

New levels and a lifetime measurement in ^{204}Tl

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The $^{205}\text{Tl}(n, 2n\gamma)$ reaction was used to populate excited states in ^{204}Tl . The γ -ray detection was accomplished with the GEANIE spectrometer, a Compton suppressed array of 26 Ge detectors. An energetic beam of neutrons was provided by the pulsed neutron source of the Los Alamos Neutron Science Center's WNR facility. The time-of-flight technique was used to determine the incident neutron energies. γ -ray excitation functions were determined from incident neutron energy of 1 MeV up to $E_n = 25$ MeV. The level scheme of ^{204}Tl was enriched and the partial level scheme and nuclear structure above the previously known 7^+ isomer at 1104-keV excitation energy were established for the first time up to $E_x \sim 2.3$ MeV. The high-spin part of the level scheme exhibits striking similarities to that of the neighboring ^{202}Tl isotope, suggesting similarities in the underlying nuclear structure. The half-life of the 7^+ isomer was measured with a more precise result ($T_{1/2} = 60.7 \pm 1.2 \mu\text{s}$), in agreement with literature values. A lower limit for the excitation energy of the $\pi h_{11/2} \nu i_{13/2}$ structure with $J^\pi = 12^-$ is proposed.

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I. INTRODUCTION

The study of excited states in the odd-odd isotopes close to the doubly magic ^{208}Pb is important for comparison of experimental results with theoretical calculations in this mass region (see, for instance, Ref. [1] and references therein). Some theoretical calculations exist in the neighboring $^{206}\text{Tl}_{125}$ [1,2], but for $^{204}\text{Tl}_{123}$ no theoretical calculations have been reported so far. The study of ^{204}Tl represents an opportunity to obtain nuclear data, establish the systematics, and increase the understanding of nuclear structure in the ^{208}Pb region through comparison with model predictions.

The low-spin level scheme of the ^{204}Tl isotope has been previously studied [3], with information for low-spin states obtained mostly from $(n_{\text{thermal}}, \gamma)$ [4] and nucleon pickup reactions [5,6]. The low-lying states have negative parity and spin in the range 0 to $4\hbar$ with configurations based on the $3s_{1/2}$ and $2d_{3/2}$ proton orbitals and the $2f_{5/2}$, $3p_{1/2}$, and $3p_{3/2}$ neutron orbitals. The highest-spin state known is the 7^+ isomer at 1104-keV excitation energy [3], which is also the lowest positive-parity state and involves the neutron $1i_{13/2}$ orbital. At higher excitations, evidence for the existence of two isomers with $J^\pi = (12^-)$ and $J^\pi = (20^+)$ was obtained by detecting delayed transitions of the ^{204}Tl fragment in a projectile-fragmentation experiment [7], but the excitation energies of these states remain unknown.

The level scheme of ^{204}Tl [3] is expected to exhibit similarities with that of ^{202}Tl [8,9]. Indeed, both level schemes have a $J^\pi = 2^-$ ground state with a half-life of 3.78 years in ^{204}Tl and 12.23 days in ^{202}Tl . A 7^+ isomer is known in both isotopes originating from the $((\pi s_{1/2})^{-1}(\nu i_{13/2})^{-1})$ configuration, an interpretation supported by the measurement

of the g factor of this isomer in both ^{202}Tl [10] and ^{204}Tl [11] isotopes. The similarities extend also to lighter odd-odd Tl isotopes. A series of low-lying 7^+ isomeric states is known in all lighter odd-odd Tl isotopes [12] down to ^{194}Tl and $J^\pi = 2^-$ ground states are the case in all these isotopes. The reported half-lives of the 7^+ isomeric states vary from a few minutes in the lighter Tl isotopes to $591 \mu\text{s}$ in ^{202}Tl [9] and $63 \mu\text{s}$ in ^{204}Tl [3]. However, the higher-spin positive-parity structure above these 7^+ isomers is known only in ^{202}Tl [8] from the β decay of the 9^- isomer of ^{202}Pb [13] which strongly populates states in ^{202}Tl with $J = 8, 9$ (the term “high-spin” is used here to describe these states because such a spin is considered high if the states are populated in a neutron-induced reaction).

In the present work we have set out to obtain more spectroscopic information in ^{204}Tl which is needed to make comparisons with nuclear model predictions and fill in systematics. The results include the following: the level scheme in ^{204}Tl was enriched, the structure above the 7^+ isomer was established, a lower limit for the excitation energy of the 12^- isomer was proposed, and the half-life of the 7^+ isomer was remeasured with better accuracy. Comparison of the ^{204}Tl partial level scheme will be made with that of ^{202}Tl .

