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Prompt γ-ray spectroscopy of isotopically identified fission fragments

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Measurements of prompt Doppler-corrected deexcitation γ rays from uniquely identified fragments formed in fusion-fission reactions of the type ^{12}C(^{238}U, ^{134}Xe)Ru are reported. The fragments were identified in both A and Z using the variable-mode, high-acceptance magnetic spectrometer VAMOS. States built on the characteristic neutron configurations forming high-spin isomers (7^- and 10^+) in ^{134}Xe are presented and compared with the predictions of shell-model calculations using a new effective interaction in the region of $Z \geqslant 50$ and $N \leqslant 82$.

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Probing the properties of the neutron-rich nuclei around doubly magic nuclei, in areas of the periodic table traditionally produced by fission, is the focus of the next generation of high-intensity isotope-separator on-line (ISOL) radioactive-beam facilities. The excitation spectra of fission fragments span a large variety of properties. These features include both few-particle excitations in spherical nuclei near closed and doubly closed shells and quadrupole, octupole, or triaxial collective excitations in deformed nuclei [1–5]. Additionally, such studies provide important inputs for the investigation of nuclear processes in stars [6,7]. The insight into exotic nuclei around doubly magic nuclei having classical shell closures (28, 50, and 82) is currently one of the milestones for nuclear physics, for understanding the evolution of shell gaps far from the valley of stability [8]. Nuclei around ^{132}Sn present a unique opportunity to study properties with only a few valence particles or holes outside doubly closed shells. Spectroscopy of states in these nuclei provides valuable constraints for matrix elements of effective residual interactions used in the shell-model calculations and thus improves the predictive power of such interactions.

Fragments originating from fissioning systems at low excitation energy e.g., produced in spontaneous fission, reactions induced by Coulomb excitation, nucleon transfer, and low-energy beams of proton/neutron on actinides, mainly allowed the spectroscopic study of nuclei produced in asymmetric mass splits. Alternatively, the spectroscopy of nuclei produced in symmetric mass splits can be studied using fissioning systems at high excitation energy and/or angular momentum as in heavy-ion fusion-fission reactions [1,2,9–12]. The prompt γ-ray spectroscopy of a particular fission fragment is challenging because of the presence of a large background originating from γ-ray transitions from hundreds of other fission fragments [1,2]. The first γ-ray-spectroscopy studies of fission fragments were reported by Cheifetz *et al.* in 1970 [13]. The advent of large γ-ray detector arrays allowed the use of high-fold γ-ray coincidences for the spectroscopy of prompt γ rays [1,2,12], and additional improvements in the sensitivity were obtained by requiring the detection of both fission fragments in a gas detector [14]. This method relies on the knowledge of the γ-ray cascades of either the nucleus under study or its complementary fragment. The use of a spectrometer for the identification of fission fragments has been restricted to the study of isomeric states [10,15,16]. The prompt spectroscopy of states populating these long-lived isomers is limited because of experimental constraints. Very recently, studies made at Legnaro National Laboratory using fission reactions induced by Coulomb excitation or transfer in the ^{136}Xe + ^{238}U system reported prompt γ-ray spectroscopy of isotopically identified light fragments [17].

This work reports on a sensitive technique for prompt γ-ray spectroscopy by using a magnetic spectrometer for isotopic identification (both A and Z) of the fragments produced in a fusion-fission reaction of ^{238}U with ^{12}C in inverse kinematics at an energy around the Coulomb barrier. The power of the technique lies in the possibility of obtaining the Doppler-corrected γ-ray spectra for both detected and undetected fission fragments in the spectrometer. This technique is applied to the study of the prompt γ-ray spectroscopy above the high-spin isomers in ^{134}Xe. The new levels found in ^{134}Xe have been interpreted based on large-scale shell-model (LSSM) calculations using the new interaction GCN5082 [18].

The experiment was performed using a 1.45-GeV ^{238}U beam with a typical intensity of $\sim 10^9$ pps, from the CSS1 cyclotron at GANIL, incident on a ^{12}C target 100 μg/cm^2 thick.

