γ-ray spectroscopy of ^{209}Tl

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States in ^{209}Tl were populated using a multinucleon transfer reaction with a ^{136}Xe beam impinging on a thick ^{208}Pb target at $E = 785$ MeV. The beam was pulsed at 825-ns intervals in order to perform isomer decay spectroscopy. The known $J^\pi = 17/2^+$ isomer in ^{209}Tl was located at 1228(4) keV and measured to have a half-life of $T_{1/2} = 146(10)$ ns. A second isomer with $J^\pi = 13/2^+$ was found to have $T_{1/2} = 14(5)$ ns. The previously suggested low-energy X and Y transitions were found to have energies 57(2) and 47(2) keV respectively, while the measurement of conversion coefficients and a new decay path make the spin assignments below the isomers experimentally firm. Correlating the delayed γ transitions with the prompt beam flash allowed the decay of states above the isomer to be found. The longer-lived isomer represents full alignment of the simplest two-particle, one-hole configuration and illuminates the remarkably weak coupling of the proton hole to the ^{210}Pb core.

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I. INTRODUCTION

The ^{208}Pb nucleus is the anchor point for much of our understanding of all heavy nuclei and has been extensively studied. Electron and hadron scattering [1–5] gives us a uniquely precise picture of the mean-field topology, while transfer reactions to immediately neighboring nuclei establish the location and purity of single-particle states and inform on the role of residual interactions [6–9]. It is surprising that the spectroscopic properties of excited states in nuclides only two or three nucleons removed from this bastion are almost totally unknown. More specifically, this deficiency applies to nuclides of lower Z, that is, $Z < 82$, and higher N, that is, $N > 126$. Colloquially, this region is called the domain "south" of ^{208}Pb. Nuclei south of ^{208}Pb are of considerable interest for both structural and astrophysical reasons. In structure, the issue centers on the robustness of the $N = 126$ spherical shell closure and its magicity. This issue is academically interesting, but has crucial importance for r-process nucleosynthesis of heavy elements. This paper reports an experiment which establishes the spins and parities of states in ^{209}Tl ($Z = 81, N = 128$) up to the previously observed isomer, which is now placed at 1228 keV.

The domain south of ^{208}Pb is poorly known as all the nuclides in this region are hard to synthesize. Heavy-ion fusion, the reaction which has allowed the proton-rich landscape to be explored, cannot make any of the nuclei of interest due to the Coulomb-induced curvature of the valley of stability. Direct light-ion reactions can only inform us about nuclides very near stability. Only fast fragmentation of heavy ions has allowed progress but only in out-of-beam studies [10,11]. Recently, heavy-ion multinucleon transfer near the Coulomb barrier has emerged as a potentially powerful tool for moving south and southeast of ^{208}Pb. Some calculations have suggested the yields are high enough for detailed spectroscopy, which with current technology means production cross sections of 100s μb [12–14]. In this work we used heavy-ion multinucleon transfer involving a ^{136}Xe beam and a ^{208}Pb target at 785 MeV $\sim$9% above the Coulomb barrier. The original goal of this experiment was to measure production cross sections, especially to probe this approach for producing very heavy nuclei, and these data have been published [15]. The present report is of a nuclide only one proton removed and two neutrons added to ^{208}Pb, so this is a small step from stability. However, significant new spectroscopic data were collected.

II. ^{209}Tl HISTORY

The understanding of excited states in ^{209}Tl is based mainly on two previous studies. First, a proton removal experiment, ^{210}Pb(t,α)^{209}Tl, located seven states in ^{209}Tl [16]. More recently, a fragmentation study of ^{238}U found an isomer in ^{209}Tl which was reported to have $T_{1/2} = 95(11)$ ns and had three delayed γ rays [11], one of which could be associated with the decay of an excited state identified in transfer. A shell-model calculation associated with the latter work indicated the isomer probably has $J^\pi = 17/2^+$ and thus at least two unobserved γ rays, labeled X and Y, should lie in the isomer decay cascade but were not observed. Measurements of the ^{209}Hg β decay [10] and ^{213}Bi α decay [17] did not add significantly to this picture.

