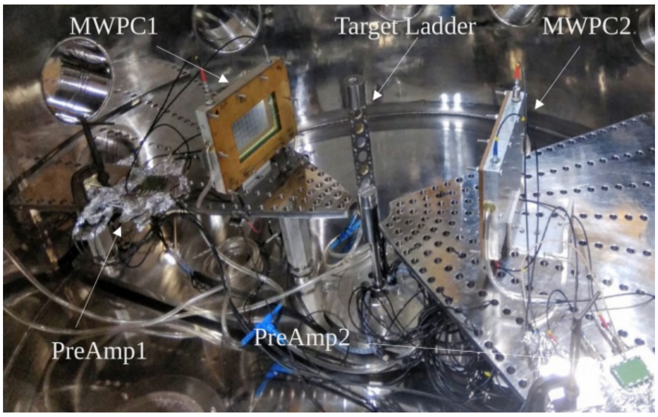


Fig.1: A photograph of the experimental setup used for fission fragment mass distribution measurement at the BARC-TIFR Pelletron LINAC facility.

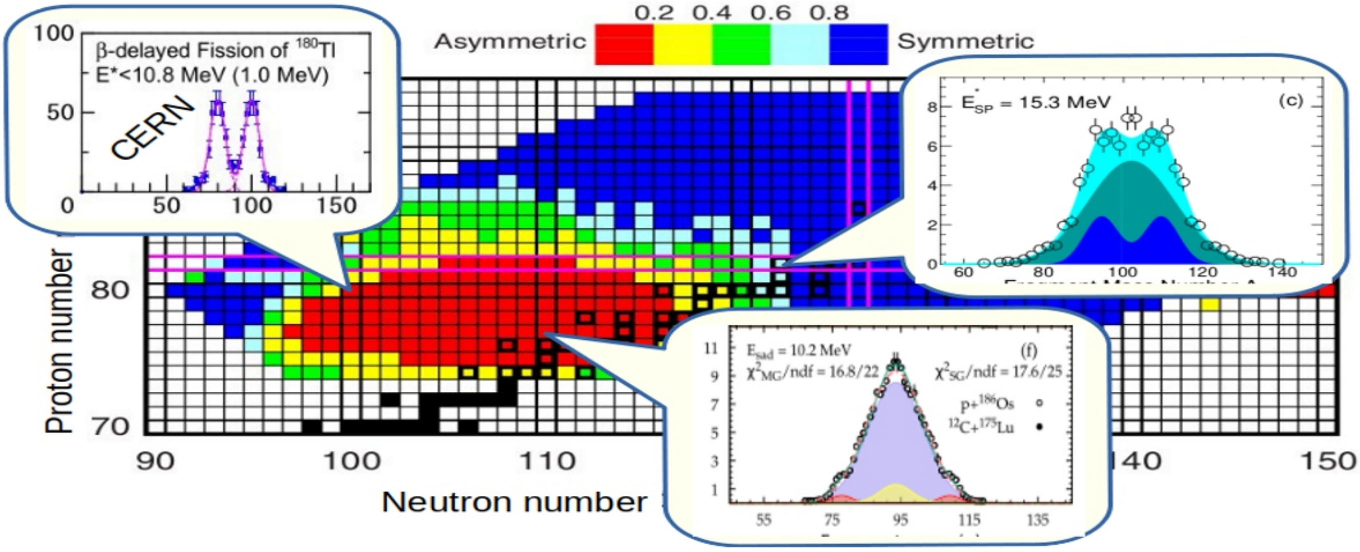


Fig.2: Brownian Shape Model (BSM) predicted ratio of the symmetric-yield to peak-yield for pre-actinide nuclei are shown in colored pixel. A few experimental mass distributions (mass number vs yields) in callouts show the experimental trend.

pre-actinide region. Several other studies [4-6] were performed using the BARC-TIFR Pelletron LINAC facility, which have improved our understanding of mass asymmetric fission in the pre-actinides and the fission process in general. A photograph of the experimental setup for the measurement of fission fragment mass distributions is shown in Fig.1. The setup consists of two large area Multi-Wire Proportional Chambers (MWPC) to measure the velocities and emission angles of the fission fragments in coincidence. Fragment masses are determined by applying momentum conservation.

