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## Signatures of the $Z = 82$ Shell Closure in $\alpha$ -Decay Process

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In recent experiments at the velocity filter Separator for Heavy Ion reaction Products (SHIP) (GSI, Darmstadt), an extended and improved set of  $\alpha$ -decay data for more than 20 of the most neutron-deficient isotopes in the region from lead to thorium was obtained. The combined analysis of this newly available  $\alpha$ -decay data, of which the  $^{186}\text{Po}$  decay is reported here, allowed us for the first time to clearly show that crossing the  $Z = 82$  shell to higher proton numbers strongly accelerates the  $\alpha$  decay. From the experimental data, the  $\alpha$ -particle formation probabilities are deduced following the Universal Decay Law approach. The formation probabilities are discussed in the framework of the pairing force acting among the protons and the neutrons forming the  $\alpha$  particle. A striking resemblance between the phenomenological pairing gap deduced from experimental binding energies and the formation probabilities is noted. These findings support the conjecture that both the  $N = 126$  and  $Z = 82$  shell closures strongly influence the  $\alpha$ -formation probability.

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In nuclear science, the constant attention to  $\alpha$  decay (and its time-reversal process of fusion reactions with  $\alpha$  particles) is due to the relative simplicity of its experimental investigation and the wealth of spectroscopic information it provides [1]. On the other hand, the  $\alpha$ -decay process is important for understanding such crucial problems in modern nuclear and astrophysics as, e.g., cluster decay (emission of nuclei heavier than  $\alpha$  particle) [2], stellar nucleosynthesis and  $\alpha$ -cluster states [3,4], and the synthesis and decay of super-heavy elements [4,5].

The gross features of  $\Delta L = 0$  (no angular momentum change)  $\alpha$  transitions (e.g., between the  $I^\pi = 0^+$  ground states of even-even nuclei) are expressed by the Geiger-Nuttall rule [6], postulated in 1911, which linearly relates the logarithm of the partial half-life  $T_{1/2}$  with the inverse square root of the  $\alpha$ -decay  $Q$  value. The Geiger-Nuttall rule was understood in 1928 by Gamow [7] and independently by Condon and Gurney [8] as due to quantum-mechanical “tunneling” of a “pre-formed”  $\alpha$  particle through a (classically impenetrable) spherical barrier. Since then, practically all theoretical and semiempirical methods treat  $\alpha$  decay as a two-step process, which involves the preformation of an  $\alpha$  particle, followed by its penetration through the barrier. Rasmussen presented in 1959 a method to extract from the measured  $\alpha$ -decay

half-life the so-called reduced  $\alpha$ -decay width, which is related to the  $\alpha$ -particle formation probability [9]. It can be used to extract nuclear structure information not only for  $\Delta L = 0$  ground-state to ground-state decay of even-even nuclei but also for the  $\alpha$  decay of odd- $A$  and odd-odd nuclei and for fine-structure  $\alpha$  decay to excited states [9]. In the analysis of the reduced widths of even-even nuclei around  $^{208}\text{Pb}$ , a strong discontinuity is observed when crossing the magic neutron number at  $N = 126$  [9]. This is also seen in the Geiger-Nuttall plots, where it is revealed through the need of different linear relations for nuclei with  $N < 126$  and  $N > 126$  [10]. Surprisingly, such a strong effect was not observed when crossing the  $Z = 82$  shell [10], leading to speculations that  $Z = 82$  is no longer a good magic number at the very neutron deficient side [11]. However, fine-structure  $\alpha$ -decay studies hinted at a  $Z = 82$  shell closure [12], and the systematics of the  $Q_\alpha$  values do not show any sign of a reduction of the  $Z = 82$  shell closure. Moreover, recent mean-square charge radii measurements show that the ground states of the even-even lead isotopes remain spherical down to  $N = 100$ , which is a fingerprint for a good shell closure [13]. This region of the nuclear chart is notorious for shape coexistence as evidenced by low-lying  $0^+$  states as band heads of rotational bands [14,15]. Furthermore, in a number of

neutron-deficient polonium isotopes, the reduced width of the  $\alpha$  decay to excited  $0^+$  states equals or is even larger than the ones corresponding to the decay to the ground state [16].