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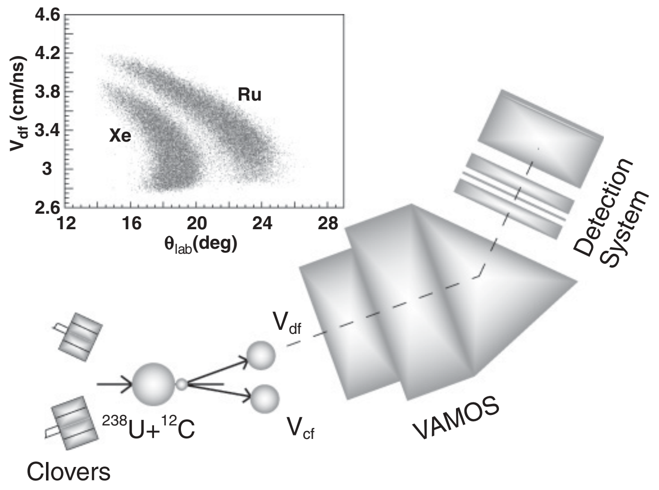

FIG. 1. Schematic layout of the experimental setup: The fission fragments were identified in VAMOS [20], and the prompt γ rays emitted from the fragments in-flight were detected in two EXOGAM clover detectors [21]. The velocities (V_{df}) as a function of the laboratory angle of the detected fragment for a typical pair of fission fragments, Ru or Xe, from the fissioning ^{250}Cf parent are shown in the inset.

This system was previously studied using ^{12}C beams [11,19]. The large-acceptance, variable-mode spectrometer VAMOS [20], placed at 20° with respect to the beam axis, was used to identify the fission fragments. These fragments are confined within a kinematic cone of $\sim$25°; only one fragment was detected per fission. A schematic layout of the experimental setup is shown in Fig. 1. The focal-plane detection system of the spectrometer consisted of two position-sensitive drift chambers separated by a distance of 1 m, a secondary-electron detector (SeD), placed between the two drift chambers, a segmented ionization chamber, and a 21-element Si wall. The Z identification was obtained from the energy loss measured in the ionization chamber and the residual energy in the Si wall. The measured parameters at the focal plane along with the known magnetic field were used to reconstruct the magnetic rigidity, mass (A), mass/charge (A/q), angle after the reaction, and path length on an event-by-event basis. The time of flight of the fragments was measured by the SeD, relative to the radio frequency of the cyclotron, and was used to obtain the velocity. The inset of Fig. 1 shows the velocities for Xe and Ru fragments detected in VAMOS as a function of the laboratory angle. The measurements were made over various settings of magnetic rigidity of the spectrometer [22]. Further details are given in Refs. [20] and [22]. These measurements take advantage of inverse kinematics, high recoil energies, and kinematic focusing of the fragments. The nuclei identified in our work ranged from the isotopes of Zn ($A = 70$ to 79) to Nd ($A = 137$ to 154) with a resolution of $\Delta Z/Z \sim 1.5 \times 10^{-2}$ [Fig. 2(a)] and $\Delta A/A \sim 6 \times 10^{-3}$. Figures 2(b) and 2(c) show a typical mass spectrum of light and heavy fragments for the isotopes of Zr and Xe, respectively.

