Evidence from α Decay That $Z = 82$ Is Not Magic for Light Pb Isotopes

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To resolve the reported anomalous behavior of the α-decay rates of ^{186,188,190,192}Pb we determined more accurate α-decay branches for these isotopes. Our data result in α-decay reduced widths whose dependence on N is similar to that observed for other elements. However, contrary to the expectation of a shell effect at $Z = 82$, this new information indicates lead nuclei to be less hindered toward α decay than mercury isotopes. It appears that midway between $N = 82$ and $N = 126$ the proton number of 82 is not magic.

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Alpha-decay transitions between ground states of doubly even nuclei are taken to represent unhindered decays. Reduced widths for these *s*-wave transitions behave in a regular fashion as a function of both neutron and atomic number. They are largest for nuclei two or four particles beyond a closed shell (with sharp minima at the shell) and they then decrease as the next closure is approached. The *s*-wave widths for ^{186,188,190,192}Pb, however, have been reported[1] to behave anomalously, i.e., they purportedly increase by a factor of 30 between ^{186}Pb ($N = 104$) and ^{192}Pb ($N = 110$) instead of decreasing as one nears $N = 126$. Theoretical calculations[2,3] have not reproduced this unusual behavior.

In Ref. 1, the [electron-capture (EC) $+\beta^+$] strengths were deduced from K x-ray intensities. A number of corrections are involved in such determinations. A more precise method entails the use of a known (EC$+\beta^+$) decay scheme. To obtain more reliable α-branching ratios we undertook the investigation of the (EC$+\beta^+$) decay schemes of these neutron-deficient lead isotopes in conjunction with studies of their α-decay properties. Results for ^{192}Pb, ^{190}Pb, and ^{188}Pb have been published in Toth *et al.*,[4] Ellis-Akovali *et al.*,[5] and Toth *et al.*,[6] respectively. Herein we present new information on ^{186}Pb and discuss the partial α half-lives for all four isotopes within the overall framework of α-decay rates for even-even nuclei with $Z \geq 78$.

While ^{188,190,192}Pb had been produced in ^{16}O $+ ^{180}$W bombardments at the Oak Ridge isochronous cyclotron, the machine parameters are such that the intense $^{16}O^{5+}$ beam could not be accelerated to a sufficiently high energy to induce the reaction ^{180}W(^{16}O, $10n$). The recent availability of energetic heavier ions from the Holifield Heavy Ion Research Facility allowed us to produce ^{186}Pb in the reaction ^{160}Dy(^{35}S, $6n$) by bombarding a 2.92-mg/cm^2-thick dysprosium metal foil enriched in ^{160}Dy to 78.9% with 200-MeV ^{32}S ions. As in the earlier studies,[4–6] reaction products were mass separated with the UNISOR (University Isotope Separator at Oak Ridge) on-line facility, collected onto an automated tape system, and transported to a counting station. Singles and coincidence γ-ray data were accumulated simultaneously with a large-volume Ge(Li) detector and a γ-x detector. A Si(Au) surface-barrier detector was used for α-particle counting. All three detectors were placed in calibrated geometries so that absolute counting rates could be determined. Production and collection cycles were 20 s in duration while the counting time was broken up into four 5-s intervals.

The α spectrum, shown in Fig. 1(a), is dominated by the single group known[7,8] to belong to ^{186}Pb α decay. The energy of this peak was determined to

