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Synthesis of N = 127 isotones through (p,n) charge-exchange reactions induced by relativistic ²⁰⁸Pb projectiles

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The production cross sections of four N = 127 isotones (²⁰⁷Hg, ²⁰⁶Au, ²⁰⁵Pt, and ²⁰⁴Ir) have been measured using (p,n) charge-exchange reactions, induced in collisions of a ²⁰⁸Pb primary beam at 1 A GeV with a Be target. These data allow one to investigate the use of a reaction mechanism to extend the limits of the chart of nuclides toward the important r-process nuclei in the region of the third peak of elemental abundance distribution.

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The production of nuclei beyond the current limits of experimental synthesis is one of the foremost subjects for nuclear physicists, providing peerless opportunities to investigate not only the properties of exotic nuclei at extreme conditions of temperature, angular momentum, and isospin but also the nuclear reactions that occur in the astrophysical processes that lead to the formation of the heaviest elements in the universe [1]. These issues will be approached in the next generation of in-flight radioactive-beam facilities [2-4], where many of the remote exotic nuclei will be accessible for the first time.

In recent decades, fragmentation and fission reactions have proved to be useful means for the production of neutron-rich nuclei throughout the nuclear chart. Light neutron-rich nuclei have been synthesized using the fragmentation of 48 Ca [5], whereas the fission reactions have been satisfactorily used to produce medium-mass neutron-rich nuclei [6-8]. The fragmentation of secondary beams of neutron-rich fission products, such as ¹³²Sn, has also been proposed to produce extremely neutron-rich nuclei [9]. The challenge of obtaining access to the heavy neutron-rich nuclei has been tackled recently using the fragmentation of heavy stable projectiles such as 208 Pb or 238 U at relativistic energies [10,11]. It is particularly difficult to reach the neutron-rich region "south"

of lead, where only a few nuclei approaching the β stability line have been synthesized to date.

The exploration of the nuclear chart in the neutron-rich region around N = 126 will shed more light on the evolution of shell closures in heavy many-body systems. It has been shown that the robustness of the N = 20 and N = 28 shells weakens with the neutron excess [12,13]. There are also indications of the weakening of the N = 50 shell gap as the proton number decreases [14,15]. In the case of the N = 82 closed shell, the appearance of the shell-quenching phenomenon for neutrondrip-line nuclei is still being debated [16–18]. The exploration of the neutron-rich N = 126 isotones is also important for the modeling of the r-process in the region of the third peak of the elemental abundance distribution. Indeed, the properties of nuclei at the $A \approx 195$ waiting point determine the flow of matter to heavier fissioning *r*-process nuclei [19].

Hitherto, attempts to synthesize the challenging N = 126neutron-rich nuclei were centered on the fragmentation of ²⁰⁸Pb and ²³⁸U. The fragmentation mechanism is characterized by large fluctuations in the neutron-to-proton ratio and the excitation energy of the projectile residue. Extremely neutronrich nuclei can be produced through the cold fragmentation channel, in which the incident projectile mainly loses protons during the abrasion stage and the excitation energy of the prefragment is below the neutron separation energy.

We propose here an alternative reaction mechanism to explore the interesting neutron-rich region around N = 126, taking advantage of (p,n) charge-exchange reactions of relativistic ²⁰⁸Pb projectiles. Thus far, our experimental knowledge of charge exchange at relativistic energies has been focused mostly on (n,p) charge exchange of intermediate- [20] and heavy-mass nuclei [21,22].