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III. THE EXPERIMENT

The present experiment was performed at the ATLAS accelerator in Argonne National Laboratory. A beam of $\sim$1 pnA of ^{136}Xe at 785 MeV impinged on a 49-mg/cm^2 ^{208}Pb target. The Couloumb barrier is 718 MeV [12], so only the first $\sim$2 mg/cm^2 of the target is used, as the energy loss of ^{136}Xe beam in ^{208}Pb target is large (30 MeV/mg/cm^2) [18]. Thus, the target was thick enough to stop the beam and all reaction products. The target was installed in Gammasphere [19], in which 93 detector modules were installed, of which 90 detectors were operational and 15 were rejected in the analysis due to inadequate performance. The experiment used the original analog electronics and HPGe count rate was limited at $\sim$10 kHz per detector. Tantalum and copper absorbers were used to reduce the prolific flux of x rays. Gammasphere was operated in constant fraction discrimination (CFD) mode to ensure good timing of events. Data were collected for 48 h in doubles trigger and 48 h in triples trigger modes. Scaling yields from the information in Ref. [15], we estimate the production cross section for the ^{209}Tl isomer was 80(20) μb, although this cross section was not provided in the original report.

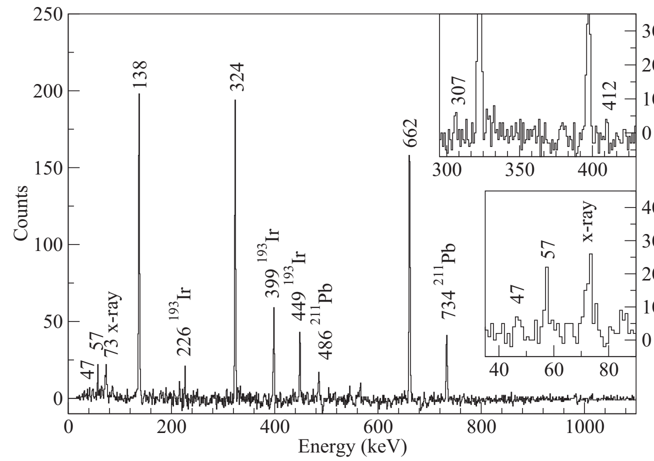

FIG. 1. ^{209}Tl γ-ray spectrum below the $T_{1/2} = 146$ ns isomer. This spectrum was obtained by summing all possible gates in the delayed γ-γ-γ matrix. Breakthrough γ transitions from other nuclei could not be completely suppressed but have been identified. The insets shows the new decay path and the new low-energy γ rays in the main cascade.

IV. ANALYSIS

The data were reduced into a series of γ-γ and γ-γ-γ matrices for analysis. Two time domains were chosen: prompt (P), which was within a 50-ns window centered on the beam burst, and delayed (D), which covered γ rays detected 31 to 750 ns after the peak of the beam burst. The energy matrices were then further gated by temporal conditions, for example, a DDD triple correlation matrix was generated in which three γ rays were found in prompt coincidence with each other, but during the delayed interval, while a PPP matrix had three γ rays in the prompt time window. Another two γ-γ-γ matrices PDD and PPD were also sorted. Those matrices will be used to correlate between transitions above and below the isomer.

