

X-ray fluorescence from the element with atomic number $Z = 120$

M.O. Frégeau,¹ D. Jacquet,² M. Morjean,^{1,*} E. Bonnet,¹ A. Chbihi,¹ J.D. Frankland,¹ M.F. Rivet,² L. Tassan-Got,² F. Dechery,³ A. Drouart,³ L. Nalpas,³ X. Ledoux,⁴ M. Parlog,^{5,6} C. Ciortea,⁶ D. Dumitriu,⁶ D. Flueraşu,⁶ M. Gugiu,⁶ F. Gramegna,⁷ V.L. Kravchuk,⁷ T. Marchi,^{7,8} D. Fabris,⁸ A. Corsi,⁹ and S. Barlini¹⁰

¹ GANIL, CEA-DSM and IN2P3-CNRS, B.P. 55027, F-14076 Caen Cedex, France

² IPNO, CNRS/IN2P3, Université Paris-Sud 11, F-91406 Orsay Cedex, France

³ CEA-Saclay, IRFU/Service de Physique Nucléaire, F-91191 Gif sur Yvette Cedex, France

⁴ CEA, DAM, DIF, F-91297 Arpajon, France

⁵ LPC, CNRS/IN2P3, ENSICAEN, Université de Caen, F-14050 Caen Cedex, France

⁶ National Institute for Physics and Nuclear Engineering, RO-077125 Bucharest-Magurele, Romania

⁷ Istituto Nazionale di Fisica Nucleare, Laboratori Nazionali di Legnaro, I-35020 Legnaro (Padova), Italy

⁸ Istituto Nazionale di Fisica Nucleare, Sezione di Padova, I-35131 Padova, Italy

⁹ Istituto Nazionale di Fisica Nucleare, and Dipartimento di Fisica dell'Università di Milano, I-20133 Milano, Italy

¹⁰ Istituto Nazionale di Fisica Nucleare, and Dipartimento di Fisica dell'Università di Firenze, I-50125 Firenze, Italy

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An atomic clock based on X-ray fluorescence yields has been used to estimate the mean characteristic time for fusion followed by fission in reactions $^{238}\text{U} + ^{64}\text{Ni}$ at 6.6 MeV/A. Inner shell vacancies are created during the collisions in the electronic structure of the possibly formed $Z=120$ compound nuclei. The filling of these vacancies accompanied by X-ray emission with energies characteristic of $Z=120$ can take place only if the atomic transitions occur before nuclear fission. Therefore, the X-ray yield characteristic of the united atom with 120 protons is strongly related to the fission time and to the vacancy lifetimes. K X-rays from the element with $Z = 120$ have been unambiguously identified from a coupled analysis of the involved nuclear reaction mechanisms and of the measured photon spectra. A minimum mean fission time $\tau_f = 2.5 \times 10^{-18}\text{s}$ has been deduced for $Z=120$ from the measured X-ray multiplicity.

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Different nuclear physics models predict islands of stability for nuclei with atomic numbers Z larger than 114 [1–7]. The most efficient way to reach (or to approach) these islands of stability should be to achieve fusion between two heavy nuclei. However, the fusion-evaporation cross-sections are so small that the synthesis of super-heavy nuclei becomes extremely difficult [8]: even if compound nuclei are formed, they will be excited and, due to their high fissility, they will predominantly decay by fission, possibly after emission of a few particles[9].

Evidence for fusion, and thus for the existence of super-heavy nuclei, is quite difficult to obtain since the fission fragments from the compound nuclei are quite similar to fragments arising from quasi-fission processes [10–17]. In quasi-fission processes, composite systems are formed, but the nucleons are not trapped within a potential well and therefore do not form compound nuclei. The transient composite systems rapidly split in two fission-like fragments that cannot be distinguished event-by-event from true fission fragments. Typical characteristic times $t_{qf} \approx 10^{-21}\text{s}$ have been inferred for the quasi-fission process from angular distributions analyses of the fission-like fragments [16, 17]. Recently, the blocking technique in single crystals has been applied to reaction time measurements for three systems [18], possibly leading to compound nuclei with $Z = 114, 120$ and 124 . For the three studied systems, elastic, quasi-elastic and deep-inelastic reactions were found to be associated with short reac-

tion times. However, for the two heavier systems, sizable cross-sections of fusion followed by fission were found, evidenced by the detection of fission fragments from nuclei with $Z = 120$ and 124 surviving more than 10^{-18}s , a blue time 3 orders of magnitude longer than t_{qf} . By contrast, no evidence for such long fission times were found for the possibly formed isotopes with $Z = 114$.

