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New Short-Lived Isotope $^{221}$U and the Mass Surface Near $N = 126$


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Two short-lived isotopes $^{222}$U and $^{222}$U were produced as evaporation residues in the fusion reaction $^{50}$Ti + $^{176}$Yb at the gas-filled recoil separator TASCA. An $\alpha$ decay with an energy of $E_\alpha = 9.31(5)$ MeV and half-life $T_{1/2} = 4.7(7) \mu$s was attributed to $^{222}$U. The new isotope $^{221}$U was identified in $\alpha$-decay chains starting with $E_\alpha = 9.71(5)$ MeV and $T_{1/2} = 0.66(14) \mu$s leading to known daughters. Synthesis and detection of these unstable heavy nuclei and their descendants were achieved thanks to a fast data readout system. The evolution of the $N = 126$ shell closure and its influence on the stability of uranium isotopes are discussed within the framework of $\alpha$-decay reduced width.

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The evidence for the presence of the $N = 126$ shell closure above $^{208}$Pb is based on systematic analyses of a limited amount of experimental data available to date like $E_\alpha$ and thus the deduced $Q_\alpha$ values, and half-life, $T_{1/2}$. More detailed information on nuclear structure can be obtained from the $\alpha$-particle preformation probability inside the nucleus [9], which microscopically quantifies the stability against $\alpha$ decay. Different variables that are equivalent to the $\alpha$-particle preformation probability in which both macroscopic observables $Q_\alpha$ and $T_{1/2}$ are simultaneously involved, can be deduced within different quantum-mechanical approaches [10,11]. Commonly, a reduced width for $\alpha$ decay, $\delta^2$ [10], which takes into account the angular momentum of the emitted $\alpha$ particle, is used.

At and below $N = 126$, reduced widths of isotones of Po ($Z = 84$) to Pa are significantly lower than those of the $N = 128–130$ isotones located beyond the closed $N = 126$ shell [10–12]. This reflects the robustness of the $N = 126$ shell closure up to Pa and the effect of the semi-magic core with $N = 126$. The evolution of this closure towards higher extremes in the proton-to-neutron ratio is less well studied, but the fading of classical shell closures has been observed in lighter regions of the chart of nuclei [2]. The $N = 126$ shell closure hosts the heaviest known stable doubly magic spherical nucleus, $^{208}$Pb, and is experimentally established up to protactinium ($Z = 91$), above which its evolution is yet poorly examined.
Signals from all the detectors were processed in a combined analog and digital (CANDI) data acquisition system comprising analog and digital branches. The shapes of the preamplified signals from the 48 horizontal strips \(n^+\) side were recorded in 50-\(\mu\)s-long traces by analog-to-digital converters with 60 MHz sampling [27]. The trace length was selected to be longer than the dead time of the analog branch of \(\approx 35\ \mu\)s. Energy resolutions (FWHM) of individual vertical strips of the stop detector coupled to the analog branch were about 30 keV for 8.7-MeV \(\alpha\) particles. The amplitudes, i.e., energies of signals stored in traces were extracted by using different types of software algorithms depending on the multiplicity of the recorded signals [28]. The best resolutions, about 40 keV, were achieved for 8.7-MeV \(\alpha\) particles that were registered as single events in the traces. The best energy resolutions of multiple \(\alpha\) events stored in a single trace with time differences down to 1 \(\mu\)s and 0.17 \(\mu\)s were \(\approx 110\) keV and \(\approx 180\) keV, respectively.

Half-lives known for \(^{222}\)U and its daughter \(^{218}\)Th and predicted for \(^{221}\)U are much shorter [13] than the 50-\(\mu\)s trace-length of CANDI. Thus, their radioactive decay signature is registered in digital traces of implantation signals (ER-like). The analysis procedure thus consists of two main steps.

(i) A correlation analysis of the analog data, searching for \(\alpha\)-\(\alpha\) chains requiring that both the ER-like and the \(\alpha\)-like events (7–18 MeV) occurred in the same pixel of the stop detector within 20 s. The results of the correlation analysis are shown in Fig. 1. The implantation of \(^{214}\)Ra, \(^{215}\)Ra, \(^{217}\)Th, and \(^{214}\)Fr was identified by their known

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\text{FIG. 1. Results from a correlation analysis of the type ER-} \alpha \text{ within } 20 \text{s: (a) Spectrum of correlated } \alpha \text{ particles registered at a bombarding energy of } 239 \text{ MeV and (b)–(d) total decay curves of the implanted nuclei. (e) Decay curve of } ^{214}\text{Fr identified in the correlation analysis of type ER-} \alpha \text{ (7–18 MeV)–} \alpha \text{ (}^{214}\text{Fr).}
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FIG. 2 (color online). Identification spectra for the separation of pileup events originating from the decay chains ER-α(7 – 18 MeV)-α(214Fr). (a) Energy spectrum from the analog part of the data. (b), (c) Energy spectra of events deduced from the digital traces. Examples of some pileup traces: (d) two α particles with full energies, (e) and (f) for traces where the second and first α particle, respectively, escaped in the backward direction and deposited only partial energy in the stop detector.