II. EXPERIMENT

The experiment was performed at the Los Alamos Neutron Science Center Weapons Neutron Research (LANSCE/WNR) facility [14]. The γ rays produced in the bombardment of a ^{205}Tl target by neutrons were measured with the GEANIE spectrometer [15].

GEANIE is located 20.34 m from the WNR spallation neutron source on the 60° -right flight path. The neutrons were produced in a $^{\text{nat}}\text{W}$ spallation target driven by an 800-MeV proton beam. The time structure consists of

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775- μ s-long “macropulses,” 16.7 ms apart, with each macropulse containing subnanosecond-wide “micropulses,” spaced every 1.8 μ s. The energy of the neutrons was determined using the time-of-flight technique. The neutron flux on target was measured with a fission chamber, consisting of ^{235}U and ^{238}U foils [16], located 18.48 m from the center of the spallation target. In the present experiment GEANIE was comprised of 11 Compton-suppressed planar Ge detectors (low-energy photon spectrometers, LEPS), 9 Compton-suppressed coaxial Ge detectors, and 6 unsuppressed coaxial Ge detectors. More information on GEANIE as well as a schematic diagram of the flight path and the beam time-structure can be found in Ref. [9].

A symmetrized, two-dimensional matrix was constructed to investigate the coincidence relationships between the γ rays and includes all (regardless of incident neutron energies) the beam-on γ - γ coincidence data obtained in the present experiment. Half-lives of the states with $T_{1/2}$ within a range of microseconds to tens of milliseconds were deduced from the beam-off data (data obtained between “macropulses”). Beam-off data are stored with times from a precision 100-ns clock based on a Stanford Research Systems Model SC10 (J grade) stabilized oscillator [17].

The ^{205}Tl sample was encased within a polystyrene disk, 2.3 cm in outer diameter and 0.5 cm thick, containing 2.2 g of Tl oxide powder, 97.0% enriched in ^{205}Tl and tightly encapsulated in the plastic capsule. The end faces of the disk were normal to the neutron beam.

III. EXPERIMENTAL RESULTS

The new levels and transitions assigned to ^{204}Tl in the present work are shown in the partial level scheme of Fig. 1. Thirteen new transitions were assigned to ^{204}Tl and placed in the level scheme. Also seen were the previously observed and placed [3] 139.9-, 146.0-, 414.1-, and 689.9-keV transitions. The 349.9- and 668.9-keV transitions were previously observed [7].

The excitation functions for all transitions in Fig. 1 were obtained except for the very weak 146.0- and 269.6-keV transitions and for the contaminated 668.9-keV transition.

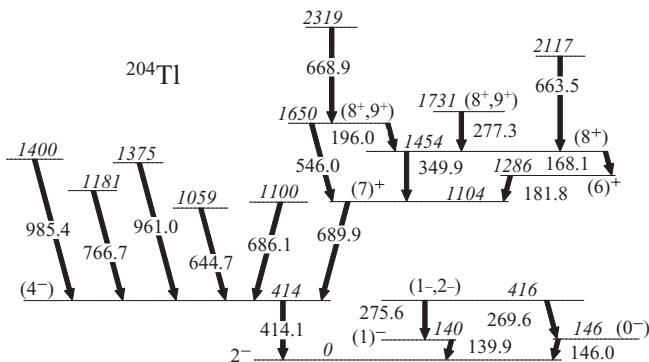


FIG. 1. Partial level scheme including all the new levels and transitions assigned to ^{204}Tl in the present work. Transition and excitation energies are given in keV.