Recently, we performed a series of experiments at the Separator for Heavy Ion reaction Products (SHIP) in GSI (Darmstadt, Germany) aimed at detailed  $\alpha$ -decay studies of the most neutron-deficient isotopes in the lead to thorium region. Several new isotopes have been produced:  $^{179}\text{Pb}$  [17],  $^{184}\text{Bi}$  [18],  $^{187-189}\text{Po}$  [19,20],  $^{192}\text{At}$  [21],  $^{193,194}\text{Rn}$  [22],  $^{197,198}\text{Fr}$  [23],  $^{208}\text{Th}$  [24], and the decay properties of more than 20 neighboring isotopes were improved and discussed in these Letters.

In this Letter, we show that by combining these new findings with the existing data, we now obtain clear evidence that, contrary to some of the previous interpretations, crossing the  $Z = 82$  shell to higher proton number strongly accelerates the  $\alpha$  decay.

While the other new isotopes were presented in Refs. [17–22,24], we report here on the  $\alpha$  decay of the new isotope  $^{186}\text{Po}$ . A detailed description of the experimental setup is given in Ref. [19]. The  $^{186}\text{Po}$  nuclei were produced after evaporation of four neutrons from the excited compound nuclei  $^{190}\text{Po}^*$  formed in the complete-fusion reaction of 230-MeV  $^{46}\text{Ti}$  ions with  $^{144}\text{Sm}$  target nuclei. The  $^{46}\text{Ti}$  beam, with a typical intensity of  $\sim 200$  pA, was provided by the UNILAC heavy ion accelerator of the GSI. Eight  $^{144}\text{Sm}$  targets, each of 96.4% isotopic enrichment and  $450 \mu\text{g}/\text{cm}^2$  thickness, were mounted on a wheel, rotating synchronously with the UNILAC macro-pulsing. After separation by SHIP, the nuclei were implanted into a  $300 \mu\text{m}$  thick,  $35 \times 80 \text{ mm}^2$  16-strip position-sensitive silicon detector, where their subsequent particle decays were measured. The  $^{186}\text{Po}$  production cross section is only  $\sim 200(70)$  pb, which corresponds to the production of a few atoms of  $^{186}\text{Po}$  per day. Within 3 days of the measurements, eight time-position correlated decay events of  $^{186}\text{Po}$  were observed, one of which is shown in Fig. 1. It starts with the implantation of the nucleus  $^{186}\text{Po}$  in the position-sensitive silicon detector,

followed by the subsequent emission of four  $\alpha$  particles ( $\alpha_1$ – $\alpha_4$ ) at the same position (within the position resolution of the detector, which is  $\sim 0.3$  mm). The measured decay energies and half-lives for the  $\alpha_2$ – $\alpha_4$  decays match well to the known daughter products of  $^{186}\text{Po}$ , namely, the isotopes  $^{182}\text{Pb}$ ,  $^{178}\text{Hg}$ , and  $^{174}\text{Pt}$ , which proves that the parent  $\alpha_1$  decay originates from this new isotope. On the basis of all eight correlation chains, an  $\alpha$ -decay energy of  $8320(15)$  keV and a half-life of  $28_{-6}^{+16} \mu\text{s}$  were deduced for  $^{186}\text{Po}$ . This is probably the lightest isotope in the Po chain that can be produced with presently available technology, as the isotope  $^{185}\text{Po}$  is expected to have sub- $\mu\text{s}$  half-life and production cross section of the order of a few pb only.

The new results in the lead to thorium region can now be analyzed with the recently developed Universal Decay Law, which describes in a consistent way the half-lives of all forms of cluster radioactivity [25]. In the Universal Decay Law, the half-life corresponding to the emission of a cluster  $c$  (in particular an  $\alpha$  particle) from a mother nucleus is evaluated starting with the microscopic expression provided by residues of the  $R$  matrix [26], i.e.,

$$T_{1/2} = \frac{\hbar \ln 2}{\Gamma_c} \approx \frac{\ln 2}{\nu} \left| \frac{H_l^+(\chi, \rho)}{RF_c(R)} \right|^2, \quad (1)$$

where  $\nu$  is the outgoing velocity of the emitted particle that carries angular momentum  $l$ . The distance  $R$  is chosen to be around the nuclear surface where the wave function describing the cluster in the mother nucleus is matched with the outgoing cluster-daughter wave function.  $H_l^+$  is the Coulomb-Hankel function [2]. Compared to the reduced width of Ref. [9], the formation probability  $|RF_c(R)|^2$  gives a more precise and unambiguous assessment of the clustering process. From Eq. (1), one can extract the experimental formation probability if the corresponding half-life has been determined.