Microscopic origin of asymmetry in fission of pre-actinides

Different theoretical models have proposed different mechanisms to explain the observed asymmetry in this region. The BSM model ascribe this to the neutron shell at 110 of the fissioning nuclei around the saddle deformation, which makes the outer saddle asymmetric. In a microscopic energy density functional (EDF) calculation, Scamps and Simenel have concluded the dominance of octupole effects, in most cases driven by neutron configurations. Another interpretation relates the asymmetric splits to prescission configurations involving molecular structures.

In Fig.2 the experimental trend is compared with the BSM model predictions. The BSM model predicts a large island of asymmetry (marked red) in the pre-actinide region. However, the experimental studies differ significantly with the predictions. It should be mentioned here the BSM predictions corresponds to excitation energy just above the fission barrier. The experimental data for ^{180}Hg also corresponds similar excitation energy. Other two data set pertaining particle induced fission has excitation energy around 10-15 MeV above the fission barrier. Though the contribution of the asymmetric component gradually diminishes with increase in excitation energy, the peak position associated with neutron or proton shell is expected to remain same. The experimental results for ^{180}Hg (in the left edge of the island) and ^{204}Pb (right edge of the island) agree well with the BSM model predictions in terms of magnitude and position of the asymmetric peak. However, the experimental results for ^{187}Ir (at the heart of the predicted island) differ significantly from the BSM model expectation. BSM model not only predicts large asymmetry, it also predicts very asymmetric fragments in low energy fission of ^{187}Ir. The experimental mass distributions of ^{187}Ir, shows an additional narrow symmetric contribution, which is in agreement with the $Z \approx 36$ stabilization in the light fragment.

While the presence of asymmetric mode in low energy fission of pre-actinide is confirmed by many experiments. Its origin is still debated and more experimental studies are required to chart the boundary and shape of the predicted island of asymmetry in the pre-actinide region. Our systematic study of the available experimental mass distributions, it was revealed that the mean value of Z of the light fragment, evaluated assuming unchanged charge density (UCD), varies

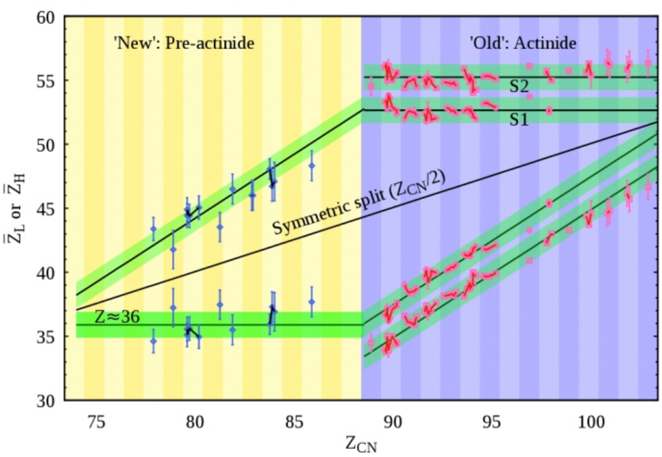


Fig.3: Evaluation of the peak position of the light and heavy fragment peaks corresponding to different asymmetric fission modes across the nuclear chart.

less as compared to that of the heavy fragment with Z the fissioning nuclei. No preferential population of neutron number was observed. Thus, it was concluded that the light fragment proton shells with $Z \approx 36$ provide the stabilization in asymmetric fission of pre-actinide nuclei [7]. The dominance of $Z \approx 36$ has been further reaffirmed in a recent GSI experiment through direct charge distribution measurement [8]. This is in contrast with the observations in the actinide region, where the proton shell stabilization in the heavy fragment was found to drive the asymmetry. The observed proton shell stabilization in the light fragment in low-energy fission of pre-actinide was also substantiated by the EDF calculation, which showed that the shell correction associated with the proton configuration in the pre-fragments dominates over that of the neutron configurations.