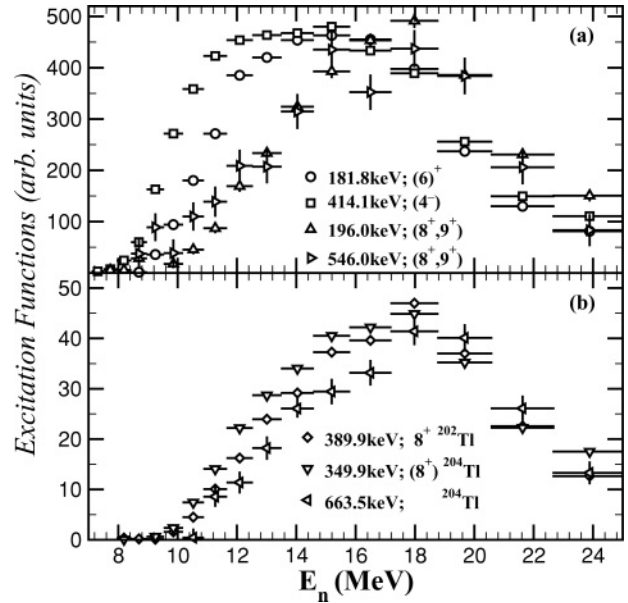


FIG. 2. Excitation functions (up to 25-MeV incident neutron energy) obtained in the present experiment for the (a) 181.8-, 196.0-, 414.1-, and 546.0-keV transitions and (b) 349.9- and 663.5-keV transitions (see Fig. 1). Additionally, in (b) the excitation function of the 349.9-keV transition is compared to that of the 389.9-keV transition of ^{202}Tl (from Fig. 7 in Ref. [9]).

Contamination of the 668.9-keV transition is due to transitions in the (n, n') and $(n, 3n)$ reaction channels in the present experiment. Examples of excitation functions obtained in the present experiment are shown in Fig. 2. The excitation functions peak between 10- and 20-MeV incident neutron energies, a characteristic of the $(n, 2n)$ reaction channel.

Evidence for the new transitions placed in the ^{204}Tl level scheme is based on the γ - γ coincidence data. The quality of the γ - γ coincidence data is shown in Fig. 3, where gates on the 349.9- and 414.1-keV transitions of Fig. 1 are shown. Correspondence for excitation functions obtained for the transitions in the present experiment to those obtained for the ^{202}Tl transitions in Ref. [9] was useful in placing the transitions that feed directly the 7^+ isomer in Fig. 1. An example is shown in Fig. 2(b), where the excitation function for the 349.9-keV transition (see Fig. 1) shows striking similarities to that obtained for the 389.9-keV transition of ^{202}Tl in the $^{203}\text{Tl}(n, 2n\gamma)$ reaction of Ref. [9], suggesting an (8^+) spin-parity assignment for the 1454-keV level in Fig. 1. Comparison with the level scheme of the neighboring ^{202}Tl isotope was also very helpful in assigning tentative spins and parities for some levels. Only the transitions for which the excitation functions matched the $(n, 2n)$ reaction threshold and shape were assigned to ^{204}Tl and placed in the level scheme. The peaks in the spectrum of Fig. 3 that are not labeled did not satisfy this condition and, hence, are deemed as contaminants. The levels in Fig. 1 are summarized in Table I.

One new low-lying level was observed in the present work, namely, the 416-keV level that deexcites in two branches via the 269.6- and 275.6-keV transitions. The existence in ^{204}Tl of the previously known 414.1-keV, 4^- level has been confirmed

TABLE I. Excitation energies, spin-parities, half-lives, and energies of the γ rays deexciting all levels in Fig. 1.

E_x^a (keV)	J^π	$T_{1/2}$	E_γ^a (keV)
0.0	2^-	3.78 y [3]	
139.9	(1^-)		139.9
146.0	(0^-)		146.0
414.1	(4^-)		414.1
415.5	$(1^-, 2^-)$		269.6, 275.6
1058.8			644.7
1100.2			686.1
1104.0	(7^+)	60.7(1.2) μs	689.9
1180.8			766.7
1285.8	(6^+)		181.8
1375.1			961.0
1399.5			985.4
1453.9	(8^+)		168.1, 349.9
1649.9	$(8^+, 9^+)$		196.0, 546.0
1731.2	$(8^+, 9^+)$		277.3
2117.4			663.5
2318.8			668.9

^aThe uncertainty on the γ -ray and excitation energies varies from 0.2 to 0.5 keV.

in the present experiment. This level was used as a base level to build up the rest of the higher-spin level scheme in Fig. 1. The excitation function obtained for the 414.1-keV transition is shown in Fig. 2(a). Five new levels at 1059-, 1100-, 1181-, 1375-, and 1400-keV excitation energy were observed to feed directly the 414-keV, 4^- level (see Fig. 1). Spins of $\sim 4\hbar$ are supported for these levels by the excitation functions of the transitions that deexcite these states, all of which were found to be similar to that of the 414.1-keV transition in Fig. 2(a). If