Figure 2(a) shows the formation probabilities  $|RF_c(R)|^2$  as extracted from the experimental half-lives and  $Q_\alpha$  values from the known ground-state to ground-state  $\alpha$ -decay transitions in even-even isotopes from platinum ( $Z = 78$ ) to thorium ( $Z = 90$ ) and neutron number ranging from  $N = 92$  to 140. From the behavior of  $|RF_c(R)|^2$  around the neutron shell closure at  $N = 126$  (e.g.,  $\pm 10$  neutrons away from the closed shell), one can deduce a global trend. Below the shell closure,  $|RF_c(R)|^2$  decreases as a function of rising neutron number, reaching its lowest values at the shell closure. When the shell closure is crossed, a sudden increase in  $|RF_c(R)|^2$  (typically by 1 order of magnitude) is observed followed by an additional but smaller increase (typically by a factor of 2) and finally saturation occurs.

To obtain a microscopic understanding of this behavior we will investigate the mechanism governing the clustering of  $\alpha$  particles at the nuclear surface. This clustering is induced by the pairing force acting among the neutrons and the protons that constitute the  $\alpha$  particle [2]. In nuclear

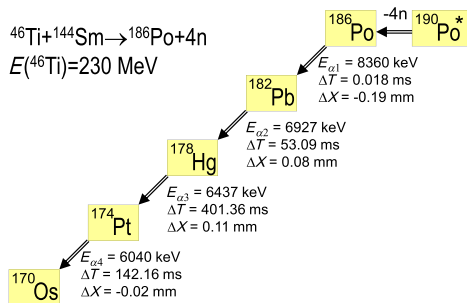


FIG. 1 (color online). An example of an  $\alpha$ -decay chain for  $^{186}\text{Po}$ . For each  $\alpha$  decay within the chain are shown:  $\alpha$ -decay energy  $E_\alpha$ , time ( $\Delta T$ ), and position difference ( $\Delta X$ ) relative to the previous decay.

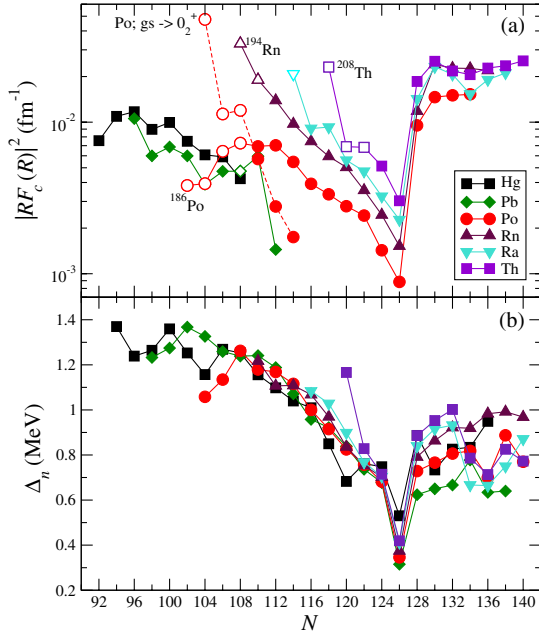


FIG. 2 (color online). (a)  $\alpha$ -particle formation probabilities for the decays of the even-even isotopes in the mercury to thorium region as a function of the neutron number  $N$  of the mother nuclei. The isotopes for which the new data were obtained in our SHIP experiments are marked by open symbols. The symbols connected by a dashed line show the  $|RF_c(R)|^2$  values for fine-structure  $\alpha$  decays of polonium isotopes to the  $0_2^+$  states in the daughter lead nuclides. (b) Neutron pairing gaps in even-even lead to thorium nuclei extracted from experimental binding energies [Eq. (3)]. The proton pairing gaps show a similar dip at the  $Z = 82$  shell closure.

systems, the pairing collectivity manifests itself through the coherent contribution of many configurations. This feature is responsible not only for the clustering of the four nucleons that eventually constitute the  $\alpha$  particle but also for the two-particle clustering, as seen in two-particle transfer reactions between collective pairing states [27,28].