V. RESULTS AND DISCUSSION

The low-lying states in ^{209}Tl are best described as two-particle, one-hole configurations relative to the ^{208}Pb core. In this case, the two particles are valence neutrons. At low excitation energies, up to the $J^\pi = 17/2^+$ isomer, they occupy the lowest neutron state above the $N = 126$ shell gap, i.e., the $2g_{9/2}$ orbit. The coupling of these two neutrons can be seen in the lowest states in ^{210}Pb, that is, a j^2 configuration for identical particles that have a residual interaction, as discussed by Schiffer and True [20]. The hole in this case is a proton hole in the $Z = 82$ closed shell. The orbits below the Fermi surface are $3s_{1/2}$, $2d_{3/2}$, $1h_{11/2}$, and $2d_{5/2}$. Accepting the single-particle shell model as valid, the spectrum of ^{207}Tl reveals the relative locations of these states below $3s_{1/2}$ as 351, 1348, and 1683 keV [21]. The coupling between the valence neutrons and the proton hole is very weak, as evidenced by the lowest-lying states in ^{208}Tl which form a one-neutron-particle–one-proton-hole $(2g_{9/2} \otimes 3s_{1/2})_{J=4,5}$ doublet separated by only 40 keV, with the higher-spin state most bound [22]. Thus, the low-lying states of ^{209}Tl may be anticipated to be a near-degenerate series of multiplets arising from coupling $3s_{1/2}$ or $2d_{3/2}$ proton holes to the ^{210}Pb neutron particle states. The lowest of these multiplet states were populated in the ^{210}Pb(t,α)^{209}Tl direct removal reaction [16], although only with the current γ-decay study does the true spin sequence emerge.

The fully aligned $\nu(g_{9/2})^2$ $J^\pi = 8^+$ state in ^{210}Pb at 1278 keV [23] is isomeric, with a half-life of $T_{1/2} = 201(17)$ ns [24], and has reduced matrix element $B(E2) = 0.7(3)$ W.u. [25]. The isomerism arises from the fact that the fastest electromagnetic de-excitation is an $E2$ decay to the $J^\pi = 6^+$ member of the multiplet, which lies only 83(3) keV lower. The $J^\pi = 6^+$ member of the decay cascade in ^{210}Pb is also isomeric; it decays by a 97(3)-keV γ transition [23] and has a half-life of $T_{1/2} = 49(6)$ ns [24], implying $B(E2) =$ 2.1(8) W.u. [25]. One may anticipate similar isomers in ^{209}Tl with $[\nu(g_{9/2})^2 \otimes \pi(s_{1/2})^{-1}]_{J=17/2,13/2}$ configurations. A candidate for the upper isomer has been reported [11] as having a half-life of $T_{1/2} = 95(11)$ ns. A goal of the current research was to locate this isomer in excitation energy by finding the missing X and Y transitions and to remeasure the isomer half-life, so to rigorously determine its spin and parity. A sample of the current data is shown in Fig. 1. It emerges that whereas the decay of ^{210}Pb is very simple, with a single cascade of four $E2$ transitions carrying away all the angular momentum, the situation in ^{209}Tl is more subtle, as the weak-coupling multiplets offer other spin-favored decay pathways which are faster and carry almost all the decay flux.

Starting at the ground state, we can use all the known data to build the decay scheme shown in Fig. 2 with little uncertainty as to the level spins and parities. Table I summarizes the levels energies, angular momentum, and parities as well as γ-ray intensities and multipolarities. The internal conversion

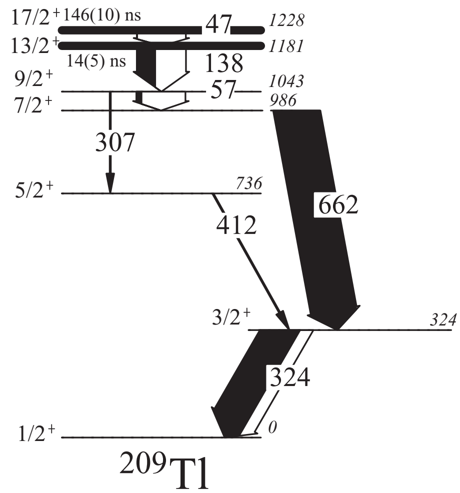

FIG. 2. ^{209}Tl decay scheme below the $T_{1/2} = 146$ ns isomer.