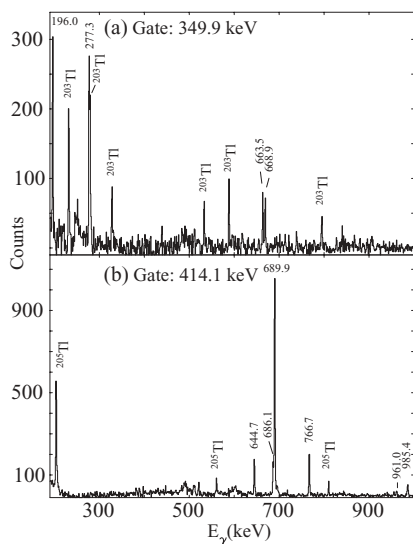


FIG. 3. Spectra gated on the (a) 349.9-keV and (b) 414.1-keV transitions of ^{204}Tl in Fig. 1. Transition energies are in keV. The presence of ^{203}Tl transitions in (a) is attributed to their contribution in the gate of the 350.2-keV transition of ^{203}Tl [18,19]. The presence of ^{205}Tl transitions in (b) is attributed to their contribution in the gate of the 415.7-keV transition of ^{205}Tl [20].

the 1375-keV level in Fig. 1 is the same as the previously reported [3] 1375.0(11)-keV, $(0, 1, 2)^-$ level, then a $(2)^-$ spin-parity is most likely for this level because the 961.0-keV transition establishes a decay path toward the 414-keV, (4^-) level besides the previously known decay path toward the 140-keV, $(1)^-$ level via the 1235-keV transition [3] (a transition for which the existence could not be confirmed in the present experiment). The 181.8-keV transition deexcites the 1286-keV level in Fig. 1 and its excitation function is shown in Fig. 2(a). The 1286-keV level was assigned as $(6)^+$ based on the previous observation of a 1289(6)-keV level in (d, t) , (p, d) reactions with $L = 6$ [3]. The excitation functions for the two transitions (196.0- and 546.0-keV) deexciting the 1650-keV level in Fig. 1 are included in Fig. 2(a) and exhibit striking similarities (within uncertainties) as is expected for transitions that deexcite the same level. Two more levels at 2117- and 2319-keV excitation energies were added to the structure above the 7^+ isomer and are the levels highest in excitation observed in the present work. The excitation function for the 663.5-keV transition in Fig. 2(b) that deexcites the 2117-keV level has an increase with neutron energy less steep than that of the 349.9-keV transition in Fig. 2(b), supporting a higher spin assignment ($> 8\hbar$) for this level. For the 668.9-keV transition that deexcites the 2319-keV level, an excitation function was not obtained from the present data because of contamination. However, this transition can be clearly seen in the spectrum of Fig. 3(a) and, moreover, was previously seen (together with the 349.9-keV transition of Fig. 1) in coincidence with ^{204}Tl fragments (see lower spectrum in Fig. 3 of Ref. [7]) in a projectile-fragmentation experiment.

The half-life of the previously known 7^+ isomer at 1104-keV excitation energy (see Fig. 1) was remeasured in the present work, taking advantage of the “beam-off” data (data obtained between macropulses). The 1104-keV level deexcites by the 689.9-keV E3 transition to the 4^- level, which in turn deexcites to the ground state via the 414.1-keV transition in Fig. 1. The decay curves of both these transitions were obtained in the beam-off data and are shown in Fig. 4. A $60.7 \pm 1.2 \mu\text{s}$ half-life is obtained from the decay curve of the 689.9-keV transition in Fig. 4(b). For the 414.1-keV transition a $61.4 \pm 2.4 \mu\text{s}$ half-life was obtained in Fig. 4(a), a value that is slightly larger than that for the 689.9-keV transition but within uncertainties. However, the 414-keV level in Fig. 1 might be fed by other isomers that can affect the half-life result for this transition obtained from the fitting in Fig. 4(a). There is some evidence for higher-lying isomers in this isotope with shorter lifetimes (12^- and 20^+ isomers with 2.6(2) μs and 1.6(2) μs half-life, respectively; see Ref. [7]), while the existence of other unknown isomers cannot be ruled out. Hence, only the half-life obtained for the 689.9-keV transition that directly deexcites the isomer is adopted here as the lifetime of the 7^+ state: i.e., $T_{1/2}(7^+) = 60.7 \pm 1.2 \mu\text{s}$. By obtaining the decay curves of transitions from known long-lived isomers, it was determined that a dead-time correction is necessary for the first $\sim 300 \mu\text{s}$ of the macropulse and such a correction was applied to the counts shown in Fig. 4.