α-decay properties [13], and the corresponding ERs were assigned for each event.

(ii) Traces of all ERs correlated to a subsequent α decay of 214Ra and 217Th, which are members of the α-decay chains originating from 223U and 221U, respectively, were checked event by event for the presence of multiple signals.

In the cases of 217Th and 214Fr, an additional ER-α-α correlation analysis was performed to reconstruct the full pattern of the α-decay chain. As an example, 214Fr was found to be the last member of the ER-α(7-18 MeV)-α(214Fr) chain with $T_{1/2} = 5.9(4)$ ms (see Fig. 1). An energy spectrum (analog branch) of the second member with $T_{1/2} = 3.5(1)$ ms is shown in Fig. 2(a). To resolve apparent pileup events, the digital traces were analyzed in the second step. Different examples of such events are presented in Figs. 2(d)–2(f). The energy spectra of the two signals allowed the separate peaks to be clearly resolved, which correspond to known α-decay energies of 222Pa and 218Ac [13]. Time differences between those signals yield $T_{1/2} = 0.96(5)$ μs, which agrees well with the literature data of 218Ac [13]. Thus, the origin of the α decay of 214Fr was attributed to the implantation of 222Pa produced as an ER via the $3\pi n$ deexcitation channel from the compound nucleus 226U*. The same digital-trace analysis was used for the extraction of α-decay properties of all other short-lived nuclei.

Significant amounts of ER traces correlated to α(215Ra) were stored with double signals. A resolved energy spectrum of the second signal and the decay curve extracted from the time difference of these signals (shown in Fig. 3) is in good agreement with the literature data for 219Th [13]. No triple signals, which would correspond to the implantation of 223U ($T_{1/2} = 55(10)$ μs [13]) from the 3n channel, were detected.

Traces with two and three signals were detected for ERs followed by α(214Ra). Traces with two signals and with very short time differences leading to $T_{1/2} = 0.16(4)$ μs (see Fig. 3) between the signals were assigned to the implantation and α decay of 218Th ($T_{1/2} = 0.117(9)$ μs [13]) produced in the α4n channel.

Eighty-one ER traces containing three signals were unambiguously attributed to the implantation of 221U followed by α decays of 222U and 218Th. One such trace is shown in Fig. 4. A half-life of 4.7(7) μs and an energy of 9.31(5) MeV were deduced for the α decay of 222U (see Fig. 3).

Twenty-six ER traces followed by subsequent α decays of 217Th and 213Ra were stored with double signals and are attributed to the implantation and α decay of the hitherto unknown 221U (see Fig. 4). A half-life of 0.66(14) μs and an α-particle energy of 9.71(5) MeV were deduced (see Fig. 3) for this isotope.

The most favored α transitions in nuclei with $N = 129$ proceed through the same $\nu 2g_{9/2}$ orbital in the mother and

FIG. 3. Energy spectra and decay curves of α particles emitted from 219Th, 218Th, 222U, and 221U. The data were extracted from the digital data branch. In the case of 218Th both double and triple-signaled traces from ER-α(214Ra) were used. All traces with time differences down to 100 ns between the preceding signal and α decay were used.

FIG. 4 (color online). Examples of traces where 222U and 221U were registered. Right panel: Enlarged figures of rapidly correlated signals.

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daughter nuclei. Thus, the observed $\alpha$ decay in $^{221}$U is attributed to such a transition. The $(9/2^+)$ state is tentatively assigned to the ground state of $^{221}$U based on systematics [13,14]. Alpha decays of both $^{221}$U and $^{222}$U are in good agreement with $Q_\alpha$ [29] and $T_{1/2}$ systematics [14].

The new $\alpha$-decay data on $^{221}$U and $^{222}$U together with literature values for Po-Th ([13,15]) allow us to construct the tendencies of $\delta^2$ in the $N = 129, 130$ isotones up to U, where the $\pi(1h_{11/2})$ orbital is fully filled. The deduced $\delta^2$ values for the $N = 124, 126-130$ isotones are shown in Fig. 5. In the case of the $N = 129$ isotones, only $\alpha$ transitions populating the same single-neutron states in the daughter nuclei were taken. For the cases at $N = 127$, the $\alpha$ transition proceeds between different initial $\nu(2g_{9/2})^1$ and final $\nu(3p_{1/2})^1$ configurations. Thus we use $\Delta \epsilon = 0$ for $N = 129$ and $\Delta \epsilon = 5$ for $N = 127$. Up to Th ($Z = 90$), the $\delta^2$ values for the $N = 124, 128-130$ isotones are larger than the $N = 126, 127$ ones, which clearly shows a strong effect of the $N = 126$ shell closure. At the same time, the $\delta^2$ values for the $N = 126, 127$ isotones, which are known up to U, smoothly rise with increasing $Z$, indicating a weakening of the semi-magic core with $N = 126$. For the $N = 129, 130$ isotones, where our new data allow extending the systematics up to U, the $\delta^2$ values decrease in contrast to the trend seen in the $N = 126, 127$ isotones. The sudden increase of $\delta^2$ values when crossing the $N = 126$ shell closure seen in lighter elements with even $A$ is no longer present at $Z = 92$. In odd-$A$ nuclei, the gap is still persisting at $Z = 92$, in contrast to the even-$A$ case. Essentially, $\alpha$ decay of the even-even nuclei, where all nucleons are paired, provides better information on their structure compared to odd-$A$ ones, where $\alpha$ decay is strongly influenced by their unpaired nucleon.

