X-Ray Fluorescence from the Element with Atomic Number $Z = 120$

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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}U + ^{64}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 a 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 $\tau_f = 2.5 \times 10^{-18}$ 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 heavy nuclei. However, the fusion-evaporation cross sections are so small that the synthesis of superheavy nuclei becomes extremely difficult [8]: Even if compound nuclei (CNs) are formed, they are excited and, due to their high fissility, they decay predominantly by fission, possibly after the emission of a few particles[9].

Evidence for fusion, thus for superheavy nucleus formation, is quite difficult to obtain since fission fragments (FFs) arising from CNs are quite similar to fragments from quasifission (QF) processes [10–17]. Composite systems are formed during QF processes, but the nucleons are not trapped within a potential well and therefore do not form CNs. The transient composite systems rapidly split in to two fissionlike fragments that cannot be distinguished event-by-event from true FFs. Characteristic times $t_{\mathrm{qf}} \approx 10^{-21}$ s have been inferred for QF processes from angular distribution analyses of the fissionlike fragments [16,17]. Recently, the blocking technique in single crystals has been applied to reaction time measurements for three systems [18], possibly leading to CNs with $Z = 114$, 120, and 124. Sizable productions of nuclei with $Z = 120$ and 124 surviving more than 10^{-18} s, a time 3 orders of magnitude longer than t_{qf}, were observed.

In the present work, x rays characteristic of the element $Z = 120$ have been sought in coincidence with FFs in the reaction ^{238}U + ^{64}Ni at 6.6 MeV/A. Characteristic x-ray emission follows 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}U + 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 to probe the superheavy element stability. Characteristic x rays were measured in coincidence with FFs in a 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/cm^2 thick ^{64}Ni target was bombarded by ^{238}U^{31+} ions ($\approx 10^8$ ions $\times$ s^{-1}) accelerated by GANIL at 6.6 MeV/A. Three adjacent telescopes detected fragments ($Z \geq 6$), beyond the grazing angle, between $\theta = 15.9°$ and 69°, at an average azimuthal angle $\varphi = 90°$ (with respect to a vertical plane). Each telescope consisted of an ionization chamber followed by a 5×5 cm^2 double-sided silicon strip detector, covering a 53 m sr 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 $\varphi = 270°$, inside the grazing angle, between 10° and 25°. Its magnetic rigidity was adjusted to allow a simultaneous detection of fissionlike 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 $\theta = 127°$ 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. 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 were obtained for triple coincidences between VAMOS, a telescope, and a germanium detector; therefore, x rays from $Z = 120$ could only be sought among the coincidences between the telescopes and the germanium detectors.

The multiconfiguration 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 associated transition probabilities lead to a lifetime $\tau_K = 2.8 \times 10^{-18}$ s. Correlation diagrams for heavy and asymmetric systems [33,34] displaying the electronic energy levels of the system as a function of the relative distance between the colliding ions show that the 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 (short dashed curves). For $\tau_{120} \lesssim 10^{-20}$ s, the very few fluorescence processes will only contribute as a background to the measured spectra: a characteristic peak cannot be observed for QF reactions.

The strongest spreading of the lines, besides piled up low energy transitions, results from the unavoidably broad electronic configuration distribution involved. MCDF shows that the characteristic energies are 4% higher for a 119+ charge-state ion than for a 1+ ion. The long dashed and full curves, calculated assuming overall Gaussian broadenings with full width half maximum (FWHM) = 4% and 6%, respectively, indicate that clean separations between the lines might only be obtained for very long fission times.

The correlations between the atomic number Z and the energy E measured by the fragment telescopes are shown in Fig. 2 for angular bins between 15.9° and 69°.

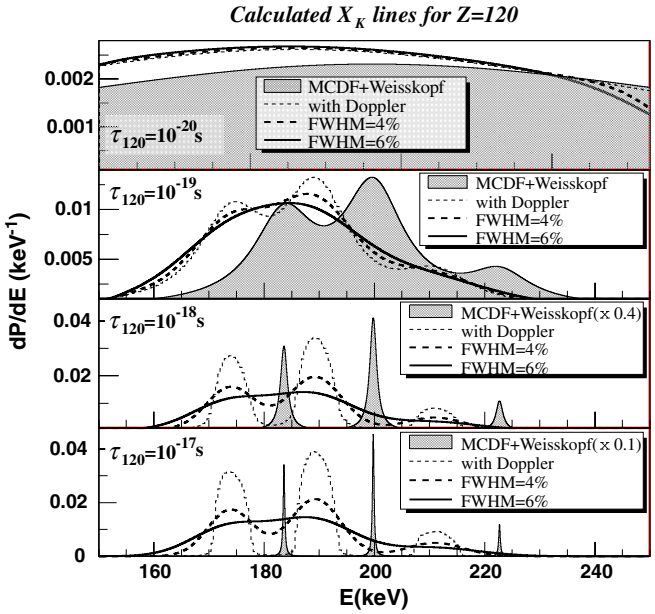

FIG. 1 (color online). Simulation of $Z = 120$ K x-ray line shape for different fission times (see text for details).