In the present work, X-rays characteristic of the element $Z = 120$ have been searched for in coincidence with fission fragments in the reaction $^{238}\text{U} + ^{64}\text{Ni}$ at 6.6 MeV/A. Characteristic X-ray emission results from the filling of inner shell vacancies created during the fusion process [19, 20]. It can only be observed if the fission time scale is long enough to permit the vacancy decay. The chosen system is very similar to the $^{238}\text{U} + \text{Ni}$ system studied in [18], thus providing us with both a confirmation of the conclusions from [18, 21] and a validation of the X-ray fluorescence technique for further explorations of the super-heavy element region. Characteristic X-rays have been measured in coincidence with fission fragments in few experiments performed to study deep-inelastic [22] or fission reaction [23, 24] times. These experiments have stressed that the main difficulty comes from the huge background created essentially by γ -rays emitted by the fragments themselves, requiring therefore high statistics to extract weak signals.

A 2 mg/cm² thick ^{64}Ni target has been bombarded by $^{238}\text{U}^{31+}$ ions ($\approx 10^8$ ions \times s⁻¹) accelerated by GANIL

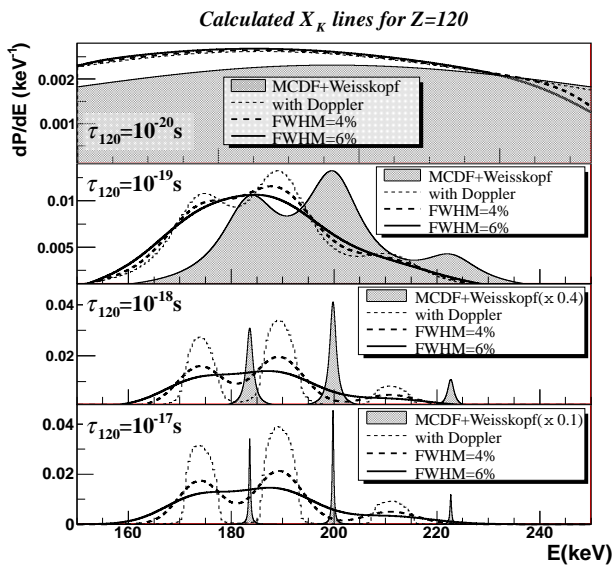


FIG. 1: Simulation of $Z=120$ K X-ray line shape for different fission times (see text for details).

at 6.6 MeV/A. Three adjacent telescopes detected heavy fragments ($Z \geq 6$), beyond the grazing angle, between $\theta = 15.9$ and 69° , at an average azimuthal angle $\varphi = 90^\circ$ (the azimuthal angles are referred with respect to a vertical plane). Each telescope consisted in an ionization chamber followed by a $5 \times 5 \text{ cm}^2$ double-sided silicon strip detector, covering a 53 msr solid angle. They provided us with the fragment detection angle, energy and atomic number (with a resolution of ± 3 for $Z = 92$). The VAMOS spectrometer [25] was operated at $\varphi = 270^\circ$, inside the grazing angle, between 10 and 25° . Its magnetic rigidity was adjusted to allow a simultaneous detection of fission-like fragments and elastically scattered projectiles. Three planar germanium detectors were operated under vacuum. They were located at 4 cm from the target, at the same polar angle $\theta = 127^\circ$ with respect to the beam direction, but at three different azimuthal angles $\varphi = 30, 150$ and 270° , covering altogether a 0.8 sr solid angle. In order to reduce the counting rate at very low energy, a 0.5 mm thick titanium foil was inserted in front of the detectors. The acquisition triggers were scaled-down for single events whereas all coincidences between detectors were registered. Nevertheless, due to the charge-state selection by VAMOS, very low statistics was obtained for triple coincidences between VAMOS, a telescope and a germanium detector and the search for X-rays from $Z = 120$ has been performed from the coincidences between the telescopes and the germanium detectors.