The above observed feature for even-$A$ nuclei indicates a weakening of the $N = 126$ shell stabilization effect for U, which can be inferred even in the absence of data on $^{221}$U ($N = 128$). In addition, $\delta^2$ values for the $N = 124$ isotones show again a notable discrepancy in U. The smallest $\delta^2$ value is found in $^{216}$U ($N = 124$), which can be argued as having a more stabilized core than the semi-magic $^{218}$U ($N = 126$).

A weakening of the $N = 126$ shell closure can also be seen in the reduction of the neutron-shell gap in elements above Pb ($Z = 82$). The neutron-shell gap, $G_n$, between the last-occupied and first-valence orbitals around $N = 126$ can be calculated as

$$G_n(Z, 126) = 2B(Z, 126) - B(Z, 125) - B(Z, 127)$$

using the known (except $^{217}$U) binding energies, $B$ [2,29]. These gaps are shown in Fig. 5 (bottom panel) where we compare experimental data ([29]) with those obtained using two different theoretical models (FRDM95 [4], HFB26 [30]). A reduction of $G_n$ as a function of $Z$ for heavier elements is observed in the experimental data, supporting the above discussed weakening of the shell closure inferred on the basis of the trends of $\delta^2$ values. $G_n$ values are significantly reduced when crossing $Z = 82$, which can be attributed to the loss of the magic $Z = 82$ partner, which leads to less stable semi-magic nuclei with $^{218}$U being the most unstable $N = 126$ isotope known to date.

Two theoretical models, FRDM95 and HFB26, were selected as representatives of the macroscopic-microscopic and microscopic approaches, respectively. The macroscopic-microscopic approach is based on corrections of calculated liquid-droplet masses to the shell structure, while the microscopic ones take into account the interactions between the nucleons with differently parametrized effective forces for the solution of the many-body quantum equations within particular approximations such as Hartree-Fock-Bogoliubov with Skyrme force (HFB26). The Finite-Range Droplet Model (FRDM95) predicts well the $G_n$ values around the doubly magic $^{208}$Pb, but fails to describe the decreasing trend for heavier elements. On the other hand, the HFB26 model does not perform as well around $^{208}$Pb, but predicts the reduction of $G_n$ towards heavier elements better. This shows the importance of interactions between the valence nucleons for the evolution of the shell closure.

The anomalous behavior observed in U thus may hint at significant changes in its structure due to nucleon-nucleon interactions, for instance the presence of non-negligible deformation as discussed theoretically in Ref. [31].

FIG. 5 (color online). Reduced $\alpha$-decay widths ($\delta^2$, [10]) of even-$A$ (a) and odd-$A$ (b) isotopes, and (c) neutron-shell gaps ($G_n$, [2]) of Po-U as a function of the $Z$. Errors originating solely from half-lives are given for U. (c) Experimental data (full dots) [29] are compared to FRDM95 (open diamonds) [4] and HFB26 (open triangles) [30] calculations. See text for details.
of 9.31(5) MeV and 9.71(5) MeV were measured for 221U. Half-lives of 4.7(7) μs and 0.66(14) μs and α-particle energies of 9.31(5) MeV and 9.71(5) MeV were measured for 222U and 221U, respectively. These were produced in the fusion-evaporation reaction 50Ti+176Yb with maximum cross sections of a few nanobarns at 47 and 54 MeV excitation energies of 226U [28]. The comparative analysis of the reduced widths and neutron-shell gaps of the Po-U isotopes shows a significant weakening of the influence of the N = 126 shell closure in U. Our findings motivate further investigations of the N = 126 shell closure by synthesizing hitherto unknown nuclei, and by detailed studies of U. The experimental technique applied in the present work allows the identification of short-lived activities in a wide range of the nuclear chart and particularly for the still unobserved 220U for which, though, the predicted half-life is only several tens of ns [14] and hence too short for surviving the flight time through TASCA. A preferable way to synthesize this isotope would be via its significantly longer lived [32] α-decay precursor 224Pu.

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