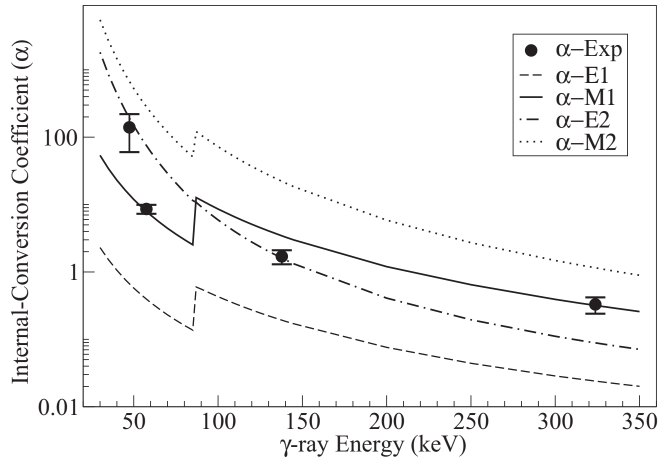

FIG. 3. The lines represent the theoretical internal-conversion coefficients for different multipole transitions in ^{209}Tl [26]. The dots represent the experimental internal-conversion coefficient. All four transitions measured in this work are consistent with pure multipoles.

coefficients deduced from intensity balance considerations are also tabulated.

A. The ground state

The ground state of ^{209}Tl has spin and parity $J^\pi = 1/2^+$, determined from its β decay, which has very strong branches to $J = 1/2$ levels in the daughter nucleus, ^{209}Pb. This assignment is completely consistent with the strong population in the (t,α) reaction, the neighboring $N = 126$ isotope ^{207}Tl, and the shell-model expectation of an uncoupled proton in the $3s_{1/2}$ state which lies immediately below the proton Fermi surface in ^{208}Pb.

B. The 324-keV state

This state was populated in the (t,α) one-proton removal reaction on a ^{210}Pb target [16]. Although the measured angular distribution of α particles is poor, and there is no spectroscopic factor, there is little doubt this is the $J^\pi = 3/2^+$ $d_{3/2}$ hole state, the equivalent of the 351-keV level in the $N = 126$ isotope ^{207}Tl. This state is also populated in the α decay of ^{213}Bi [17] and the β decay of ^{209}Hg [10]. The new 412-keV decay to this level slightly increases the population of the state, and so influences the inferred conversion coefficient. The data allows us to extract the coefficient for the 324-keV transition to be $\alpha_{\text{tot}}^{\text{Exp}} = 0.33(9)$. This is consistent with the theoretical expectations of a pure $M1$ decay ($\alpha_{M1}^{Th} = 0.32$) [26], as any significant $E2$ contribution to the decay decreases the degree of conversion ($\alpha_{E2}^{Th} = 0.089$) [26] as shown in Fig. 3. This measurement is consistent with the recent measurement of $\alpha_{\text{tot}}^{\text{Exp}} = 0.36(6)$ [11]. It is also consistent with a direct measurement of the electron-to-γ ratio following the α decay of ^{213}Bi which determined $\alpha_{\text{tot}}^{\text{Exp}} = 0.40(9)$ [27]. The group who made this latter measurement also made a later determination of $\alpha_{\text{tot}}^{\text{Exp}} = 0.131(15)$ [27], which is inconsistent with the other three measurements, as it has a claimed precision which makes it a statistical outlier. This smaller value, if correct, implies a significant component of the decay is of $E2$ character. The analogous transition in magic ^{207}Tl is almost pure $M1$ in character, consistent with a conversion coefficient of $\alpha_{\text{tot}} \sim 0.3$. In addition, both the γ-ray measurements would have severe intensity balance issues if the smaller conversion coefficient were correct.

TABLE I. The properties of γ rays in ^{209}Tl inferred from this research.