Within the BCS approach, the two-particle formation amplitude is proportional to  $\sum_k u_k v_k$ , where  $u_k$  and  $v_k$  are the standard occupation numbers. To this, one has to add the overlaps of the corresponding proton and neutron radial functions with the  $\alpha$ -particle intrinsic wave function on the nuclear surface [2]. For neighboring nuclei, these overlaps of radial wave functions do not differ strongly from each other and may be considered constant. On the other hand, the corresponding pairing gap is given by

$$\Delta = G \sum_k u_k v_k, \quad (2)$$

where  $G$  is the pairing strength. We thus find that the  $\alpha$  formation amplitude is proportional to the product of the proton and the neutron pairing gaps. This implies that the pairing gaps can serve as a signature of the change in clusterization as a function of the nucleon numbers.

To probe this conjecture, we compare the formation probabilities extracted from the experimental half-lives to the corresponding pairing gaps. The latter can readily be obtained from the experimental binding energies [29] as

$$\Delta_n(Z, N) = \frac{1}{2} [B(Z, N) + B(Z, N - 2) - 2B(Z, N - 1)]. \quad (3)$$

These gaps are shown as a function of the neutron number in Fig. 2(b). One indeed sees a striking similarity between the tendency of the pairing gaps in this figure with the  $\alpha$ -particle formation probabilities. This similarity makes it possible to draw conclusions on the tendencies of the formation probabilities. The nearly constant value of  $|RF_c(R)|^2$  for mercury and lead for neutron numbers  $N \leq 114$  is due to the influence of the high- $j$   $i_{13/2}$ ,  $h_{9/2}$ , and  $f_{7/2}$  orbitals at the lower end of the  $N = 82$  to  $126$  shell. As these highly degenerate shells are being filled, the pairing gap [Eq. (3)] and, therefore, the formation probability, remain constant. A quite sharp decrease of formation probability and pairing gap happens as soon as the low- $j$  orbitals such as  $2p_{3/2}$ ,  $1f_{7/2}$ , and  $2p_{1/2}$  start to be filled between  $N = 114$  and  $N = 126$ . Finally, at  $N = 126$ , the pairing reaches its lowest value.

As the neutron pairing gap  $\Delta_n$  varies smoothly, the two-neutron clustering in the mercury, lead, polonium, radon, and radium isotopes is all of a similar character. The new data obtained, i.e., for those with neutron number between 102 and 110, show an enhanced  $\alpha$ -formation probability for the radon, radium, and thorium isotopes compared to that of mercury and lead. This behavior is a clear manifestation of crossing the  $Z = 82$  shell. The importance of the new experimental data reported here can be assessed by noticing that, in previous analyses [10,11], the behavior discussed above was not observed just due to the lack of experimental data for the region of crossing  $Z = 82$ .

The most neutron-deficient polonium isotopes behave differently. For  $A(\text{Po}) \leq 196$  ( $N \leq 112$ ) the formation probabilities are only slightly larger than or similar to the corresponding ones in the lead isotopes. Furthermore,  $^{186}\text{Po}$  shows a value that is even 40% smaller than the one in  $^{184}\text{Pb}$ . As shown in Refs. [14–16], this is due to the mixing of normal and intruder configurations in the ground states of the most neutron-deficient polonium isotopes. These intruder configurations correspond to proton pair excitations across the  $Z = 82$  gap and induce deformation in the polonium ground states as recently shown experimentally by charge radii measurements [30,31], whereas the lead ground states remain spherical [13]. This leads to a retardation in the ground-state to ground-state  $\alpha$  decay of the neutron-deficient polonium isotopes in contrast to the decay towards the excited  $0^+$  states in lead [14]. When adding to Fig. 2(a) the  $|RF_c(R)|^2$  values for  $\alpha$  decays to the  $0_2^+$  states, a clear gap between lead and polonium similar to the radon and radium case is evident.



