

# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

## Letter of Clarification to the ISOLDE and Neutron Time-of-Flight Committee

### **P-344: Study of the Di-nuclear System $^A\text{Rb} + ^{209}\text{Bi}$ ( $Z_1 + Z_2 = 120$ )**

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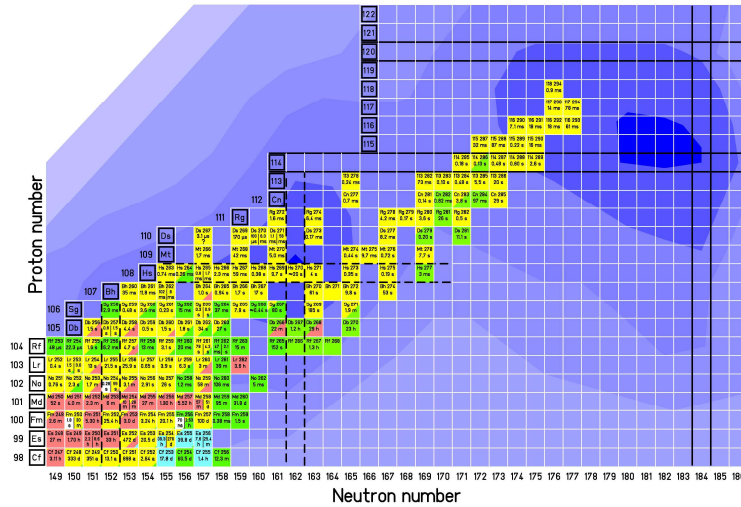
### **Abstract**

We present a Letter of Clarification concerning our approved proposal P-344 and related addendum P-344-ADD-1. The proposal aims at the search for predicted shell closures in the region of superheavy elements at  $Z=120$ ,  $N=184$ . This will be accomplished by studying quasi-fission and fusion-fission products created in collisions of nuclear systems with a total proton number  $Z=120$  and neutron number  $N=184$ . Originally, we proposed reactions of  $^{95}\text{Rb} + ^{209}\text{Bi}$  to reach compound systems  $^{304}120^*$ . Meanwhile, the technical developments at ISOLDE enable the production and post-acceleration of neutron-rich Ni beams at intensities which allow us to switch to the more asymmetric system  $^{66}\text{Ni} + ^{238}\text{U} \rightarrow ^{304}120^*$ . The aim of the addendum P-344-ADD-1 is to switch the reaction system from Rb+Bi to Ni+U. Following the request of the INTC, with this document we answer the open questions, summarize the experimental goal and method, and explain why we favour the reaction Ni+U.

**Total of granted shifts for P-344: 12**

**Remaining shifts: 12**

## I. Goal of the experiment



**Figure 1.** Chart of Nuclides showing the known isotopes of superheavy elements. The blue background represents shell correction energies (i.e. fission barriers) calculated in the macroscopic-microscopic model which assumes the magic numbers at  $Z=114$ ,  $N=184$ . Isotopes close to these numbers reveal relatively high fission barriers (“island of stability”). If the magic proton number is  $Z=120$  instead of 114, like predicted by mean-field models, the island of stability would be shifted toward larger  $Z$ , accordingly.

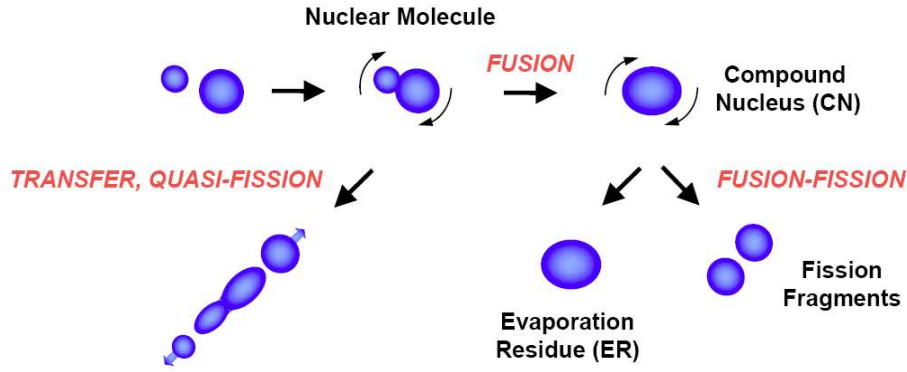
New spherical shell closures are predicted by theoretical model calculations in the region of superheavy nuclei. Most models expect a neutron shell closure at  $N=184$  while for the proton shell the results diverge and arrive at values of  $Z = 114$  or  $120$  or  $126$ . A double shell closure in that region will be reflected by an increase of the fission barriers for the respective isotope and its neighbors. Consequently, those nuclei will reveal an enhanced stability against fission and will form a so-called “island of stability” (Fig. 1).