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

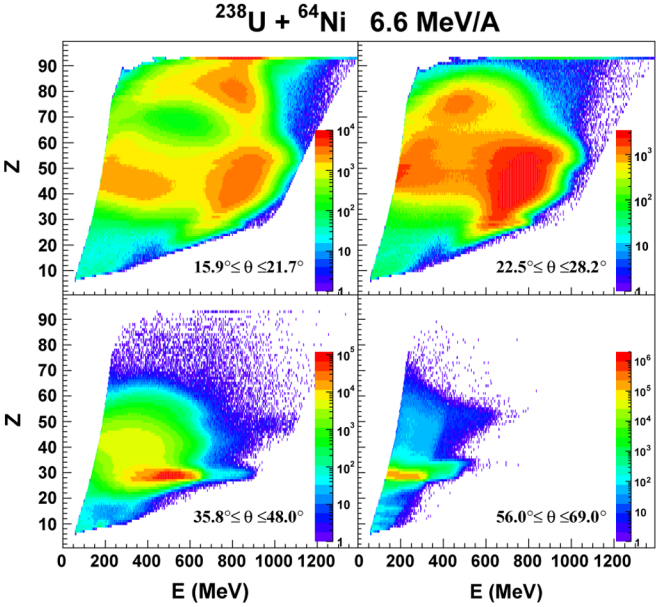

FIG. 2 (color online). Atomic number versus energy for the ions identified in the three telescopes.

were evidenced at 20° [18]) is thus exclusively populated by fragments arising from capture reactions (either QF or fusion followed by fission). The coincidences with VAMOS performed in the present experiment confirm that no FFs from uraniumlike nuclei fill this Z region, but the rather poor Z resolution hindered for $Z > 80$ a perfect separation between fragments from capture reactions and uraniumlike nuclei from deep-inelastic reactions. For $30 \lesssim Z \lesssim 65$, the detected fragments arise either from a sequential fission of excited uraniumlike nuclei or from capture reactions. The fragments with $Z \approx 28$ correspond to the inelastic or elastic scatterings of the target.

The energy spectra of photons measured by the germanium detector at $\varphi = 270°$ 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 the 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 μs gate following the coincidence window and, 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 is observed. The peaks present shoulders toward 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_α x rays. The $4^+ \rightarrow 2^+$ transition at 103.5 keV can be hardly seen after a random coincidence correction. For the coincidences with $35 \leq Z \leq 90$, two peaks show up 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 fission or QF fragments.

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 uraniumlike 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 a photon energy spectrum 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 FF 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