E_i	E_f	J_i^π	J_f^π	E_γ	I_γ [a]	$\alpha_{\text{tot}}^{\text{Exp}}$	Mult.
324	0	$3/2^+$	$1/2^+$	323.9(1)	78(3)	0.33(9)	$M1$
736	324	$5/2^+$	$3/2^+$	411.5(2)	2(1)		$M1$
986	324	$7/2^+$	$3/2^+$	661.8(1)	100(5)		$E2$
1043	986	$9/2^+$	$7/2^+$	57.5(20)	11(2)	8.6(13)	$M1$
1043	736	$9/2^+$	$5/2^+$	307.5(2)	2(1)		$E2$
1181	1043	$13/2^+$	$9/2^+$	137.8(2)	39(2)	1.7(4)	$E2$
1228	1181	$17/2^+$	$13/2^+$	47.4(20)	0.6(3)	140(80)	$E2$

[a] γ-ray intensities corrected for efficiency and normalized to the 662-keV γ ray.

C. The 736- and 947-keV states

The next two excited states were originally reported in the (t,α) study [16]. They arise from coupling the $3s_{1/2}$ proton hole to the $J^\pi = 2^+$ first excited state in ^{210}Pb forming a $J^\pi = 3/2^+,5/2^+$ doublet. Neither the α-particle angular distributions nor cross sections identify which state is which. The current experiment can address this issue, as we have found an isomer decay path which passes through the lower state, which must have $J^\pi = 5/2^+$, as will be discussed in Sec. V D. Our γ-ray measurement places this level at 735.4(5) keV. Consequently, the partner 947-keV level will very likely have $J^\pi = 3/2^+$, though not seen in this current work. The centroid of the $J = 3/2^+,5/2^+$ doublet lies at 841 keV, somewhat higher than the core $J^\pi = 2^+$ state, which lies at 799 keV. This may partially arise from two-level mixing of the $J^\pi = 3/2^+$ states at 324 and 947 keV, which will depress the 324-keV state (from its natural position at 351 keV) and elevate the 947-keV state. The ^{209}Hg β-decay study [10] reported 208- and 739-keV γ rays from these states, both of which are completely absent in our data. This is puzzling, as ^{209}Hg is thought to have a $J^\pi = 9/2^+$ ground-state spin and a Q value of 4.99 MeV, so it is difficult to understand how the now-known main isomer decay path was not at all populated in β decay, while these proposed less-favored states were populated. Inspection of the published β-decay data [10] shows the two reported transitions to be statistically very marginal, and their assignment must be questioned.

D. The 986- and 1043-keV states

These states were not populated in the (t,α) study, probably as their angular momentum is too high to be efficiently reached in transfer. However, they both lie on the isomer decay path. The lower state, at 986 keV, was observed in the original decay study [11] and proposed to have $J^\pi = 7/2^+$. In our data we find the only decay is to the 324-keV state by a 662-keV γ transition. Intensity balances are consistent with this being a pure $E2$ transition. The 986-keV level is fed by a 57(2)-keV γ transition. This is the previously unobserved transition reported as X in the original isomer study [11]. The conversion coefficient for this transition can be inferred. We measure $\alpha_{\rm tot}^{\rm Exp} = 8.6(13)$. This result is consistent with an $M1$ transition ($\alpha_{M1}^{Th} = 7.9$) as shown in Fig. 3. Any higher multipolarity would make the 1043-keV level isomeric. Thus, the new transition indicates the 1043-keV state has $J^\pi = 9/2^+$, and the decay is an $M1$ spin flip transition. The 986- and 1043-keV doublet can be interpreted as the $s_{1/2}$ proton hole being coupled to the $J^\pi = 4^+$ 1098-keV two-neutron state in ^{210}Pb. The centroid of this doublet lies at 1014.5 keV, in good agreement with the assignment. We find that the 1043-keV level also has an $E2$ decay to the $J^\pi = 5/2^+$ state at 736 keV, reminiscent of the core $E2$ cascade. This parallel decay branch, although small, helps solidify the decay scheme and possible spin sequence. Taking the $M1$ matrix element from the low-lying spin-flip $B(M1: 4^+ \rightarrow 5^+) = 2.1(3)$ W.u. [28] in ^{208}Tl and the neutron-rearrangement $E2$ matrix element from the ^{210}Pb core $B(E2: 4^+ \rightarrow 2^+) = 4.8(9)$ W.u. [25], and scaling for mass, conversion, and γ ray energies, one estimates the $E2$ branch to be 1.1%, consistent with the experimentally observed branching ratio 2.6(16)%. Clearly, the spin-flip decay is the fastest decay path, so it diverts most of the isomer decay flux.