The Multi Configuration Dirac Fock approach (MCDF) [26–28] has been used to calculate the energies and transition probabilities of K X-rays from $Z=120$ [29, 30]. MCDF predicts, for a 1^+ ionization state, three

dominant K-rays ($K_{\alpha 2}$ at 183.6, $K_{\alpha 1}$ at 199.8 keV and $K_{\beta 1}$ at 222.7 keV), in good agreement with previous calculations [31, 32]. The transition probabilities of the associated K vacancies lead to a lifetime $\tau_K = 2.8 \times 10^{-18} \text{ s}$. Correlation diagrams for heavy and asymmetric systems [33, 34] displaying the electronic energy levels of the system as a function of the relative energy distance between the colliding ions show that emission from orbitals of intermediate molecular states [35] does not contribute to X-ray peaks since its energy changes rapidly with the interatomic distance both in the entrance and exit channels. Applying Weisskopf theory [35–37], a sizable broadening of the characteristic lines results from the finite lifetime τ_{120} of the $Z=120$ system. The filled curves in Fig. 1 show that the well separated lines predicted by MCDF progressively merge into a single broad peak when τ_{120} decreases. The disappearance of three separated lines is still enhanced by the Doppler broadening, as shown by the short dashed curves. For $\tau_{120} \lesssim 10^{-20} \text{ s}$ the very few fluorescence processes will only contribute as a background to the measured spectra: a characteristic peak cannot be observed for quasi-fission reactions.

The strongest smearing effect of the measured lines, besides piled up low energy transitions, results from the unavoidably broad electronic configuration distribution involved. Indeed, MCDF calculations have been performed for various electronic configurations and the characteristic energies are shifted up by 4% for a $119+$ charge state ion with respect to a $1+$ ion. The long dashed and full curves, calculated assuming overall gaussian shaped broadenings with $\text{FWHM} = 4\%$ and 6% , respectively, indicate that clean separations between the lines might only be obtained for very long fission times, furthermore with very high statistics.

The correlations between the atomic number Z and the energy E measured by the fragment telescopes are shown in Fig. 2 for selected angular bins between 15.9 and 69° . The overall behavior is in agreement with previous measurements [18] in which the reaction mechanisms had been identified thanks to a 4π detection of all charged products. For the most forward bins, deep-inelastic reactions are seen for $Z \approx 92$, separated from a distinct region between $Z \sim 65$ and $Z \sim 90$. The 4π detection performed in [18] showed for the latter region a multiplicity of 2 heavy fragments whose sum of atomic numbers is 120 accompanied by a negligible amount of light particles and clusters. This Z region (in which fusion-fission events were evidenced at 20° [18]) is thus exclusively populated by fragments arising from capture reactions (either quasi-fission or fusion followed by fission). The coincidences with VAMOS performed in the present experiment confirm that no fission fragments from uranium-like nuclei fill this Z region, but the rather poor Z resolution hindered for $Z > 80$ a perfect separation between fragments from capture reactions and uranium-like nuclei from deep-inelastic reactions. For $30 \lesssim Z \lesssim 65$, the

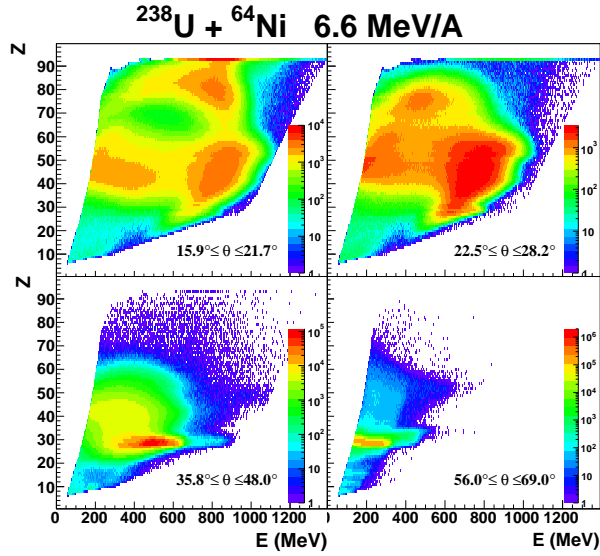


FIG. 2: Atomic number versus energy for the ions identified in the three telescopes.

detected fragments arise either from a sequential fission of excited uranium nuclei or from capture reactions. The fragments with $Z \approx 28$, correspond to more or less inelastic or elastic scattering of the target. Elastically scattered fragments become highly dominant in the most backward angular bin, beyond the target grazing angle.