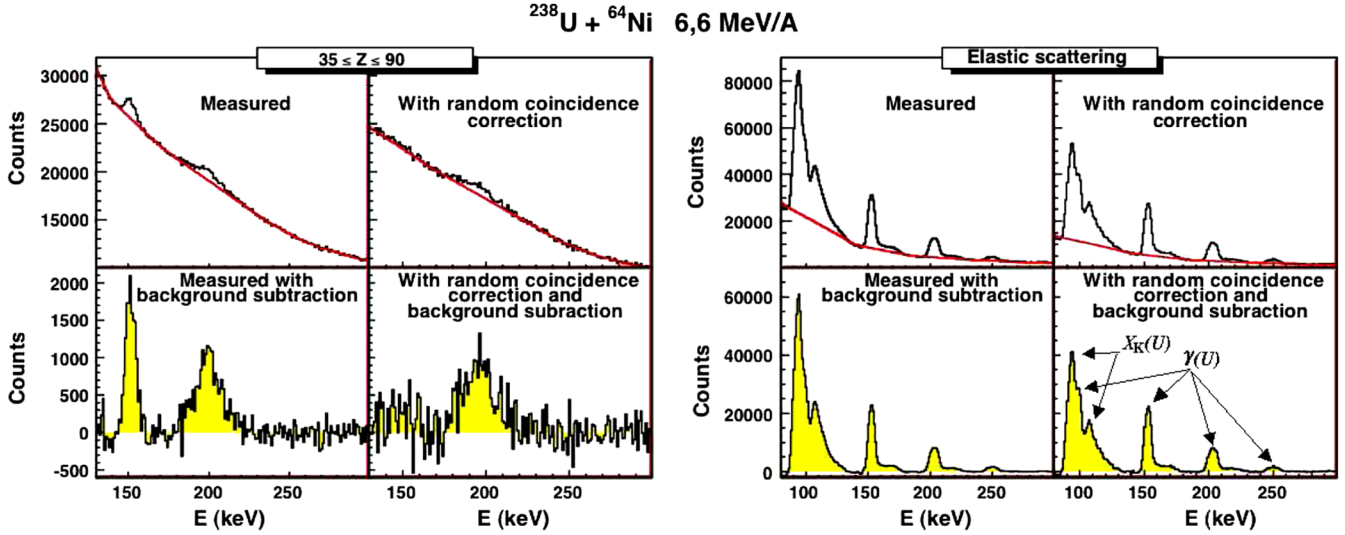

FIG. 3 (color online). Photon spectra at $\varphi = 270°$ in coincidence with fragments with $35 \leq Z \leq 90$ for the left part and with elastic scattering reactions for the right part.

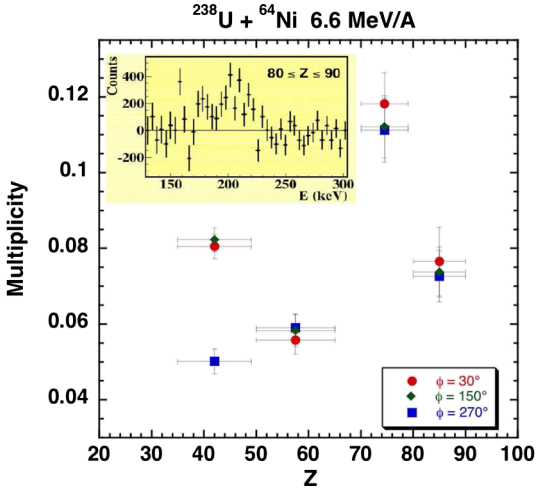

FIG. 4 (color online). Photon multiplicity between 175 and 225 keV taking into account the detection efficiency. Only the statistical errors are indicated. The horizontal bars indicate the Z integration ranges. The insert presents, for the bin with the lowest statistics, the photon energy spectrum recorded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

shows that the same photon multiplicity is measured between 175 and 225 keV 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 QF events. 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]) imply that all the protons of the projectile and the target constitute this composite system. Since the 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 weeding out targetlike and projectilelike nuclei as well as FFs from uraniumlike nuclei, 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 QF).

A K x-ray multiplicity $M_{120} = 0.11 \pm 0.02$ can be inferred for $70 \leq Z < 80$, considering the statistical error and the systematic error arising from the 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. Then, 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 is inferred from the uranium K x-ray multiplicity P_{el} for projectile elastic scattering detected by VAMOS, considering the similar atomic impact parameters associated with elastic scattering and with fusion. Since only the incoming part of the trajectory must be taken into account in the case of fusion, the simple approximation $P_K = P_{\mathrm{el}}/2$ 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 gives, therefore, only access to a minimum τ_{120} value and to a minimum 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 contribution of the unseparated 103.5 keV γ ray from ^{238}U, the K_β yield has been inferred from the tabulated ratio between K_α and K_β lines. Then, the contribution of K x rays following the internal conversion of uranium γ rays has been subtracted as in [22], leading to $P_{\mathrm{el}} = 0.27 \pm 0.07$. Assuming for P_{el} a dependence on the projectile velocity similar to the one observed for the K-shell ionization cross section [39], our value, although slightly higher, is just consistent with previous measurements[40,41].

Considering the large uncertainties on P_K and M_{120} along with 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 $\tau_{120}^{\mathrm{min}} = 2.5 \times 10^{-18}$ 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 $\tau_{120}^{\mathrm{min}}$ is at least 100 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 CN. Very long fission times imply huge nucleon exchanges between the partners during the contact step.

Therefore, the composite systems are inevitably driven toward a total equilibration of all their degrees of freedom and CNs are formed. The high inferred percentage of fusion-fission among the detected capture reactions seems in contradiction with the commonly assumed strong QF dominance 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 have been indeed done in order to infer a fusion-fission cross section for our system [44], but they are definitively not supported by the reaction time measurements and the Z distribution which imply mass asymmetric fissions, at least in the angular ranges covered in the present experiment and in [18].

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

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