E. The 1181-keV state

The 138-keV γ decay from the 1181-keV level is the next transition in the isomer decay cascade. The original isomer experiment [11] suggested this to be of $E2$ nature, based on a measurement of its conversion coefficient. In this work we determine $\alpha_{\rm tot}^{\rm Exp} = 1.7(4)$. A pure $E2$ decay is expected [26] to have $\alpha_{\rm tot}^{Th} = 1.6$, so there is little doubt this assignment is correct and the spin and parity of the 1181-keV level is $J^\pi = 13/2^+$ as shown in Fig 3. Again, this decay is a close analog of the ^{210}Pb core $J^\pi = 6^+$ to 4^+ which has energy of 98 keV. The $J^\pi = 6^+$ state in ^{210}Pb is isomeric with a half-life of 49(6) ns [24]. We find that the time distribution of all the γ rays below the 1181-keV state are consistent with the presence of two higher-lying isomers, the lower state being best fitted with $T_{1/2} = 14(5)$ ns, as shown in Fig. 4, and implying reduced matrix element $B(E2) = 4.2(18)$ W.u.

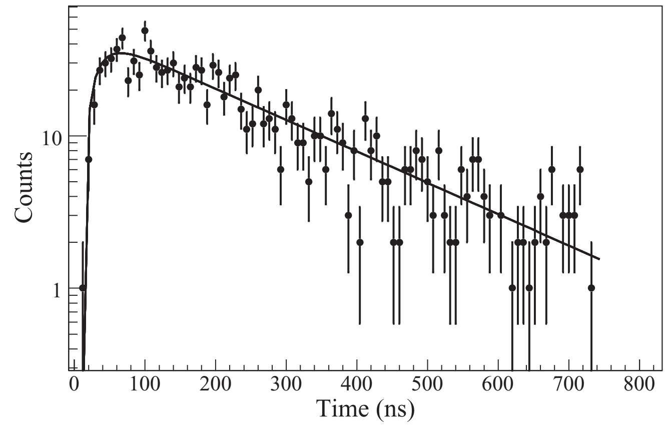

FIG. 4. The time difference between the prompt and delayed transitions with the fitted decay curve of the $17/2^+$ and $13/2^+$ ^{209}Tl isomers shown.

F. The 1228-keV, $T_{1/2} = 146(10)$ ns isomer

The fully aligned $\nu(g_{9/2})^2$ $J^\pi = 8^+$ state in ^{210}Pb lies at 1278 keV [23] and has $T_{1/2} = 201$ ns [24]. One may expect a similar isomer in ^{209}Tl, fully aligned to $J^\pi = 17/2^+$. Another low-energy $E2$ transition, called Y in the original isomer decay experiment [11], was suggested to depopulate the isomer and cause the isomerism. We have found some evidence for this transition at 47.4(20) keV. This would place the isomer at 1228(4) keV. The transition is highly converted ($\alpha_{E2}^{Th} = 200$) [26], and also highly absorbed in the Gammasphere chamber and detector absorbers, and so only has 15(8) counts in the gated spectrum. Nonetheless, after efficiency corrections, the measured internal conversion is $\alpha_{\rm tot}^{\rm Exp} = 140(80)$, consistent with the expected $E2$ conversion coefficient as shown in Fig. 3. The large uncertainty in this inferred conversion coefficient comes mainly from the scant counts in the candidate peak. A more tailored experiment could undoubtedly improve this result.