The  $N$  and  $Z$  values where the island of stability is located are still not confirmed experimentally. Several isotopes with  $Z=114$  are known but they are seven or more neutrons far from the  $N=184$  shell (Fig. 1) and nuclei with  $Z=120$  or more were still not observed at all.

The only way to synthesize superheavy nuclei well beyond  $Z=100$  is fusion-evaporation reactions. But there are some serious bottlenecks which prevent reaching the island of stability, even on a long-term time scale:

- (i) Cross-sections of fusion-evaporation residues (ER) drop with increasing  $Z$ . Model calculations predict some 10 fb for isotopes with  $Z=120$ . This results in irradiation times of half a year or more to observe a single nucleus, even at beam intensities of  $10^{12}$  projectiles/s.
- (ii) Fusion-evaporation reactions with non-radioactive beams lead to neutron-deficient ERs and do not allow to reach the neutron number  $N=184$ .
- (iii) Fusion-evaporation reactions with some neutron-rich radioactive ion beams (RIBs) would allow to reach  $N=184$  but are not applicable because of the small RIB intensities.

We suggested a so far unique experimental approach to obtain information about the island of stability. It is not based on the direct production and study of  $Z=120$ ,  $N=184$  isotopes but profits from the complex reaction mechanism in heavy ion collisions combined with the application of neutron-rich radioactive ion beams.



**Figure 2.** Evolution paths of a heavy nuclear system. For details see text.

Figure 2 illustrates the evolution of a heavy nuclear system toward fusion. The first step is the capture of projectile and target nucleus due to the nuclear force. It leads to the formation of a composite system, usually termed as dinuclear system (DNS) or nuclear molecule. After DNS formation, which is accompanied by full kinetic energy dissipation, the system evolves by exchanging nucleons and energy. The nuclei can undergo complete fusion and form a compound nucleus (CN) which de-excites by evaporating nucleons, or by fission (Fusion-Fission, FF). But the DNS can also scission in two fragments before the CN is formed (Quasi-Fission, QF).

The cross-section,  $\sigma_{ER}$ , for the formation of a specific Evaporation Residue (ER) is determined by the strength of the QF and FF channels and can be written as

$$\sigma_{ER} = \sigma_{cap} \cdot P_{CN} \cdot P_{survival}$$

$\sigma_{cap}$  is the capture cross-section,  $P_{CN}$  the probability for CN formation and  $P_{survival}$  the probability that the CN survives fission and an ER is formed.  $\sigma_{cap}$ ,  $P_{CN}$  and  $P_{survival}$  depend on several parameters like beam energy, angular momentum, proton number or Coulomb barrier of the system.

The QF and FF cross-sections grow with increasing proton number and increasing Coulomb barrier. In (super)heavy systems the QF and FF channels dominate and  $\sigma_{ER} \ll \sigma_{QF} + \sigma_{FF}$ , which explains the tiny cross-sections of superheavy ERs.

Our experimental approach is to study QF and FF reactions from which we expect information about possible shell closures at  $Z=120$ ,  $N=184$ . Capture cross-sections are on the scale  $\sigma_{cap} = (10 - 100)$  mb, meaning that  $\sigma_{QF} + \sigma_{FF} \approx \sigma_{cap}$  is on the same scale. This allows QF and FF studies with good statistics even at beam intensities of  $10^6$  particles/s.

## II. Experimental approach

QF and FF reactions in (super)heavy systems have been extensively studied during the past years by different groups, including ourselves. Many experimental data and model calculations are available for nuclear systems up to  $Z=120$ . The results reveal some common trends.

- FF fragments and QF fragments which result from long-living ( $> 10^{-20}$  s) DNS reveal very similar properties, namely, symmetric mass distributions, isotropic angular distributions and full dissipation of kinetic energy. If the DNS or CN is driven by shell effects, they are revealed by the mass and total kinetic energy (TKE) distributions of these events.

- With increasing  $Z$  and increasing entrance channel Coulomb barrier  $Z_p \cdot Z_t$ , the values for  $\sigma_{\text{cap}}$  and  $P_{\text{CN}}$  decrease. As a consequence, the contribution of CN-like events also decreases.

- Experimental data and model calculations of QF and FF reactions reveal that shell effects do not only act in ERs, but also in long-living DNS and CN at low excitation energy ( $E^* < 40$  MeV). They are visible in the mass distributions of QF and FF fragments which have maxima around magic nuclei like  $^{132}\text{Sn}$  or  $^{208}\text{Pb}$ .