The evolution of the peak position of the light and the heavy asymmetric peaks is shown in Fig.3. It has rendered a striking connection between the pre-actinide and the actinide region, establishing the general dominance of proton shell in low-energy fission. It rises a fundamental question as why the proton configuration dominates in sharing of nucleons during fission process. Is the Coulomb repulsion between the protons keep the away from the neck, allowing specific shell configuring to manifest, while the neutron sub-systems are disturbed due to the presence of the neutron-rich neck. As discussed earlier, the position of the light fragment peak remains stable at $Z \approx 36$ and the mean Z of the heavy fragment increase monotonically taking them away from the position of the symmetric split as the Z of the fissioning nucleus increases. At the boundary of the pre-actinide and actinide a role reversal takes place, where the proton shell stabilization in the heavy fragment starts to dominate, forcing the light fragment to take the excess protons.

Role of Entrance Channel Dynamics in Fission of Pre-actinides

Dynamics also plays a very vital role in nuclear reaction in general and in heavy-ion induced fusion-fission reaction in particular. Dynamics in the entrance channel in heavy-ion fusion fission reaction might delay the equilibration of some of the degrees of freedoms, preventing formation of compound nucleus and altering the outcomes substantially. Fission fragment mass distribution is a sensitive probe to study the entrance channel dynamics, particularly the mass flow dynamics Quasifission, a non-compound (non-equilibrated) nuclear process, is being studied experimentally as well as theoretically with great vigor as it hinders formation of super-heavy elements. It strongly depends on the entrance channel parameters like charge product (or mass asymmetry), deformation of the colliding nuclei, shell closure and neutron excess in addition to the compound nucleus (CN) fissility. There are extensive experimental evidence of entrance dependence in fission of systems with higher fissility and entrance channel charge product. However, its extent in the sub-Pb region remained unexplored and largely neglected. Investigation of this aspect is essential for an accurate modeling of the excitation energy dependence of the microscopic effects discussed earlier. Particularly, ignoring quasifission might lead to ambiguity in the inferred multimodal fission recently observed in the pre-actinide region.

In order to address this issue, experiments were carried out to measure the fission fragment mass and total kinetic energy distribution for ^{191}Au, populated in i) ^{16}O+^{175}Lu and ii) ^{37}Cl+^{154}Sm reactions utilizing the BARC-TIFR Pelletron facility. Beam energies was so chosen that the excitation energy and the angular momentum populated are similar for both the reactions. Details of the measurements can be found in Ref. [4].

In Fig.4 the measured fragment mass distributions for both the systems are compared. As can be seen from the figure, the measured mass distribution for heavier projectile (^{37}Cl) is much wider as compared to the lighter projectile (^{16}O) induced reaction. The observed difference is much larger than that expected due to the small change in excitation energy and angular momentum. The statistical dependence of the mass distribution widths on excitation energy and angular momentum is also determined in the same experiment. Thus, it provided compelling evidence of presence of quasi-fission in the sub-lead region. The characteristic of the quasi-fission contribution is obtained by subtracting the distribution for ^{16}O+^{175}Lu reaction form that for ^{37}Cl+^{154}Sm reaction. The quasi-fission contribution, which has dynamic origin, is also found to overlap with microscopic asymmetric component. The Di-Nuclear System (DNS) model calculation, which reproduces the observed quasi-fission probability and its distribution, has revealed the persistence of shell effects in the emerging light fragments of the di-nuclear system. Thus, the study demonstrated for the first time that not only the microscopic shell effects, but the dynamics in the entrance channel also has a significant role in influencing the fission of nuclei in the newly identified island of mass asymmetry.

Comparison of the experimental mass ratio widths of neutron deficient nuclei near Pb is shown in Fig.5. The mass ratio is defined as the ratio of the fragment mass to the mass of the fissioning nucleus. The fitted mass ratio widths for most of the heavier projectile (^{35,37}Cl, ^{40,48}Ca and ^{48}Ti) induced and lighter projectile (^{13}C, ^{16}O and ^{24}Mg) Ca and Ti induced reactions involving both spherical as well as deformed targets exhibit significantly larger widths as compared to C - Mg induced reactions. Further, all the systems involving ^{154}Sm (deformed) target with heavy beams show an increase in the width with