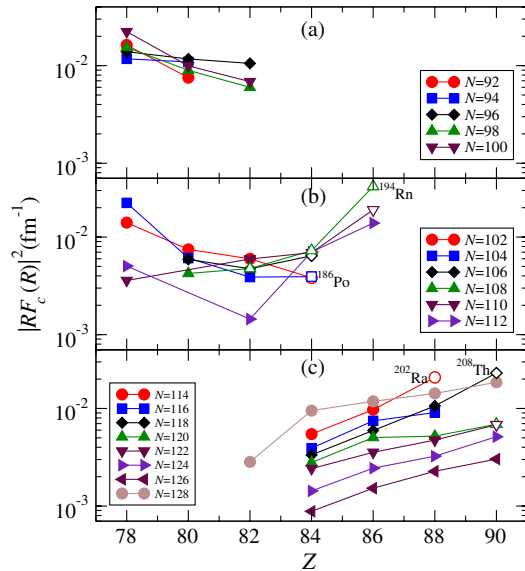


FIG. 3 (color online). Alpha-particle formation probability as a function of the proton numbers  $Z$  of the parent nuclei for three regions, see the main text. The middle panel shows the region where experimental data cover the crossing of the  $Z = 82$  shell gap. The isotopes for which the new data were obtained in our SHIP experiments are marked by open symbols.

Figure 3 shows  $|RF_c(R)|^2$  as a function of  $Z$  for a number of isotones. The information on the isotonic chains is much more limited compared to that of the isotopic chains. Therefore, the data are presented in three different areas. Above  $Z = 82$  [Fig. 3(c)] and below  $Z = 82$  [Fig. 3(a)], one notices the typical decreasing behavior of  $|RF_c(R)|^2$  above and below the shell closure, as discussed above. Figure 3(b), where the crossing of the  $Z = 82$  shell is effectively shown, indicates the typical increase in  $|RF_c(R)|^2$  not between lead and polonium isotopes but between lead and radon and higher  $Z$  isotones. The sudden increase in the formation amplitude, by moving away in either direction from  $Z = 82$ , is smaller compared to the effect at  $N = 126$ . This is because around  $^{208}\text{Pb}$  both proton and neutron shell closures intervene. As can be seen from Fig. 2(a), the increase in  $|RF_c(R)|^2$  beyond  $N = 126$  attenuates when going to higher  $Z$  values. But it has to be stressed that the minimum in the formation probability seen in this figure is a signature of the closeness of the  $Z = 82$  shell. Its effect would be greatly magnified if it would be possible to reach a neutron magic number, e.g.,  $N = 82$ .

We can now propose a generic form for the  $\alpha$ -particle formation amplitude as a function of nucleon (proton or neutron) number. When the nucleons are filling a new major closed shell (e.g.,  $N$  between 82 and 126), the  $\alpha$ -particle formation amplitude is nearly constant as high- $j$  orbitals are filled first. As soon as the low- $j$  orbitals are filled, the formation probability smoothly reduces until one reaches again a closed proton or neutron configuration,

i.e., the upper boundary of the major shell. Here, a minimum is reached. Crossing the closed shell induces a steep increase followed by an approximately constant trend discussed above. However, when strong particle-hole excitations across closed shells are encountered, this “generic” form of the  $\alpha$ -particle formation probability is altered as one clearly sees in the light polonium isotopes. Such effects, however, do not invoke a disappearance of the influence of the  $Z = 82$  shell gap on the  $\alpha$ -decay probability.

In conclusion, we reported on the identification of  $^{186}\text{Po}$ , the most neutron-deficient polonium isotope known so far, and measured its  $\alpha$ -decay properties. Combining these data with our recently obtained results for the neutron-deficient isotopes with  $Z > 82$ , we extracted the  $\alpha$ -particle formation probabilities following Ref. [27]. We discussed the formation probability in the framework of the pairing force acting among the protons and the neutrons forming the  $\alpha$  particle and could thus show a striking resemblance between the phenomenologically deduced pairing gap and the formation probabilities. This clearly indicates the influence of the  $N = 126$  and  $Z = 82$  shell closures on the  $\alpha$  formation amplitude around  $^{208}\text{Pb}$ . To further investigate these findings, it would be interesting to reduce the uncertainties on the  $\alpha$ -decay properties of  $^{192,194}\text{Pb}$  and measure the  $\alpha$  decay of even-even lead isotopes up to  $^{210}\text{Pb}$ . This nucleus, according to our calculations, should have an  $\alpha$ -particle formation probability similar to that of  $^{210}\text{Po}$ .

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