## II.1 Requirements

- $Z_p + Z_t = 120$ , where  $Z_p, Z_t$  are the proton numbers of projectile and target nucleus
- $Z_p \cdot Z_t$  as small as possible:  $^{66}\text{Ni} + ^{238}\text{U} \rightarrow Z_{\text{Ni}} \cdot Z_{\text{U}} = 2576$ ;  $^{95}\text{Rb} + ^{209}\text{Bi} \rightarrow Z_{\text{Rb}} \cdot Z_{\text{Bi}} = 3071$
- as large as possible mass asymmetry  $\eta_A = (A_t - A_p)/(A_t + A_p)$ ; asymmetric systems are preferred:  $\eta_A = 0.566$  for  $^{66}\text{Ni} + ^{238}\text{U}$  and  $0.375$  for  $^{95}\text{Rb} + ^{209}\text{Bi}$
- beam intensity:  $I_{\text{beam}} > 10^5$  pps on target

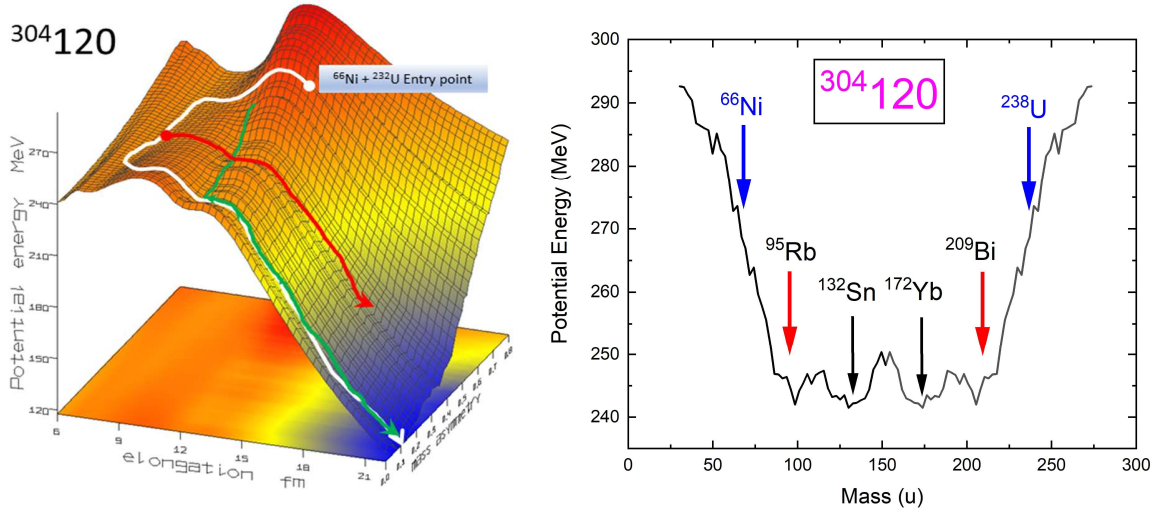
At the time of our proposal P-344, Rb+Bi was the optimal available system. Meanwhile, neutron-rich Ni beams were announced with sufficiently large intensity. The combination  $^{66}\text{Ni} + ^{238}\text{U}$  has a smaller entrance channel Coulomb barrier than Rb+Bi which results in an enhancement of those reaction products from which we expect the clearest signatures for shell effects, namely QF fragments from long-living DNS and FF fragments.

## II.2 Planned measurements

With the present experiment we are not looking for a signature that the element  $Z=120$  was synthesized. We will look for signatures of shell closures in the mass-energy distribution of the two-body products (QF and FF). This can be understood if we look at the potential energy surface (PES) at the scission point of the DNS.

Figure 3 (left) shows the PES for a  $^{304}120$  system represented as a function of the elongation of the DNS and the mass asymmetry of the binary fragments. Minima in the PES reflect shell closures in QF or FF fragments. Also represented are few possible paths of the DNS evolution. The white path leads to the compound nucleus that may eventually fission (FF). The red path is a possible alternative FF decay. The green one is the path that goes through QF instead of FF.

The asymmetric decay around mass asymmetry 0.1 results from the formation of the doubly-magic  $^{132}\text{Sn}$  and its partner  $^{172}\text{Yb}$ . It can occur in FF as well as QF reactions and leads to a local minimum in the PES. The other local minimum is created by the doubly magic fragment  $^{208}\text{Pb}$  and its partner  $^{96}\text{Sr}$ , and should predominantly be populated in FF reactions.