The charge-exchange reactions involving heavy relativistic projectiles are described as peripheral nuclear collisions [23]

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between the projectile and target in a participant-spectator context [20]. Two-body nucleon-nucleon interactions induce the charge exchange between the projectile and target nuclei. There are two charge-exchange mechanisms [24] responsible for the modification of the nuclear charge. The first is a quasielastic isospin-exchange collision between a projectile nucleon n_p and a target nucleon n_t , where n_p receives the total kinetic energy of n_t ending up in the phase volume of the projectile prefragment, with the exchange of a virtual π^{\pm} or ρ^{\pm} meson; the second is an inelastic isospin-exchange collision where a nucleon from either the target or the projectile is excited into the $\Delta(1232)$ -resonance state by the exchange of a virtual pion between both nuclei. Subsequently, the Δ resonance decays, emitting a real pion. Only when the emitted pion is charged and escapes from the nucleus will the charge exchange be effective. It was shown that the contribution of these two mechanisms can be observed in the velocity distributions of the charge-exchange products [22].

In this work we provide a systematic comparison of the ²⁰⁸Pb single (p,n) charge-exchange cross sections of ²⁰⁷Hg, ²⁰⁶Au, ²⁰⁵Pt, and ²⁰⁴Ir with the simulated fragmentation yields of the same isotopes using ²³⁸U projectiles. The data reveal an approach to studying the primary structure of the neutron-rich N = 126 isotones from the β decay of the mother N = 127 nuclei, produced in (p,n) charge-exchange reactions of relativistic ²⁰⁸Pb projectiles.

The heavy neutron-rich nuclei were produced at GSI Darmstadt through (p,n) charge-exchange reactions between a 208 Pb projectile at 1 A GeV and a Be target 1.6 g/cm² thick. The SIS-18 synchrotron delivered the primary beam with a repetition cycle of 10 s and a spill length of 2 s, with intensities of up to 10^9 ions/spill. The reaction products were analyzed with the magnetic spectrometer fragment separator (FRS) [25], specifically tuned to transmit ²⁰⁶Au along the central trajectory. The FRS is a high-resolution forward spectrometer with a momentum-resolving power of 1500 for an emittance of 20π mm mrad, a momentum acceptance of 1.5%, and an angular acceptance around its central trajectory of 15 mrad. The spectrometer is divided into two symmetrical stages in order to preserve the achromatism of the system. In the first section, the residues are selected according to their mass-over-charge ratio A/Q, with a resolution given by the dispersion of their magnetic rigidity $B\rho$ and the acceptance of the spectrometer. A profiled Al degrader is placed at the intermediate image plane to separate the transmitted nuclei with an additional bend that depends on the ratio of the magnetic rigidities in the first and second sections of the separator [26].

The nuclei traversing the FRS were identified according to their mass-over-charge ratio A/Q and their atomic charge Q. The former was determined from the magnetic rigidity and the velocity of the fragments, whereas the latter was deduced from the energy loss registered in two multisampling ionization chambers (MUSIC) located at the final image plane of the spectrometer. The magnetic rigidity was obtained from the positions of the fragments at the intermediate and final image planes, measured using two plastic scintillators that also provided the time of arrival of the fragments at both focal planes and then their velocity.

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The relativistic energies employed in the current experiment favored that the equilibrium charge state of the fragmentation residues produced in the target was nearly fully stripped. However, because of the large atomic numbers of the selected nuclei, their probability of ionization in their interaction with the optical elements and detectors present in the FRS was enhanced. The charge-state equilibrium close to fully ionized nuclei was restored by using three stripping Niobium foils: the first behind the target (220 mg/cm²), the second behind the energy degrader at the intermediate image plane of the separator (105 mg/cm²), and the third in between the two MUSIC chambers (230 mg/cm²).

In spite of the high electronic affinity of the Niobium, a small percentage of the reaction residues remained in their hydrogen-like charge state in the first and second sections of the FRS, as well as in the MUSIC chambers. Their presence hence affected the identification of the nuclei of interest in A/Q, with the FRS, and in atomic number, with the MUSIC chambers. In particular, (A - 3, Z) residues with a hydrogenlike ionic charge state (Q = Z - 1) have a similar magnetic rigidity as fully stripped (A, Z) nuclei. Since the neutron-rich (A - 3, Z) fragments typically have production cross sections around two orders of magnitude larger than the cross sections of the isotopes with three more neutrons (A, Z), a few percent of the production of (A - 3, Z) fragments with a hydrogen-like ionic charge state could represent almost 100% contamination in (A, Z) fully stripped nuclei. Moreover, the ionic charge changes within the gas of the ionization chambers contributed to degrade the energy resolution achieved with these detectors.