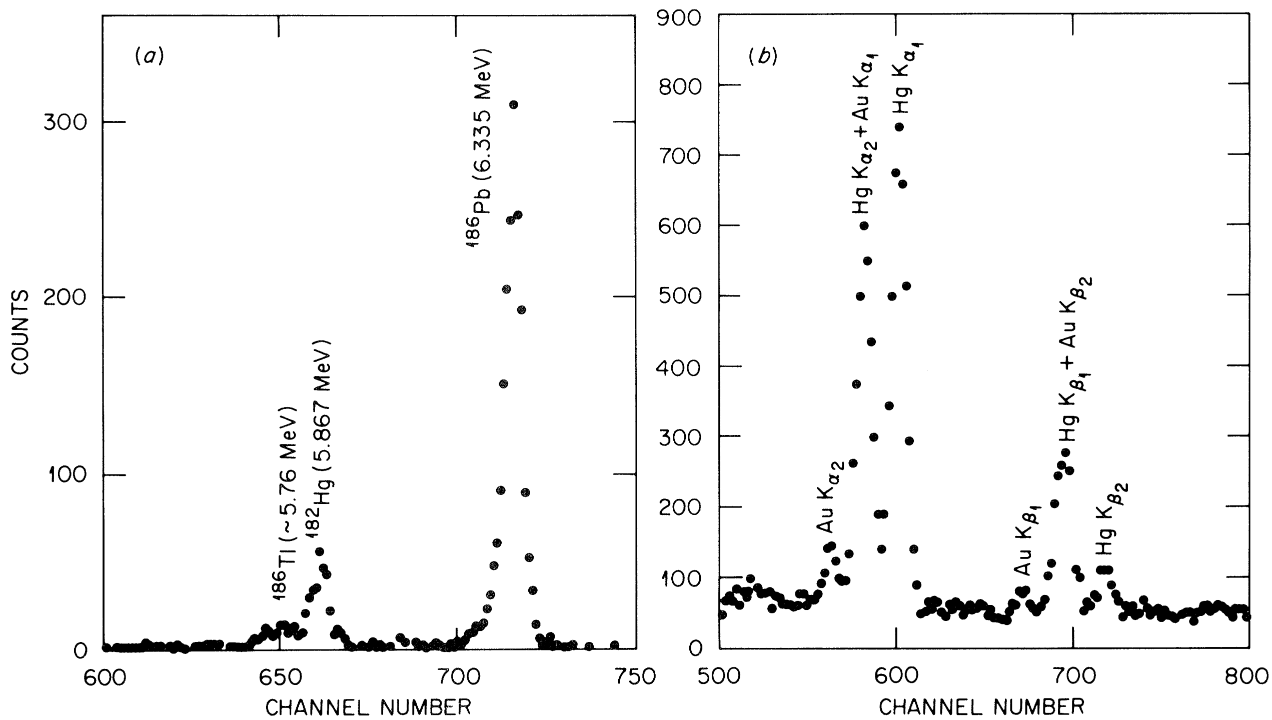

FIG. 1. (a) Singles α-particle spectrum and (b) total coincidence x-ray spectrum, measured at $A = 186$.

be 6335 ± 10 keV; previously measured energies are 6320 ± 20 (Ref. 7) and 6332 ± 10 keV (Ref. 8). The α group decayed with a half-life of 4.7 ± 0.1 s in agreement with the value of 4.79 ± 0.05 s reported by Schrewe *et al.*[8] but in disagreement with the value 8 ± 2 s published by Le Beyec *et al.*[7] The two less intense peaks in Fig. 1(a) belong to the ^{186}Pb α-decay daughter, ^{182}Hg (see e.g., Hansen *et al.*[9]), and to ^{186}Tl (see Ijaz *et al.*[10]).

Gamma-ray spectra clearly showed the presence of ^{181}Tl and ^{186}Hg (Béraud *et al.*[11] and Cole *et al.*[12]) produced in reactions involving charged-particle plus neutron evaporation, but no transition that could be assigned to ^{186}Pb on the basis of either half-like or K-x-ray coincidences. There was no indication of a short-lived component, consistent with a 4.7-s half-life, in the decay curve of the annihilation radiation peak. Thallium K x rays were also not observed. Here the situation was somewhat blurred by the presence of lead x rays due to fluorescence. The lead $K\alpha_2$ peak has an energy just 0.07 keV below that of the thallium $K\alpha_1$ x-ray group. (The thallium $K\alpha_2$ peak was masked by the much more intense mercury $K\alpha_1$ x rays.) However, the intensity of the lead $K\alpha_1$ x rays summed over the 20-s counting period, when corrected by the theoretical $K\alpha_1/K\alpha_2$ ratio, accounted for all of the events seen in the peak at 72.80 keV with no indication of thallium $K\alpha_1$ x rays. A small excess of counts in this peak was observed during the first 5-s counting interval, but it was adequately accounted for by the K-shell electron conversion of the $E3$ 374.0-keV transition associated[11,12] with the 3.0-s ^{186}Tl isomer. Figure 1(b) illustrates the absence of thallium K x rays. It shows the total coincidence spectrum, in the neighborhood of the K-x-ray energy region, accumulated in the γ-x detector. While gold and mercury K x rays are seen in Fig. 1(b), there are no thallium x rays from either ^{186}Pb $(EC+\beta^+)$ or ^{186}Tl isomeric decay. [No transitions have been observed (Refs. 11 and 12 and present study) in coincidence with the ^{186}Tl $E3$ γ ray; if any exist their energies must be less than the thallium K-shell binding energy.] Finally, we note that the ^{186}Tl isomer apparently has a low spin.[12] One would therefore expect most of ^{186}Pb $(EC+\beta^+)$ decay to proceed either directly or through intermediate states to this isomer. The decay curve for the 374.0-keV γ ray showed a single (3.0 ± 0.2)-s component with no hint of a parent-daughter, growth and decay, correlation, further indicating an absence of ^{186}Pb $(EC+\beta^+)$ decay.