The Doppler-corrected γ-ray spectra, in coincidence with the isotopically identified fragments, were obtained using two fully Compton-suppressed segmented clover detectors placed at backward angles (Fig. 1). The angle between the γ ray (from the segment of the relevant clover detector) and the velocity vector of the emitting fission fragment were used to obtain the γ-ray energy in the rest frame [23]. Two velocity vectors were used for each event to obtain the γ-ray spectrum of either (i) the fragment detected in VAMOS (V_{df}) or (ii) the undetected complementary fragment (V_{cf}). The former was measured in the VAMOS spectrometer, whereas the latter was deduced from the measured velocity (V_{df}) and mass A by assuming two-body kinematics. The typical energy resolution was around 4 keV for 200-keV and 13 keV for 1.2-MeV γ-ray transitions. The total photo-peak efficiency of the two detectors was $\sim$0.8% at 1 MeV. Plotted in Figs. 2(d) and 2(e) are the Doppler-corrected γ-ray spectra obtained using the velocity vector of the detected ^{100}Zr and ^{138}Xe fragments in the spectrometer, respectively. The known γ-ray transitions of the ground-state bands of ^{100}Zr [14] and ^{138}Xe [24] can be clearly seen. Also labeled in Fig. 2(e) are broad transitions originating from ^{100}Zr. This is one of the complementary fragments of ^{138}Xe, originating from the fission of ^{242}Pu produced in an α-transfer reaction after evaporation of neutrons. The large width of these peaks originates from the incorrect Doppler correction used for Zr because the velocity and angle were taken for the detected ^{138}Xe, as mentioned earlier. The absence of γ-ray transitions from the Ru isotopes in Fig. 2(e) indicates that the most neutron rich Xe isotopes observed in this work are not populated by fusion-fission reactions, in contrast to the lighter Xe isotopes (see the following).

In Fig. 3(a), the mass of the detected fragment in the spectrometer is plotted as a function of the Doppler-corrected γ-ray energy for the Ru isotopes. The γ-ray spectra of the Ru isotopes can as well be obtained by detecting its complementary fragments (Xe) in the spectrometer and by obtaining the velocity vector V_{cf} for Ru from the measured quantities and the reaction kinematics. This is demonstrated in Fig. 3(b), where ^{133}Xe was identified in VAMOS, but the velocity vector used for Doppler correction was that of the undetected (in the spectrometer) complementary fragment. Transitions in $^{106-109}$Ru [25] and the incorrectly Doppler-corrected γ-ray transitions originating from ^{133}Xe are shown in the figure. It can be seen from the figure that ^{133}Xe is dominantly produced in coincidence with ^{108}Ru, leading to an average total (pre- and post-scission) neutron multiplicity of nine. A similar value was obtained with other fusion-fission fragment combinations (Ce-Zr, Ba-Mo, Te-Pd, and Sn-Cd). The Doppler-correction method illustrated allows a clear distinction between γ-ray transitions from nuclei detected in the spectrometer and its complementary fragment and opens avenues for studying heavier nuclei, whose direct identification in the spectrometer could be limited.

This technique allows for an unambiguous identification of each isotope over a wide range of elements produced in heavy-ion fusion-induced fission reactions. Thus, this improved sensitivity allows for the study of the prompt spectroscopy of states (having lifetimes $\leqslant$1 ns) in weakly populated and/or unknown neutron-rich nuclei. This will also additionally provide a unique access to study relatively high angular momentum states in stable nuclei that are very difficult to populate. ^{134}Xe presents an interesting case having