**Figure 3.** (left) Potential energy surface (PES) computed by the two-center shell model at the scission point for the compound system  $^{304}120$ ; (right) PES at the scission point

Figure 3 (right) is a cut through the PES at the scission point. The local minima in the potential provide maxima in the mass distribution. The experimental observation of the valley corresponding to  $^{132}\text{Sn}+^{172}\text{Yb}$  is the first step to look for signatures of  $Z=120$  and  $N=184$  shell closures. It is revealed by an increase of the cross-section of Sn-like and Yb-like events (“Sn peak”).

One advantage of this measurement is that even though the FF cross-section may be only few microbarn, QF which has cross-sections of (10 - 100) mb, can still give us the same signature for the Sn valley and for shell closures at  $Z=120$  and  $N=184$ . In other words, we do not need to form a compound nucleus to check the presence of the stability valley.

The relative contributions of QF and FF are more complex to be determined because of the complete overlap for some of the paths. Both, QF fragments which originate from long-living DNS and FF fragments reveal the same signatures like e.g. symmetric fragment mass distributions and small TKE. Therefore, it is necessary to measure in successive experiments also an excitation function and an angular distribution.

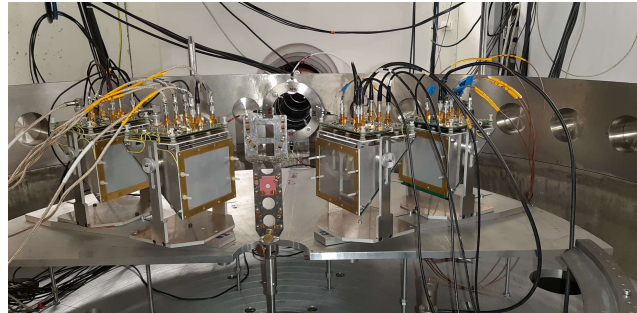
Following Fig. 3 (right), another important advantage of the reaction  $^{66}\text{Ni}+^{238}\text{U}$  over  $^{95}\text{Rb}+^{209}\text{Bi}$  can be deduced. Both reactions evolve mostly toward symmetric masses, namely there will only be a small contribution of target-like fragments heavier than the target and projectile-like fragments lighter than the projectile. If we choose the reaction  $^{95}\text{Rb}+^{209}\text{Bi}$  only two peaks should be observed, one corresponding to  $^{132}\text{Sn}$  and the other to  $^{172}\text{Yb}$ . Instead, in the case of  $^{66}\text{Ni}+^{238}\text{U}$  we would observe two additional peaks, corresponding to the double magic  $^{208}\text{Pb}$  and its partner  $^{96}\text{Sr}$ . The results in the significantly larger FF cross-section in Ni+Pb. Therefore, in the case of the  $^{66}\text{Ni}$  induced reaction we would observe an additional signature of the shell closures. If such signatures would not be found at a beam energy close to the Coulomb barrier, there would be no reason to run an excitation function.

### III. Experimental setup

The measurements will be performed with the two-arm spectrometer TOSCA (Time-Of-flight-sub-nano-second Spectrometer for Charged radiation Applications) which was constructed at the

University of Naples (Italy). It will replace the CORSET spectrometer of JINR Dubna (Russia) which was originally foreseen for our experiment.

An image of the TOSCA spectrometer is shown in Fig.4. It is a TOF spectrometer that implements the 2-Velocity method to measure the mass and the energy of the two fragments. What is measured is the time-of-flight and the position of each fragment from which the velocity vector is deduced. By applying the two-body kinematics the mass and energy of the fragments are obtained. The system can reach a mass resolution of 1 u and a time resolution of 60 ps (FWHM) with the use of digital electronics. TOSCA has been employed already in one experiment at GSI in 2022 and four experiments at JYFL in 2023, among them the above mentioned studies of QF and FF reactions in  $^{88}\text{Sr} + ^{208}\text{Pb}$ .



**Figure 4.** The TOF spectrometer TOSCA at JYFL (Finland). The system has two arms each with a start and stop detectors sensitive to position and based on microchannel plates.

#### IV. Answers to the Questions of the INTC

*• The intensity of the Ni beams are not higher than the Rb ones; the experimental plans should be revised accordingly.*

The following tables show the yields of Ni and Rb isotopes which are of interest for our experiments. The yields are given after contacting members of the ISOLDE technical team. To reach the N=184 neutron number we must apply  $^{66}\text{Ni}$  or  $^{95}\text{Rb}$ , respectively. The denoted yields for these isotopes show that the intensity of  $^{66}\text{Ni}$  is larger than the one of  $^{95}\text{Rb}$ .