In order to approach this challenge, the atomic charge-state changes of the nuclei traversing the FRS were determined by measuring the difference in magnetic rigidity in the intermediate degrader, quantified according to the variable $\Delta E_{deg} = (\gamma_1 - \gamma_2)uA/Q$, with γ being the relativistic factor in the first (1) and second (2) sections of the FRS, A being the mass number of the ion, and u being the atomic mass unit. Also, the ionic charge changes occurring in the stripping Niobium foil placed between the two MUSIC chambers were identified by comparing the energy losses registered by the two MUSICs. In this case, the fully stripped ions correspond to the maximum energy loss signal Q^{max} measured by any of the two chambers. A detailed description of the method can be found in Ref. [27].

Figure 1 shows the two-dimensional cluster plot of the variables ΔE_{deg} and Q^{max} for a FRS magnetic setting centered on ²⁰⁶Au. In Fig. 1, the four lines indicate events compatible with four ionic charges inside the FRS. The dotted line, labeled (1e-0e), corresponds to fragments leaving the target as hydrogen-like ions and losing the electron at the intermediate degrader. The dashed line labeled (0e-1e) represents nuclei leaving the target fully stripped but gaining one electron in the energy degrader at the intermediate image plane. The solid line labeled (0e-0e) corresponds to fully stripped nuclei in both sections of the FRS. Finally, the dashed-dotted line labeled (1e-1e) indicates nuclei leaving the target with one electron and keeping this electron all along the FRS.

The excellent magnetic resolution of the spectrometer allows the separation of the different charge states, even between the (0e-0e) and (1e-1e) contributions to a large extent,



FIG. 1. (Color online) Correlation plot of the effective energy loss in the intermediate degrader and the atomic charge for a FRS magnetic setting centered on ²⁰⁶Au. Each labeled set of clusters corresponds to a different charge-state configuration.

as Fig. 1 illustrates. The only misidentifications may be given by two contributions: first, the (A - 3, Z) nuclei that acquire one electron in the target, conserve it along the FRS, and lose it at the exit of the FRS or in the stripper foil between both MUSIC chambers and, second, the (A, Z + 1) nuclei that gain one electron at the target and keep it in their flight through the FRS and the two MUSIC chambers. Fortunately, the probability for these combinations is rather low, producing a moderate contamination, as explained below and shown in Table I.

The mass identification matrix of the A/Q ratio as a function of the atomic charge Q^{max} for fully stripped nuclei is shown in Fig. 2. The N = 127 isotones produced through (p,n) charge-exchange reactions are labeled in Fig. 2. The high mass resolution obtained after applying the identification procedure allows one to determine the production cross sections of the transmitted nuclei with high accuracy. The small amount of contamination still present in the (0e-0e) distribution can be inferred from the clusters of dots appearing on the right side of 207 Hg and 206 Au in Fig. 2. Their A/Q and Q^{max} values correspond to those of 208 Hg and 207 Au, but their yields can only be understood as being due to the hydrogen-like contaminants traversing the FRS (which are 205 Hg for 208 Hg and 204 Au and 207 Hg for 208 Au).

The expected rates of each contaminant nucleus are calculated as the product between its fragmentation yields and the survival probability of the corresponding charge-state configuration along the beam line. In this work, the code

TABLE I. Single (p,n) charge-exchange cross sections and charge-state contaminants (percentages are given in parentheses).