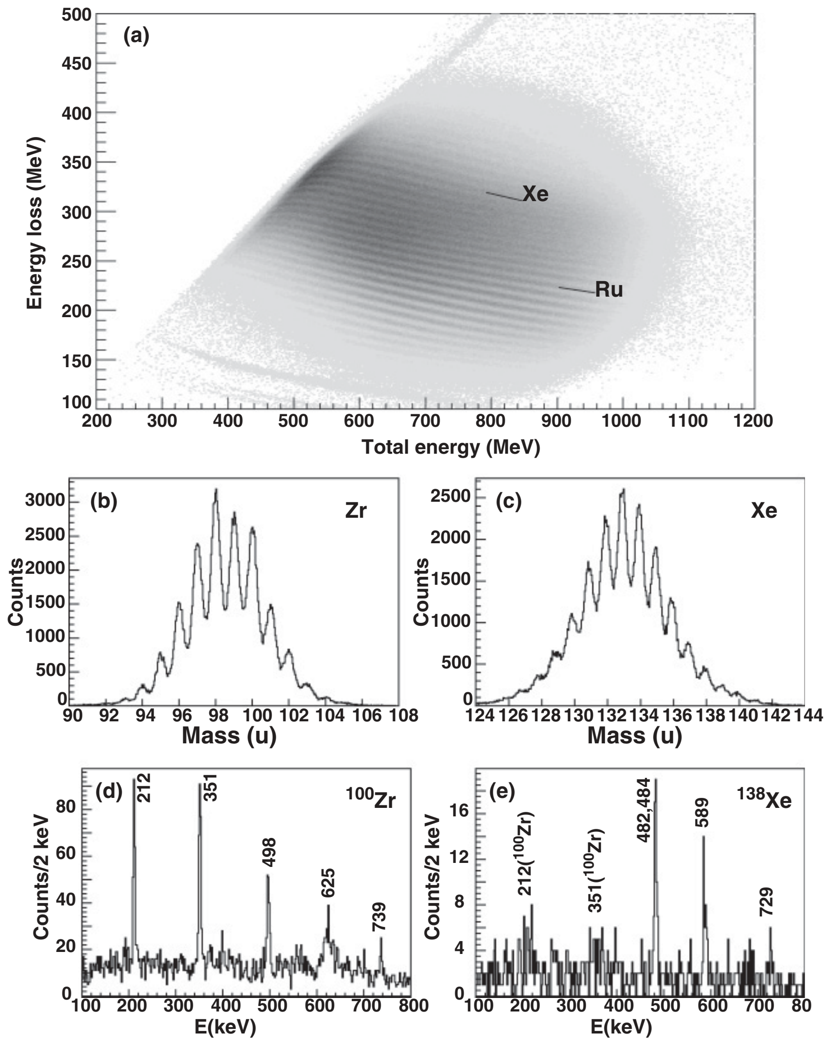

FIG. 2. Identification plots. (a) Two-dimensional spectrum of energy loss vs total energy. (b) Mass distribution obtained for the Zr isotopes. (c) The same as (b) for Xe isotopes. Doppler-corrected γ-ray spectrum obtained using the velocity of the detected fragment (V_{df}) for (d) ^{100}Zr and (e) ^{138}Xe. In panel (e), γ-ray transitions from ^{100}Zr can also be seen (see text).

four valence protons and a pair of neutron holes outside the doubly magic ^{132}Sn. Recently, detailed spectroscopy of the low-lying states in ^{134}Xe have been reported [26]. Two isomers, one at 10^+ with a lifetime of 5 μs [27] and another at 7^- with a lifetime of 290 ms, are known in ^{134}Xe [28]. The higher spin yrast states above these isomeric states are not known. This is also the case for the isotones ^{130}Sn and ^{128}Cd, where only the yrast states are measured below the 10^+ isomeric state [16,29]. We present the application of this technique to the prompt spectroscopy of ^{134}Xe above these isomers.

Figure 4 shows the Doppler-corrected γ-ray spectrum of ^{134}Xe. The γ-ray transitions originating from the complementary fragments of Ru isotopes can also be seen. Five γ-ray transitions [218(1), 320(1), 1323(2), 612(1), and 1101(2) keV] can be clearly identified in ^{134}Xe. The first three γ-ray transitions were reported in Ref. [30] as being in coincidence with each other and were tentatively assigned to ^{135}Xe. Our method of identification makes an unambiguous assignment of these transitions to ^{134}Xe. The two additional γ rays (323.1 and 541.8 keV) reported in Ref. [30] with relatively low intensities were not observed in this work. The intensities of all the observed γ rays are comparable in our work. Since the 612- and 1101-keV transitions were not reported in Ref. [30], they do not belong to the earlier established cascade. No previously known transitions [27] below the isomers in ^{134}Xe were observed in our study. Therefore, the observed γ-ray transitions necessarily feed the isomer(s). The 7^- and 10^+ isomers are expected to have the structure with dominant neutron configuration based on $(\nu h^{-1}_{11/2}d^{-1}_{3/2})$ and $(\nu h^{-2}_{11/2})$, respectively. Any transition between the aforementioned configurations would necessarily involve a large multipolarity and a change of parity leading to large lifetimes [27] and, hence, would not be observed in our work. States of higher spins built on these isomers will necessarily involve the rearrangement of the valence protons because the configuration of the neutrons is constrained. The first excited state in ^{136}Xe (2^+ state at 1313 keV [31]) can be understood as originating from a proton excitation because the configuration of neutrons corresponds to the $N = 82$ shell closure. Therefore, the first state above the 7^- or 10^+ isomer in ^{134}Xe is expected to have an excitation energy higher by a similar amount (1313 keV). Hence, the 612-keV transition was placed above the 1101-keV transition. The placement of the 1323-keV transition is also consistent with the previous argument. The lack of correlations between the γ rays above and below the long-lived isomers prevents us from attributing