Isotope	Yield / $\mu\text{C}$	Yield on target / pps	$T_{1/2}$
Ni-65 $\rightarrow$ $^{303}\text{120}^*$ (N=183)	$7 \cdot 10^7$	$5 \cdot 10^6$	2.5 h
<b>Ni-66 <math>\rightarrow</math> <math>^{304}\text{120}^*</math> (N=184)</b>	<b><math>1 \cdot 10^8</math></b>	<b><math>8 \cdot 10^6</math></b>	<b>55 h</b>
Ni-67 $\rightarrow$ $^{305}\text{120}^*$ (N=185)	$7 \cdot 10^5$	$5 \cdot 10^4$	21 s

Isotope	Yield / $\mu\text{C}$	Yield on target / pps	$T_{1/2}$
Rb-94 $\rightarrow$ $^{303}\text{120}^*$ (N=183)	$5 \cdot 10^8$	$4 \cdot 10^7$	2.7 s
<b>Rb-95 <math>\rightarrow</math> <math>^{304}\text{120}^*</math> (N=184)</b>	<b><math>2 \cdot 10^7</math></b>	<b><math>1.6 \cdot 10^6</math></b>	<b>377 ms</b>
Rb-96 $\rightarrow$ $^{305}\text{120}^*$ (N=185)	not available	---	203 ms



- *What is the requested beam energy?*

We are planning to run at an energy of 5.2 MeV/u, slightly (~1%) above the Coulomb barrier. This results in an excitation energy of the dinuclear system or CN, respectively, of 27 MeV.

$$E_{\text{beam}} = 5.2 \text{ MeV/u} \rightarrow E^*(\text{CN}) = 27 \text{ MeV}$$

- *What is feasible within the allowed 12 shifts? One beam at one energy, as initially allowed in P-344? Or do they want to use different beams at different energies?*

We are planning to run at one beam and one energy:  $^{66}\text{Ni}$  @ 5.2 MeV/u

- *The transmission rate is of the order of 5%, half of what the authors have accounted for in their proposal.*

With 5% transmission we arrive at the yields given in the above tables, namely,  $8 \cdot 10^6$  pps for  $^{66}\text{Ni}$  and  $1.6 \cdot 10^6$  pps for  $^{95}\text{Rb}$ . Still open is the question if 5% losses can be assumed also for  $^{95}\text{Rb}$  because of its short half-life of 377 ms (larger losses during charge breeding? breeding times?)

- *What will be the activity of the U target, which they plan to use?*

We will use  $^{238}\text{U}$  targets with thicknesses of a few  $100 \mu\text{g}/\text{cm}^2$ . The activity is well below 10 Bq and the targets can be handled like non-radioactive material. No special safety measures are required. Also, we have longstanding experience in the use of  $^{238}\text{U}$  targets.

- *Secondly, the Ni beams will contain some contaminants e.g. Ga. Moreover, the yields for these beams may not be stable over time. Will this be an issue for the experiment? The proponents should contact the technical groups at ISOLDE for more detail and account for this within their plans.*

We need more details on the contaminant Ga, namely what is the isotope and the expected energy (same as Ni energy?). With this information we know, if we can eliminate the reactions with Ga in the data analysis or if we have to install a TOF system before the target.

- *Third, it is not clear what the proponents hope to observe during their experimental campaign. What is expected to be a signature of the synthesis of the element  $Z=120$ ? In the addendum, it is mentioned that the cross section for the evaporation residue will be very small. What will be the experimental signature of the difference between the fusion-fission and the quasi-fission processes? Overall, once the possible running conditions have been established, what specific science goals will be achieved during the 12-shift experiment, noting that it is unlikely that all of the science proposed for the original 42-shift experiment will be met?*

see text in section II.2

## V. Literature

In the following are some articles which can give the referees additional information about the physics of QF and FF reactions and experimental technique.

1. G.G. Adamian et al., Eur. Phys. J. A **56**, 47 (2020).
2. S. Heinz et al., Eur. Phys. J. A **58**, 114 (2022).
3. E.M. Kozulin et al., “Evidence of quasifission in the  $^{180}\text{Hg}$  composite system formed in the  $^{68}\text{Zn}+^{112}\text{Sn}$  reaction”, Phys. Lett. B 819 136442 (2021).
4. E. Vardaci et al., Journal of Physics G Nuclear and Particle Physics **46**, 103022 (2019).
5. K.V. Novikov et al., Investigation of fusion probabilities in the reactions with  $^{52,54}\text{Cr}$ ,  $^{64}\text{Ni}$ , and  $^{68}\text{Zn}$  ions leading to the formation of  $Z = 120$  superheavy composite systems, Phys. Rev. C **102**, 044605 (2020).