The energy spectra of photons measured by the germanium detector at $\varphi = 270^\circ$ in coincidence with elastically scattered target nuclei recoiling between 56 and 69° , and with fragments with $35 \leq Z \leq 90$ are presented in Fig. 3. The top-left panels present the spectra as measured, whereas the bottom-left panels present the spectra after background subtraction. The high counting rates make mandatory random coincidence corrections. The random coincidence energy spectrum has been determined from the photon spectra measured when a fragment triggered the acquisition and no coincidence with a germanium detector was detected during a 500 ns coincidence window. The photon energy spectra were then acquired during a $6 \mu\text{s}$ gate following the coincidence window, thus equivalent to a randomly opened counting gate. An iterative correction procedure has been applied using this random coincidence spectrum, leading to the spectra shown in the right parts of Fig. 3. For elastic scattering, above 130 keV, the characteristic pattern of uranium decay via rotational bands after Coulomb excitation is observed (rays expected at 246.2 , 201.5 and 151.7 keV for ^{238}U , applying Doppler shifts corresponding to the average detection angle). The three peaks have shoulders towards high energy due to piled up 20 keV uranium L X-rays. The lower energy part of the spectrum is dominated by the uranium $K_{\alpha 1}$ X-ray expected with Doppler shift at 94.5 keV mixed with the $K_{\alpha 2}$ ray expected at

90.4 keV and by the $K_{\beta 1}$ ray expected at 106.4 keV. The decay of the 4^+ uranium level of the rotational band by a 103.5 keV γ can be hardly seen at 98.7 keV only after random coincidence correction. For the coincidence with $35 \leq Z \leq 90$, besides low energy uranium X-rays (not shown), two peaks can be seen in the measured spectrum around 150 keV and 200 keV, possibly reminding one of the γ -rays from uranium. However, the 200 keV peak is much broader than the 150 keV one and also much broader than the 200 keV peak observed either in coincidence with elastic scattering or in inclusive measurements. Furthermore, the random coincidence correction reduces strongly the peak at 150 keV (as expected for an uranium γ -ray since the time scale for rotational band decay is much longer than the time scale for fission), whereas the 200 keV peak is only slightly affected in a narrow energy range. The broad peak observed between 175 and 225 keV is therefore populated by true coincidences with fragments of fission or quasi-fission.

The origin of the peak at 200 keV has been investigated considering 4 bins of atomic numbers: $35 \leq Z < 50$, $50 \leq Z < 66$, $70 \leq Z < 80$ and $80 \leq Z < 91$. The two first bins are populated by fragments arising either from uranium fissions or from capture reactions whereas the two others are only populated by fragments from capture reactions (with some contamination from uranium-like nuclei for the last one, due to Z resolution). Despite large statistical errors due to the poor signal to noise ratio, a peak at 200 keV could be unambiguously identified for the three germanium detectors and the four Z bins. This is illustrated by the insert in Fig. 4 that presents the energy spectrum measured by the germanium detector at $\varphi = 30^\circ$ in coincidence with $80 \leq Z < 91$, the case with the lowest statistics. No significant differences in the peaks registered at $\varphi = 30, 150$ and 270° could be observed, except for $35 \leq Z < 50$ where a contribution of a γ -ray from a uranium fission fragment could be identified around 180 keV from the differences in the Doppler shifts for the various azimuthal angles involved. Except for this bin, Fig. 4 shows that the same photon multiplicity between 175 and 225 keV is measured for the three detectors. All these observations lead to the conclusion that the 200 keV peak observed in coincidence with $50 \leq Z < 91$ arises from a composite system moving in the beam direction (no difference in the Doppler effects at different detection angles, excluding thus any emission from the detected or complementary fragment). It must be noted in addition that the highest multiplicity in Fig. 4 is observed between $70 \leq Z < 80$, as expected for an emission by the composite system since this Z region is the only one exclusively populated by $Z = 120$ fission and quasi-fission events[18]. Considering the very low charged particle ($Z < 6$) multiplicities measured in coincidence with $70 \leq Z < 80$ (see the above discussion about Fig.2 and [18]), all the protons of the projectile and of the target constitute this composite system. Since the