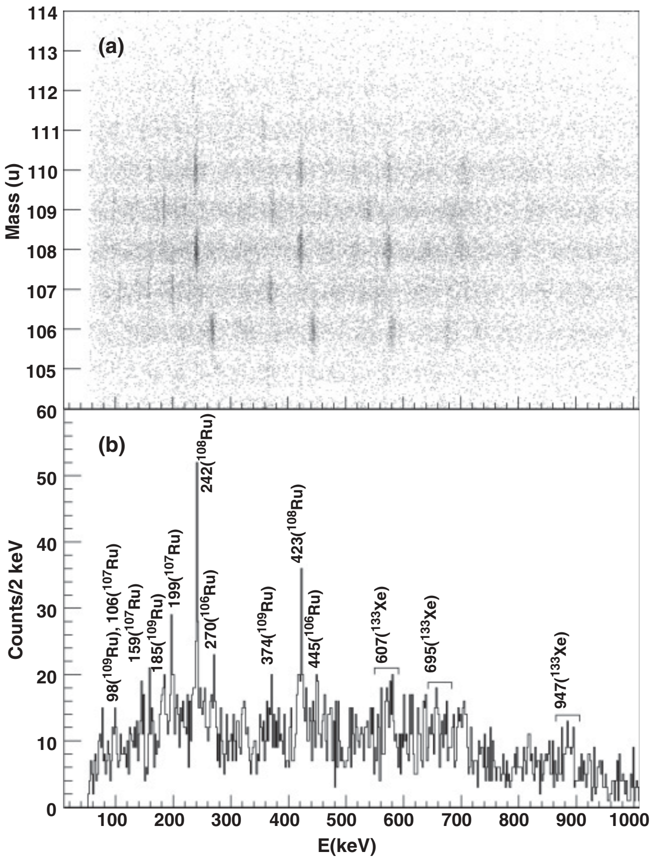

FIG. 3. γ-ray spectra of the Ru isotopes: (a) The mass of Ru as a function of Doppler-corrected γ-ray energy obtained using $V_{\rm df}$. (b) Doppler-corrected γ-ray spectra obtained using $V_{\rm cf}$ from the detected ^{133}Xe.

the currently observed cascades to a particular isomer. Hence, LSSM calculations will be used as a guide.

LSSM calculations have been carried out with the code NATHAN [32,33], using the recently proposed shell-model interaction GCN5082 [18] within a valence space, comprising of $g_{7/2}$, $d_{5/2}$, $s_{1/2}$, $d_{3/2}$, and $h_{11/2}$ orbitals for both neutrons and protons. The GCN5082 interaction was derived from a realistic G matrix based on the CD-Bonn potential and then corrected empirically to reproduce a set of 320 low-spin states in 87 nuclei with $Z \geqslant 50$ and $N \leqslant 82$ [18]. It has been applied to the study of double-β emitters in this region [34] and in the description of the so-called mixed-symmetry states in $N = 80$ isotones [35]. In this Rapid Communication, we only discuss the LSSM results for negative-parity states above the 7^- isomer and the positive-parity states above the 10^+ isomer in ^{134}Xe. A good agreement was obtained with the known levels below the isomers in ^{134}Xe [18,35]. Figure 5 shows the proposed experimentally derived level scheme that was obtained using the measured γ-γ coincidences of Ref. [30], the relative intensities of our work, and the shell-model calculations. The results of the LSSM calculations are also shown in Fig. 5.