Several of the decay transitions could be fitted to determine the isomer half-life. The careful selection of energy-selected, background-subtracted, triple-coincident events suppressed random time coincidences in the time difference spectrum, to the extent that the event rate fell to <1 count/ns at the end of the count cycle. Fits including a small constant background were statistically poorer but agreed with the best fit shown. The half-life is measured in this work to be $T_{1/2} = 146(10)$ ns, as shown in Fig. 4, which is longer than the previously reported value. In order to understand this discrepancy we captured the data from Fig. 1 of Ref. [11] and refitted the data points for the beginning of the curve to avoid background subtraction. We find their data is consistent with our longer half-life. The reduced transition probability of the 47-keV transition between the $17/2^+$ and the $13/2^+$ levels is $B(E2) = 1.1(4)$ W.u.

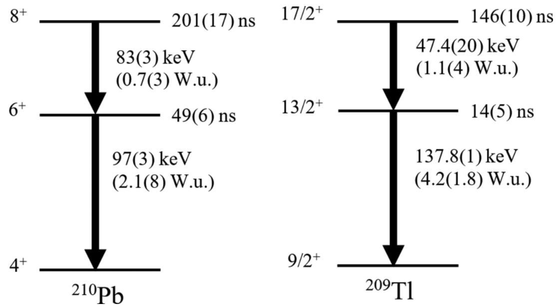

FIG. 5. Comparison between the spectroscopic properties of the isomers in ^{210}Pb and ^{209}Tl.

compared to 0.7(3) W.u. in ^{210}Pb [25] as illustrated in Fig. 5. Shell-model calculation were performed in Reference [11] and the reduced transition probability for this transition was calculated to be 0.96 W.u., which is in good agreement with our measurement and the 47-keV assignment. ^{209}Tl has slightly enhanced $B(E2)$ transition rates compared to ^{210}Pb, presumably due to the valence proton increasing the effective charge for transitions in ^{209}Tl.

VI. CONFIGURATIONS ABOVE THE ISOMER

Six prompt γ rays were found above the $J^{\pi} = 17/2^{+}$ isomer using the PDD matrix. Those are shown in Fig. 6. The intensities of these γ rays as well as the PPD matrix were used to produce the decay scheme shown in Fig. 7. Four of the γ rays we located above the isomer have previously been reported in a conference proceeding [29]. The side-feeding transitions of 250 and 516 keV helped fix the ordering of the main cascade, as they were not coincident with the highest decays. The structure of these states is anticipated to be due to the excitation of the neutrons to states above the $\nu g_{9/2}$ level and their coupling to the proton hole. The spectrum of excited neutron states has been experimentally observed in ^{211}Pb [23]. However, the coupling leads to a myriad of states and our current data on ^{209}Tl is too statistically marginal to allow any spin assignments which would allow the exact configurations to be inferred.

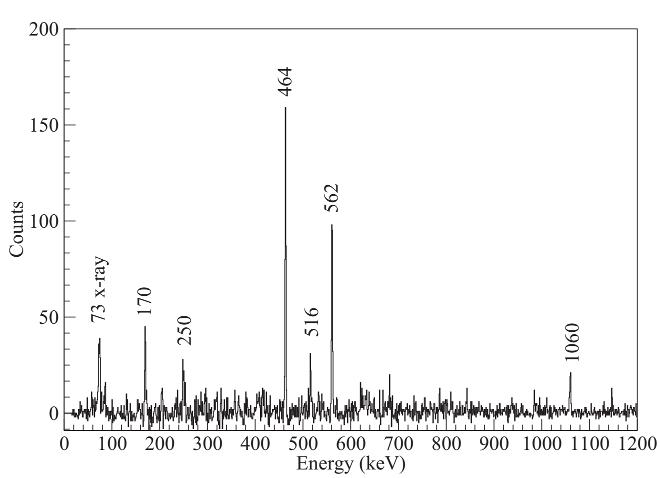

FIG. 6. Prompt γ-ray transitions obtained above the $17/2^{+}$ ^{209}Tl isomer, by gating on the 324- and 662-keV delayed transition in the prompt-delayed-delayed γ-γ-γ matrix.