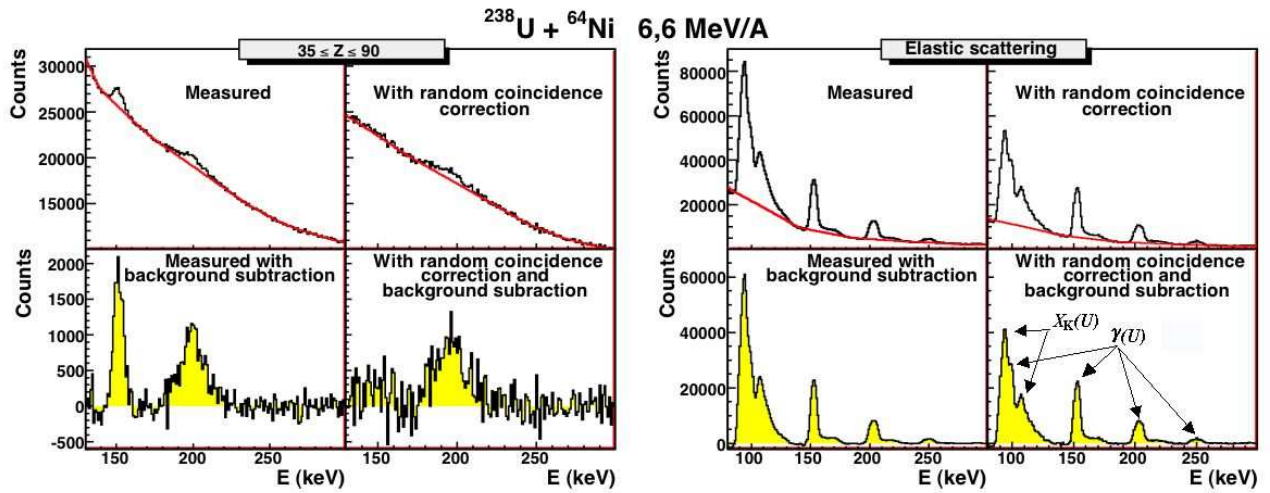


FIG. 3: Photon spectra registered at $\varphi = 270^\circ$ in coincidence with fragments with $35 \leq Z \leq 90$ for the left part and with elastic scattering reactions for the right part.

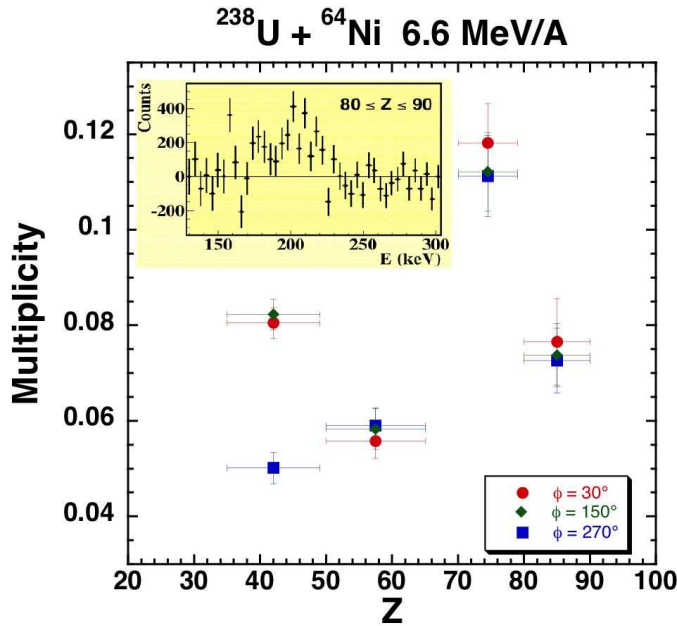


FIG. 4: Photon multiplicity between 175 and 225 keV taking into account the detection efficiency. Only the statistical errors are indicated. The horizontal bars indicate the Z integration ranges. The insert presents, for the bin with the lowest statistics, the energy spectrum measured at $\varphi = 30^\circ$ after background subtraction and random coincidence correction.

most probable energy of the broad line at 200 keV is located between the MCDF values calculated for a $Z = 120$ $K_{\alpha 1}$ line for the two extreme charge-state configurations $1+$ and $119+$, the 200 keV peak can be unambiguously attributed for $70 \leq Z < 80$ to K X-rays emitted by the $Z = 120$ element. Indeed, for this Z selection that weeds out target-like and projectile-like nuclei as well as uranium-

like fission fragments, the analysis presented above shows that the random coincidences have been efficiently suppressed and excludes any scenario in which 200 keV photons would be emitted by the detected fragment or its partner, whatever the reaction mechanism is (compound fission, or quasi-fission).