The shell-model calculations confirm that these high-spin states above the isomers (7^- and 10^+) indeed have dominant contributions of $(\nu h_{11/2}^{-1} d_{3/2}^{-1})$ and $(\nu h_{11/2}^{-2})$ neutron

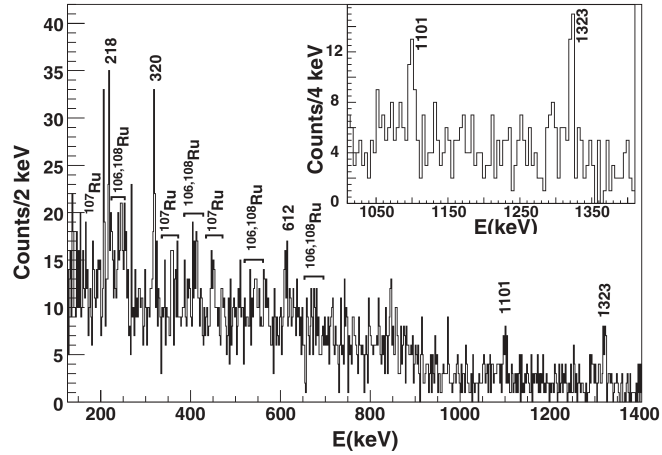

FIG. 4. Doppler-corrected γ-ray spectrum of ^{134}Xe. The background originating from the γ-ray transitions from the complementary Ru isotopes is also labeled. The inset shows an expanded region of the high-energy part of the spectrum in a higher binning.

configurations. The positive-parity states shown in Fig. 5 are built on the $(\nu h_{11/2}^{-2})$ configuration where the contribution of neutron holes coupled to the maximum spin ($10\hbar$) is more than 70%. The contribution of the protons for these states is dominated by the $(\pi d_{5/2}^{1} g_{7/2}^{3})$ configuration, which varies from 30% to 60% between the 12^+ and 14^+ states. Similarly, for the negative-parity states, the dominant $(\nu h_{11/2}^{-1} d_{3/2}^{-1}, 7^-)$ configuration contributes more than 85%, and for the protons,

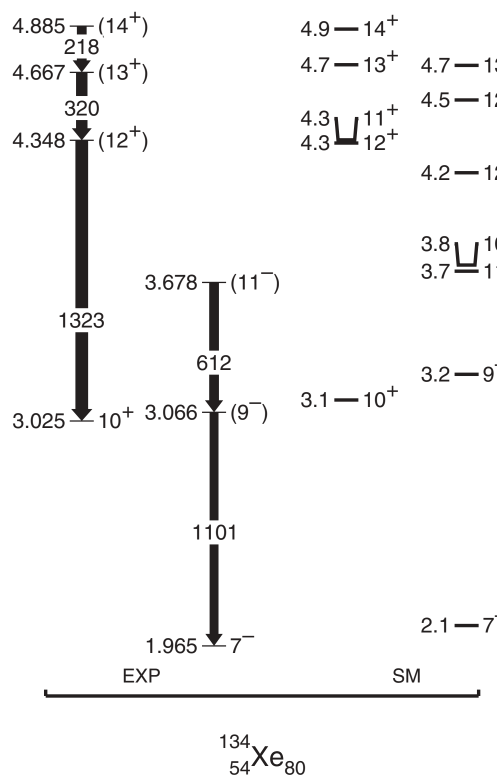

FIG. 5. Experimental and calculated level schemes of ^{134}Xe above the 7^- and the 10^+ isomers (see text).

the dominant contribution of the $(\pi g^4_{7/2})$ configuration is 30% and 46% for the 9^- and 11^- states, respectively.

In summary, this work demonstrates the power of using a large-acceptance spectrometer, coupled with γ-ray detectors, to perform the prompt spectroscopy of fission fragments with the direct unambiguous isotopic identification. This work has allowed us to study and improve the spectroscopy of ^{134}Xe above the high-spin isomers and to verify the predictions of a new shell-model interaction. The spectroscopy of certain neutron rich-isotopes populated in this experiment for various elements was previously unattainable using in-beam γ-ray spectroscopy methods. In perspective, the use of inverse kinematics coupled with an upgrade of the VAMOS detection system and the full EXOGAM array will allow for the spectroscopy of nuclei that are currently inaccessible.

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