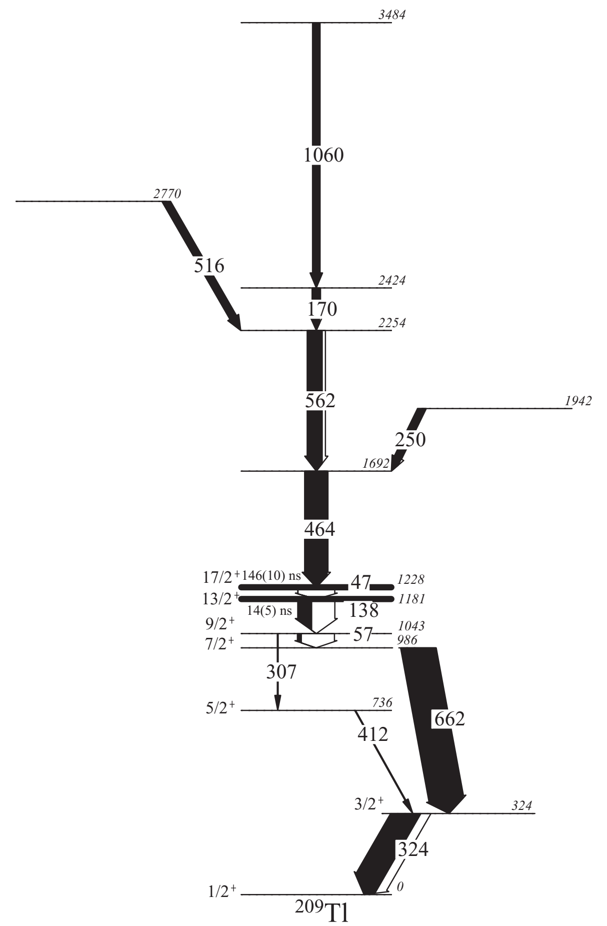

FIG. 7. ^{209}Tl Decay scheme for prompt and delayed transitions.

VII. CONCLUSIONS

^{209}Tl was populated about five times more intensely than a previous study [11]. The new data reveal several new decays which confirm the previous suggestions of spin and parity, as well as locate the isomer, its decays, and its structure. The longer isomer was remeasured and a second isomer was found in the decay chain. The $E2$ transition rates from the isomer are shown in Fig. 5 and are slightly enhanced when compared to ^{210}Pb, consistent with shell-model predictions. We have located a cascade of γ rays above the isomer. Although no angular momentum assignment could be made, analogy with ^{211}Pb would suggest the states arise from neutron excitations beyond the $2g_{9/2}$ orbit. However, more extensive data will be needed to understand their properties.

In all, the shell-model works very well for ^{209}Tl, at the 10s-keV level. This may be expected in a nuclide so close the ^{208}Pb core. The challenge remains to move further from the doubly magic core and find where it softens. This is particularly important along the $N = 126$ closed neutron shell, in nuclei like ^{205}Au and ^{203}Ir, and also in the $N = 128$ isotones, ^{207}Au and ^{205}Ir.

This multinucleon transfer approach appears viable for performing detailed spectroscopy in nuclei south and southeast of ^{208}Pb, although the actual production cross sections are lower than some of the theoretical predictions reported in Ref. [15] and decrease faster as one moves far from stability. For Xe+Pb, an increase of beam energy would undoubtedly increase the yield of ^{209}Tl and its neighbors, as production would be increased through using more of the target thickness. It is not clear if the production cross section has any strong energy dependence. Other combinations, like Hg+Pb or Hg+Hg, appear to be more appealing for producing exotic nuclides and more likely to have enhanced yields along $N = 126$, although the background from fission and quasifission will be considerably enhanced, which may impede spectroscopy of this type.

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