A K X-ray multiplicity $M_{120} = 0.11 \pm 0.02$ can thus be inferred for $70 \leq Z < 80$, taking into account the statistical error as well as the systematic error arising from detection efficiency determination. As asserted by Fig. 1, a characteristic K X-ray can only be observed for fission times $\tau_{120} \geq 10^{-19}$ s. A more accurate estimate of the minimum mean fission time can be reached assuming for $Z = 120$ independent exponential distributions for the fission time and for the vacancy decay and a fluorescence yield equal to 1. In this case, a simple correlation between τ_{120} and the vacancy lifetime τ_K can be deduced: $\tau_K = \tau_{120} \times (P_K/M_{120} - 1)$, where P_K is the K vacancy creation probability during the fusion process. P_K has been inferred from the uranium K X-ray multiplicity P_{el} for projectile elastic scattering detected by VAMOS, considering the similar atomic impact parameters associated to elastic scattering and to fusion. Since only the incoming part of the trajectory must be taken into account in the case of fusion, the simple approximation $P_K = P_{el}/2$ has been made. Considering a coherent addition of the incoming and outgoing K electron excitation amplitudes in the case of elastic scattering would lead to a slightly lower P_K value [20, 38], resulting in longer fission times τ_{120} . This approximation leads thus to a minimum value for the fission time and therefore to the smallest proportion of fusion among the capture reactions. In order to determine P_{el} , the K_{α} yield has been derived from a gaussian fit to the measured K_{α} peak in coincidence with elastically scattered projectiles. To suppress the contri-

bution of the unseparated 103.5 keV γ -ray from ^{238}U , the number of K_β emission has been inferred from the tabulated ratio between K_α and K_β yields. Then, the contribution of K X-rays resulting from internal conversion of the uranium rotational E2 cascade has been subtracted as in [22]. This procedure leads to $P_{el} = 0.27 \pm 0.07$, the large uncertainty resulting mainly from the procedure itself and from the detection efficiency determination. This P_{el} value is slightly higher than the one that can be inferred from existing measurements and calculations for similar systems at somewhat lower energies [39, 40]. Nevertheless assuming for P_{el} a dependence on the projectile velocity similar to the one observed for the K-shell ionization cross section [41] our values are just consistent with the previous measurements.

Taking into account the large uncertainties on P_K and M_{120} as well as the one on the MCDF vacancy lifetime ($\pm 20\%$) resulting from the large amount of possible electronic configurations, and assuming all the detected X-rays arise from atoms with a single nuclear lifetime, a minimum mean fission time $\tau_{120}^{min} = 2.5 \times 10^{-18}$ s can be inferred. Conversely, assuming a bimodal time distribution with very fast reactions for which no X-ray can be emitted and with very long fusion-fission reactions for which all the existing vacancies decay before fission, a minimum percentage of 53% can be inferred for fusion followed by fission among the detected capture reactions leading to fragments with $70 \leq Z < 80$.

It must be stressed that the minimum mean fission time τ_{120}^{min} is at least hundred times longer than the longest lifetimes of giant composite systems calculated in transuranium ion collisions [42]. Our asymmetric system presents by contrast to the systems of [42] a potential well corresponding to the compound nucleus and very long fission times imply huge nucleon exchanges between the partners during the contact step. Therefore, the composite systems are inevitably driven towards a total equilibration of all their degrees of freedom and compound nuclei are formed. The high inferred percentage of fusion-fission among the detected capture reactions seems in contradiction with the commonly assumed strong dominance of quasi-fission mechanisms for such heavy systems. However, this latter assumption comes essentially from extrapolations of mass-angle correlations measured for lighter systems [16, 43], assuming that symmetric fission follows fusion. Such assumptions and extrapolations have been indeed done in order to infer fusion-fission cross-section for our system [44], but they are definitively not supported by the reaction time measurements and the Z distribution that imply mass asymmetric fissions, at least in the fragment angular range covered in the present experiment and in [18].

The present work confirms thus previous fission time results obtained by a quite different experimental technique and provides us with evidence for transiently formed unbinilium elements characterized by their elec-

tronic inner shell structure.

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* Electronic address: morjean@ganil.fr

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