Our data therefore suggest a ^{186}Pb α/total branching ratio of 100%, a value which is much larger than the 2.4% branch estimated[7] from expected cross sections for the production of ^{186}Pb in heavy-ion reactions. On the basis of their measured ^{186}Pb and ^{189}Pb α branches Hörnshoj *et al.*,[1] in their discus-

TABLE I. Alpha-decay properties of lead isotopes.

	E_α (keV)	$T_{1/2}$ (min)	α branch	δ^2 (MeV)
			Current data[a]	
^{192}Pb	5112(5)	3.5(1)	$5.7\times10^{-5}(10)$	$0.049^{+0.014}_{-0.012}$
^{190}Pb	5577(5)	1.2(1)	$9.0\times10^{-3}(20)$	$0.094^{+0.036}_{-0.028}$
^{188}Pb	5980(5)	22(2)	0.22(7)	$0.114^{+0.055}_{-0.041}$
^{186}Pb	6.335(10)	4.7(1)	1.0	0.09
^{184}Pb	...	...	...	...
			Earlier data	
^{192}Pb	5110[b]	2.3(5)[c]	$6.9\times10^{-5}(24)$[d]	0.094
^{190}Pb	5580(10)[c]	1.2(2)[c]	$2.1\times10^{-3}(7)$[d]	$0.021^{+0.017}_{-0.010}$
^{188}Pb	5980(10)[e]	24.5(15)[f]	$3.3\times10^{-2}(11)$[d]	$0.015^{+0.009}_{-0.007}$
^{186}Pb	6320(20)[g]	8(2)[h]	4.8×10^{-2} [d]	0.0028
^{184}Pb	6632(10)[i]	0.55(6)[i]	1.0(estimate)	0.06

[a]Data are from Refs. 4–6, and this study.
[b]Ref. 14; other value: 5060(30) (Ref. 7).
[c]Ref. 7.
[d]Ref. 1.
[e]Ref. 7; other values: 5975(15) (Ref. 15); 5990(15) (Ref. 8).
[f]Ref. 15; other value: 26(2) (Ref. 7).
[g]Ref. 7; other value: 6332(10) (Ref. 8).
[h]Ref. 7; other value: 4.79(5) (Ref. 8).
[i]Ref. 8.

sion of α widths, increased the ^{186}Pb ratio by a factor of 2, i.e., to 4.8%. This augmented branch is still much less than our value.

In discussing α-decay rates we consider them within the theoretical formalism developed by Rasmussen[13] wherein decay probabilities are represented by a reduced width, δ^2. Decay energies, half-lives, and α branches are needed to compute the reduced widths. In Table I we compare these quantities for ^{186,188,190,192}Pb as obtained in our current investigations with data from Refs. 1, 7, and 8, and from Siivola[14] and Hornshøj *et al.*[15] Large discrepancies exist, not only for the ^{186}Pb branching ratio, but for the ^{188}Pb and ^{190}Pb α branches as well. As a result, the corresponding widths are increased by factors of 32, 7, and 4.5. Our width for ^{192}Pb, on the other hand, is less than the value based on the earlier data because the nuclide's half-life is 3.5 min, rather than 2.3 min as reported by Le Beyec *et al.*[7] and used in Ref. 1. Even though the ^{184}Pb α branch has not been measured we have included in Table I its half-life and decay-energy data[8] and calculated a reduced width by assuming a branching ratio of 100%.