In the projectile-fragmentation experiment of Ref. [7] the 349.9- and 668.9-keV transitions were assigned to the ^{204}Tl fragment, but not placed in the level scheme, and are reported

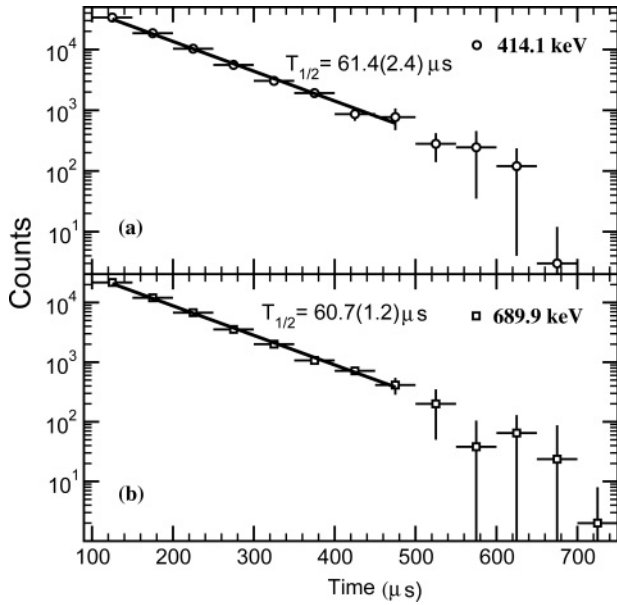


FIG. 4. Decay curves obtained from the beam-off data in the present experiment for (a) the 414.1-keV and (b) the 689.9-keV transitions (see Fig. 1). The half-life deduced from the fitting of the curves is shown in both cases. Points with less than 300 counts (later than 500 μ s in time) were not included in the fitting because of increased uncertainties in reliably fitting a well-defined Gaussian peak in the γ -ray spectra. The peaks for both transitions diminish below detection ability in the γ -ray spectra after $\sim 700 \mu$ s in time, i.e., after ~ 11 half-lives of the 7^+ isomer.

as part of the decay path of a suggested (12^-) isomer with 2.6(2)- μ s half-life. The placement in the present work of both transitions in the level scheme above the 7^+ isomer, with the 668.9-keV transition decaying from the level at 2319-keV excitation energy, suggests that this excitation energy is a lower limit for the excitation energy of the (12^-) isomer, i.e., $E_x(12^-) > 2319$ keV.

IV. DISCUSSION

The half-life value of $60.7 \pm 1.2 \mu$ s obtained in the present work is more precise than previous reports, and it agrees within uncertainties with the 63(2) μ s value adopted in the literature

[3] for the 7^+ isomer in ^{204}Tl . This latter value is the result of evaluation from previous measurements of the half-life of this isomer, namely, the 62(5) μ s value reported in Ref. [21], the 63(2) μ s value in Ref. [22], and the 65(3) μ s value in Ref. [23]. The present result, with its smaller statistical uncertainties, is in agreement, within uncertainties, with two of the three previous measurements. The uncertainty weighted mean of the three previous measurements and the present one gives a half-life value of $61.7 \pm 1.0 \mu$ s.

There exist similarities in E_x , J^π , and γ -decay patterns of ^{204}Tl levels to the levels reported in ^{202}Tl [8,9]. In both isotopes the low-lying negative-parity states are well known [3,8,9] with configurations based on the coupling of the $3s_{1/2}$ and $2d_{3/2}$ proton orbitals to the $2f_{5/2}$, $3p_{1/2}$, and $3p_{3/2}$ neutron orbitals. Moreover, for the newly observed low-lying 416-keV level, a very similar level is known in ^{202}Tl at 402-keV excitation energy [9]. The 490.47-keV level [8] of ^{202}Tl is the first configuration that involves a proton hole in the $d_{3/2}$ orbital, namely, $((\pi d_{3/2})^{-1}(\nu f_{5/2})^{-1})$, and is the corresponding level to the 414.1-keV level in Fig. 1, whereas similar levels were observed to feed both levels in the present work and in Ref. [9]. Above the 7^+ isomer, the 1286-keV level in Fig. 1 is the corresponding level to the ^{202}Tl level at 1099-keV excitation energy [8,9] and both levels originate from the same configuration as the lower-lying 7^+ isomers, namely, $((\pi s_{1/2})^{-1}(\nu i_{13/2})^{-1})$. From the coupling of the proton hole in the $d_{3/2}$ and $d_{5/2}$ orbitals to the $i_{13/2}$ neutron hole, three additional states with $J = 8^+$ and 9^+ were reported in ^{202}Tl [8,9] at 1340-, 1552-, and 1676-keV excitation energies; the corresponding states were also observed in the present experiment in ^{204}Tl at 1454-, 1650-, and 1731-keV excitation energies. This is supported by the comparison of the corresponding excitation functions obtained for the transitions in the present experiment with those obtained for the ^{202}Tl transitions in Ref. [9]. Hence, the high-spin structure above the 7^+ isomer in both ^{202}Tl and ^{204}Tl is very similar at least up to ~ 1.7 -MeV excitation energies.