Nucleus	σ (mb)	Contaminants
²⁰⁷ Hg	$0.11(5)10^{-1}$	²⁰⁷ Tl (3%), ²⁰⁴ Hg (1%)
²⁰⁶ Au	$0.36(12)10^{-3}$	²⁰⁶ Hg (3.5%), ²⁰³ Au (2%)
²⁰⁵ Pt	$0.57(18)10^{-5}$	²⁰⁵ Au (12%), ²⁰² Pt (5%)
²⁰⁴ Ir	$0.19(16)10^{-6}$	²⁰⁴ Pt (13%), ²⁰¹ Ir (26%)



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FIG. 2. (Color online) Two-dimensional cluster plot of the atomic charge as a function of the A/Q ratio.

GLOBAL [28] has been used to calculate the production probabilities of the charge-state contaminants after the target, the intermediate and final image planes, and the two MUSIC chambers. Finally, the percentage of contamination introduced by these hydrogen-like nuclei in the cross sections of the N =127 isotones is determined as the ratio between the expected production of contaminants and the yields of the N = 127 nuclei. Table I summarizes the charge-state contaminants for each N = 127 isotone, followed by the percentage of contamination in the measured cross sections in parentheses. For proton numbers over 78, the production rates of N = 127 isotones exceed by far the yields of hydrogen-like contaminants, thus confirming unambiguously their production through (p,n)charge-exchange reactions.

The production rates of the nuclear species investigated were obtained by measuring the number of ions transmitted to the final image plane of the FRS. The production cross sections were determined after correcting these yields for losses due to beam attenuation and secondary reactions in the different materials placed throughout the FRS for losses due to the rejection of the charge states in the data analysis and for losses due to an incomplete transmission of the ions in the image planes. The measured yields were normalized to the fragmentation cross section of ²⁰²Ir, measured in a previous experiment under analogous conditions [10], in order to account for the total number of incident projectiles and the dead time of the data acquisition system. The corrections for beam attenuation, secondary nuclear reactions, and charge states were calculated for each nuclear species using the codes KAROL [29] and GLOBAL [28], whereas the transmission correction was applied to the residues partially transmitted in one or both focal planes. The uncertainties in beam attenuation and secondary reactions amount to 10% [29], and the uncertainties associated with the correction for incompletely stripped ions and transmission amount to 5% and 20%, respectively [27].

Table I summarizes the charge-exchange cross sections of the N = 127 isotones. The uncertainties associated with these measurements include the statistical and the systematic effects. The systematic uncertainties embody the errors related to the yield corrections and the reference cross section.



FIG. 3. Production cross sections of charge-exchange N = 127 isotones (dots, this work) and N = 126 fragmentation residues (triangles, from Ref. [10]) as a function of the number of protons removed from the projectile ²⁰⁸Pb. The lines represent fragmentation cross sections of N = 127 (solid line) and N = 126 (dashed line) isotones obtained with the COFRA [30] code for reactions induced by ²³⁸U and ²⁰⁸Pb projectiles, respectively.

Figure 3 shows the evolution of the measured chargeexchange cross sections for the four N = 127 isotones (dots) as a function of the number of protons removed from the primary beam, ²⁰⁸Pb. Figure 3 also shows the cross sections of the analogous N = 126 isotones (triangles) synthesized by cold fragmentation of ²⁰⁸Pb [10]. The cross sections of the N = 127nuclei produced by fragmentation of ²³⁸U have been simulated with the code COFRA [30] (solid line) for comparison. We have also simulated the fragmentation cross sections of the N = 126 isotones from ²⁰⁸Pb beams (dashed line). In this last case, the perfect agreement with the measured data illustrates the predictive power of this code.

Looking in detail, one can observe that the production yields of the N = 127 isotones show the same slope as the cross sections of the N = 126 isotones. This indicates that the proton removal channels dominate the trend followed by the measured charge-exchange cross sections. Indeed, the constant ratio between the measured charge-exchange yields and the fragmentation yields of the analogous proton removal channel is expected since the charge-exchange mechanism takes place in peripheral nuclear collisions between projectile and target and, in the case of the inelastic channel, the isospin exchange can be only detected if the pion escapes from the nuclear surface, which is less likely to occur than the abrasion of a proton.