Figure 2 shows s-wave reduced widths for nuclei with Z from 78 to 100 plotted as a function of N. Information used to calculate the reduced widths for isotopes with $A \geqslant 196$ is taken from Refs. 16 and 17. Data for lead, mercury, and platinum nuclei are taken from Table I; Refs. 8, 9, and 15; and Schneider *et al.*[18] and Ellis, Toth, and Carter,[19] respectively. One sees in Fig. 2 the regularity of the reduced widths as a function of neutron number with the extremely sharp break at $N=126$. This discontinuity has been shown (see, e.g., Mang[20]) to be a shell-structure effect. A less pronounced minimum is seen at the subshell closure at $N=152$. The lead anomaly is indicated by the dashed line which connects the ^{186,188,190,192}Pb widths calculated from the earlier data summarized in Table I. The widths for ^{186,188,190,192}Pb computed from our data are shown by the open points. It is clear that they have a dependence on N which is similar to that observed for other elements. However, these new data indicate neutron-deficient lead isotopes to be less hindered toward α decay than mercury isotopes, contrary to the expectation of a shell effect at $Z=82$.

Our results seem to be related to the disappearance of the $Z=82$ gap in the vicinity of $N=114$ which has been predicted[21] on the basis of Hartree-Fock-Bogoliubov calculations of proton single-particle energies, and to the existence of varying shapes in mercury and platinum isotopes in this mass region. In ^{182,184,186,188}Hg it has been shown[22] that well-deformed prolate bands cross the slightly oblate ($\epsilon \sim 0.1$) ground-state bands. Platinum nuclei with $A < 190$, on the other hand, are believed[23] to be prolate in their ground states. If one supposes that ^{186,188,190,192}Pb also have slightly oblate ground states then α transitions from oblate mercury to prolate platinum isotopes would be expected to be

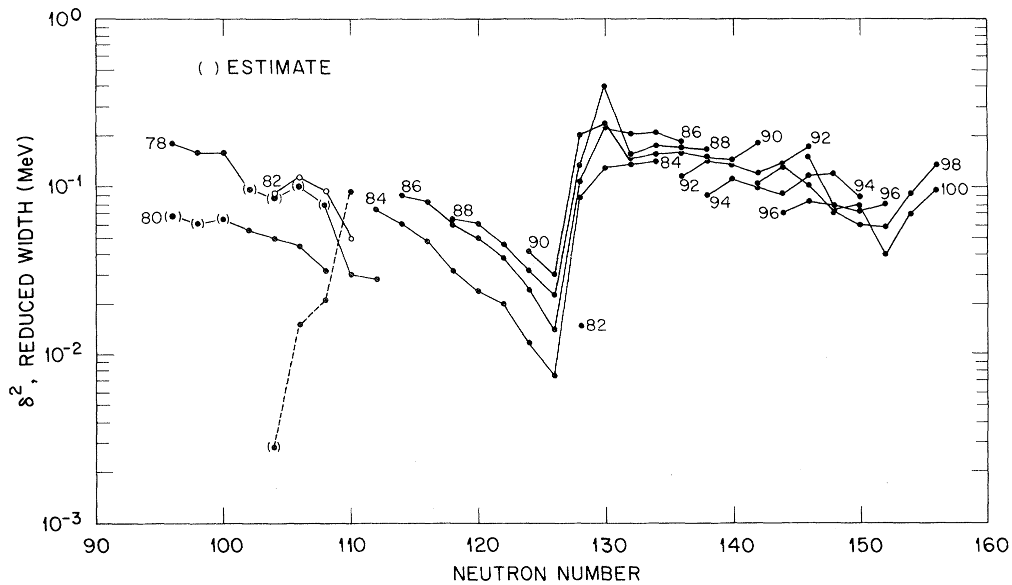

FIG. 2. Reduced widths for s-wave α transitions plotted as a function of N for isotopes with Z from 78 to 100. The dashed line connects widths for ^{186,188,190,192}Pb calculated from earlier data. Open points for $Z=82$, connected by the full line, are widths for ^{186,188,190,192}Pb calculated from our experimental results.

hindered whereas lead α decays, which do not involve shape changes between the parent and daughter nuclei, would not be. (Low-lying 0^+ excited states in ^{192,194,196,198}Pb have been described[24] by oblate two-particle, two-hole configurations; their ground states, however, have been assumed to be spherical.) Further detailed calculations of α-decay rates incorporating shape changes are needed. If such theoretical results do indeed agree with our experimental observations, then the study of α-decay rates may prove to be a useful tool to deduce information concerning nuclear shapes. Finally, we note that the ^{184}Pb reduced width (see Table I) is appreciably smaller than our widths for ^{186}Pb and ^{188}Pb which seems to indicate the reappearance of the $Z=82$ gap for $N<102$.

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