At even higher excitation energies the existence of the $\pi h_{11/2}\nu i_{13/2}$ and $\pi h_{11/2}\nu i_{13/2}^2 f_{5/2}$ configurations has been tentatively proposed [7] and deemed responsible for the (12^-) and (20^+) isomers, respectively. The excitation energies of these isomers remain unknown, with the 20^+ isomer expected to lie at a higher excitation energy than the 12^- isomer.

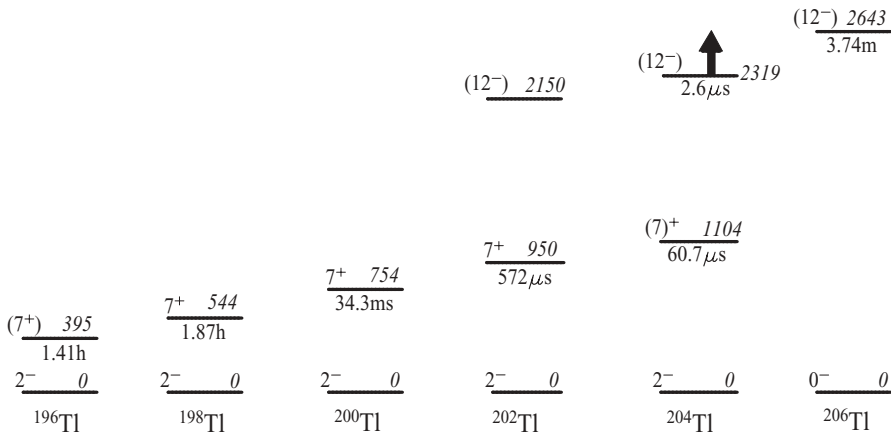


FIG. 5. Systematics of the 7^+ isomers and the 12^- levels in the odd-odd $^{196-206}\text{Tl}$ isotopes. The excitation energy of the 12^- isomer in ^{204}Tl is only a lower limit and this is represented by the upward pointing arrow. Data are taken from Refs. [3,8,24–27] and the present work.

From the combination of the results of the present experiment with the results of the projectile-fragmentation experiment of Ref. [7] only a lower limit for the excitation energy of the (12^-) level can be obtained; hence, a lower limit for the excitation energy of the $\pi h_{11/2} \nu i_{13/2}$ configuration can be suggested at 2319 keV. The systematics for the (12^-) levels and the 7^+ isomers in odd-odd Tl isotopes from ^{196}Tl to ^{206}Tl is shown in Fig. 5. The excitation energy of the 7^+ isomers decreases as one moves away from the closed neutron shell. The same behavior is observed in the excitation energies of the (12^-) levels in ^{202}Tl and ^{206}Tl (~ 2150 keV [8] and 2643 keV [24], respectively). Hence, an upper limit for the excitation energy of the (12^-) level in ^{204}Tl can be also suggested, only tentatively, based on the systematics of Fig. 5, i.e., $2319 \text{ keV} < E_x(12^-) < 2643 \text{ keV}$.

V. SUMMARY

In summary, states of ^{204}Tl with up to ~ 2.3 MeV excitation energy were studied using the $^{205}\text{Tl}(n, 2n\gamma)$ reaction. The data were taken using the GEANIE spectrometer and the neutron beam of the Los Alamos Neutron Science Center's WNR facility. The deduced γ -ray excitation functions combined

with γ - γ coincidence information were used to enrich the higher-spin part of the level scheme. The nuclear structure and partial level scheme above the previously known 7^+ isomer at 1104-keV excitation energy were established and the half-life of this isomer was remeasured. The level scheme of ^{204}Tl exhibits similarities to that of the neighboring ^{202}Tl isotope up to at least ~ 1.7 -MeV excitation energy, a fact that is expected based on the similar underlying nuclear structure in both isotopes. At higher excitation energies, a lower limit for the excitation energy of the $\pi h_{11/2} \nu i_{13/2}$ configuration was deduced.

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