We also learn from Fig. 3 that the (p,n) charge exchange of lead beams is the optimal mechanism to produce the N = 127 isotones with a proton number larger than or equal to 78, whereas for lower Z the fragmentation of ²³⁸U (solid line) is more adequate.

In order to get a better understanding of the (p,n) chargeexchange mechanism, the velocity distributions for ²⁰⁵Au and ²⁰⁶Au are shown in Fig. 4. These two reaction products were produced in the collision ²⁰⁸Pb + ⁹Be at 1 A GeV. While the residue ²⁰⁵Au was most commonly produced by the removal of three protons, the nucleus ²⁰⁶Au was unequivocally synthesized through a (p,n) charge-exchange



FIG. 4. (Color online) Longitudinal velocity distributions of 205 Au (dashed line) and 206 Au (solid line) fragments and of the beam nuclei 208 Pb (dotted line) in the reference frame of the latter.

interaction plus the removal of two protons. Figure 4 also shows the velocity of the lead projectile to illustrate the contribution of the momentum dispersion and the energy straggling of the fragment separator to the measured velocities. These observables were obtained from the magnetic rigidity $B\rho$ of the nuclei in the intermediate image plane and were further transformed into the reference frame of the projectile nucleus in the center of the target by Lorentz transformation. The transformed velocities were corrected by the energy loss of the projectile in the first half of the target and the energy loss of the residue in the second half of the target and the first stripping Nb foil. The momentum resolution is then given by the resolution in $B\rho$, $\Delta B\rho/B\rho \approx 5 \times 10^{-4}$, which improves the time-of-flight resolution by one order of magnitude.

The longitudinal velocities of the fragmentation residues decrease as the number of protons removed from the projectile increases [30]. Looking at Fig. 4, the velocity distribution of the three-proton removal product ²⁰⁵Au is closer to the projectile velocity than the distribution of the two-proton removal and one (*p*,*n*) charge-exchange residue ²⁰⁶Au. The lower velocities in the charge-exchange nucleus ²⁰⁶Au can be explained as being due to the excitation of the Δ resonance in the inelastic charge-exchange channel. In this case, the velocity of the reaction products is reduced because of the mean energy transferred to the pion, by approximately 300 MeV.

Under the present experimental conditions, the energy shift induced by the excitation of the Δ -resonance state and the subsequent emission of the pion corresponds to an average velocity shift of approximately 0.03 cm/ns. Although the thickness of the target used in this work prevents an accurate unfolding of the measured distributions as performed in Ref. [22], the distribution of ²⁰⁶Au shows a clear structure at smaller velocities in the range compatible with the shift induced by the inelastic charge-exchange channel. The appearance of this slower component in the velocity distribution of ²⁰⁶Au can then be considered as a clear sign pointing

to charge exchange as the production mechanism for the N = 127 nuclei.

Four N = 127 isotones have been synthesized unequivocally in (p,n) charge-exchange collisions in the reaction 208 Pb + 9 Be at 1 A GeV. The lower velocities of those nuclei compared to others produced in pure nucleon removal collisions indicate a considerable role of the inelastic charge-exchange channel through the excitation of the Δ resonance. Given a systematic comparison, the measured charge-exchange cross sections of 208 Pb exceed the simulated fragmentation cross sections of 238 U for the $A = 205 \rightarrow 207$, N = 127 isotones. For lower-mass isotones, the fragmentation of 238 U results in a more appropriate production mechanism.

Summarizing, this work represents a step further in the attempts to expand the chart of nuclides toward the neutron drip

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line in the region around N = 126. Indeed, charge exchange has proved to be an optimum reaction mechanism to produce neutron-rich nuclei "south" of lead, of particular importance not only for investigating the evolution of shell closures in heavy nuclear systems but also for understanding the *r*-process stellar nucleosynthesis.

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