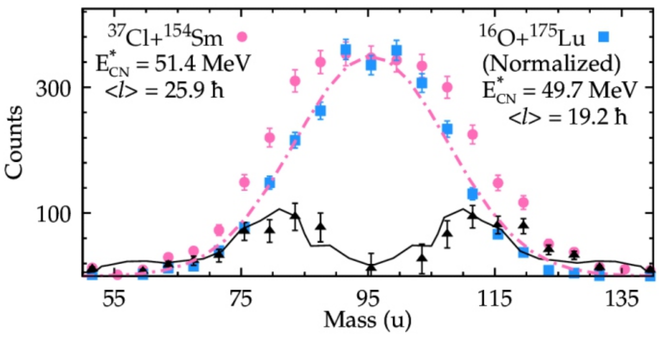


Fig.4: Experimentally measured fission fragment mass distributions for ^{16}O+^{175}Lu and ^{37}Cl+^{154}Sm reactions with similar excitation energy and angular momentum are compared. The difference between the two distributions is shown in black triangle. The continuous black line is the Di-Nuclear system (DNS) model prediction of quasi-fission contribution for the heavier system.

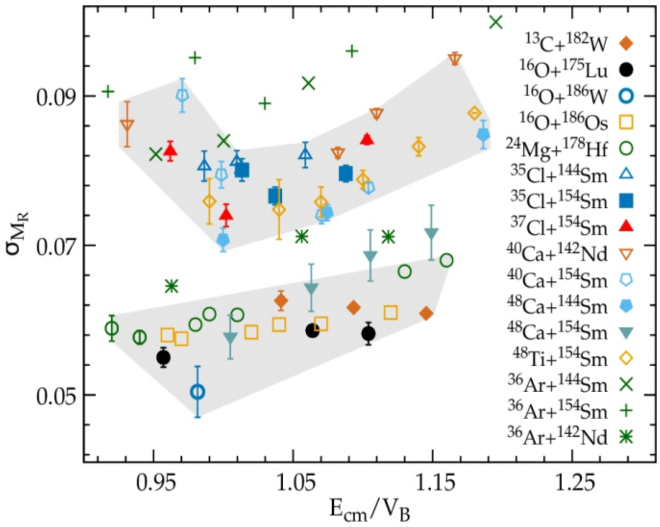


Fig.5: Width of the mass ratio distributions as a function of energy in centre-of-mass (E_{cm}) relative to the Coulomb barrier energy (V_B) are compared. The region of C, O, Mg and Cl, Ca (except $^{48}Ca+^{154}Sm$) are shaded separately to highlight the difference among them.

decreasing energy below the Coulomb barrier. In case of neutron rich $^{48}Ca+^{154}Sm$ system, the quasifission exhibits signature of fast time scale, i.e., observation of mass-angle correlation in asymmetric splits, which are clearly separated from the fusion-fission (symmetric) products. The widths of the symmetric distributions are found to be comparable to those of lighter ion induced reactions, thus having no significant contribution from quasifission in the symmetric region. While no such distinctly separate quasifission contribution is observed for $^{48}Ca+^{144}Sm$ and $^{40}Ca+^{154}Sm$, widths of the symmetric distribution for these systems are found to be larger as compared to those for $^{48}Ca+^{154}Sm$ system and other lighter ion induced reactions. The above induced reactions show distinctly different behavior as shown by the shaded regions. In general, Cl, comparison indicates that most of the systems involving heavier projectile are having contribution from the quasi-fission process.

Summary and outlook

Rapid progress was witnessed in last few years in low-energy fission of pre-actinides. Observation of mass asymmetric fission has provided an opportunity to test the fission models beyond the actinide region to improve their reliability. Different theoretical models have suggested different mechanism as the underlying driving force for the asymmetry. Our study suggests, proton shell stabilization in the light fragment mainly govern the asymmetric split.

Acknowledgments

We acknowledge the accelerator staff for the smooth operation of the machine. Technical help from P. Patale and A. Kumar during experiments are also acknowledged.

References

[1] A. Andreyev *et al.*, Phys. Rev. Lett. 105 252502 (2010).

[2] P. Möller *et al.*, Phys. Rev. C 91, 044316 (2015).

[3] R. Tripathi *et al.*, Phys. Rev. C 92 024610 (2015).

[4] S. Gupta *et al.*, Phys. Lett. B 803, 135297 (2020).

[5] S. Dhuri *et al.*, Phys. Rev. C 106, 014616 (2022).

[6] V. Kumar *et al.*, Phys. Rev. C 109, 014613 (2024).

[7] K. Mahata *et al.* Phys. Lett. B 825, 136859 (2022)

[8] Morfouace *et al.*, Nature 641, 339 (2025).