Title

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^{176}Lu: An Unreliable s-Process Chronometer

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A level scheme of ^{176}Lu up to ~1400 keV excitation energy is deduced from a gamma-gamma coincidence experiment and previously published particle transfer data. 170 gamma-ray transitions are placed between 85 levels, confirming many of the previously established levels and some of the decay scheme. A level at 838.5 keV ($J^{\pi}=5^{-}$, $t_{1/2} < 10$ ns) decays with substantial strength to both the ground state (7^{-}, 4.08×10^{10} yr) and the 122.9 keV isomer (1^{-}, 3.7 hr). The presence of this level guarantees the thermal equilibration of ^{176}Lu^{g,m} for temperatures greater than 3×10^{8} K and therefore during s-process nucleosynthesis. The resulting extreme temperature sensitivity of its effective half-life rules out the use of ^{176}Lu as an s-process chronometer. The use of ^{176}Lu to determine s-process temperatures is discussed.

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Introduction

^{176}Lu is one of the few naturally occurring radio-nuclides that have survived from the era of nucleosynthesis. It's present isotopic abundance[1] is 2.6% and its half-life is 4.08 x 10^{10} yr[2]. The spectrum of gamma rays from the ground-state decay of ^{176}Lu nuclei in a foil of natural lutetium observed by a 1.3 cm thick planar germanium detector is shown in Figure 1.

As shown in Fig. 2, ^{176}Lu can be produced only via the slow neutron capture process (s process). The stable isobars ^{176}Yb and ^{176}Hf shield this nucleus from rapid neutron capture and proton capture contributions. The s-process production path in the vicinity of ^{176}Lu is also indicated in the figure. Due to the the long half-life of the ground state, ^{176}Lu^{g}, it was suggested that ^{176}Lu would be a candidate for a s-process chronometer[3,4]. However, there exists a much shorter lived isomer at 122.9 keV ($J^{\pi}=1^{-}, t_{1/2}$ = 3.7 hr)[1]. As Figure 3 shows, the large spin difference between these two levels prevents decays from the isomer to the ground state; rather the isomer β decays to ^{176}Hf. The presence of this isomer could affect the decay of ^{176}Lu in astrophysical environments, providing a method of communication exists between the two levels. An example of this communication is illustrated in Figure 3 where an additional level of intermediate spin is populated and subsequently decays to both the ground state and the isomer. The time scale for obtaining equilibration between the isomer and g.s. is determined by the rate of excitation of the mediating level, its spin, parity and excitation energy, and its decay properties, as well as the half-lives of the g.s. and isomer. In the stellar environment where the s process occurs, nuclei are believed to be subjected to temperatures of the order of a few x 10^{8} K. At these temperatures one can expect that the tails of the thermal distribution should populate levels up to ~ 1 MeV.

The presence of such an equilibration path would severely compromise the usefulness of ^{176}Lu as an s-process chronometer due to the effective decay constant, λ_{eff}, being temperature sensitive. Such a mediating level lying between 662 keV and 1332 keV excitation energy can be inferred from the photoexcitation work of Norman *et al.*[5] In these experiments, ^{176}Lu^{m} activity was observed following the irradiation of a ^{nat}Lu foil with ^{60}Co γ-rays, but not following irradiation with a ^{137}Cs source. Our aim in this experiment was, therefore, to determine the level scheme of ^{176}Lu up to approximately 1 MeV to search for levels which could serve as a mediating level between the ground state and isomer. We pursued this goal using the method of coincident gamma ray detection. Concurrently and independently another group pursued a different technique to establish the level scheme, obtaining similar results and identical conclusions[6,7].

Experiment

We used the ^{176}Yb(p,n)^{176}Lu reaction to populate levels in ^{176}Lu. An 8-MeV proton beam was provided by Lawrence Berkeley Laboratory's 88-Inch Cyclotron. The beam energy was chosen to maximize the yield of ^{176}Lu while limiting other reaction products. The target was a 2 mg/cm^2 metallic foil enriched to 97.04% ^{176}Yb. Data collection was count-rate limited and required that beam currents were kept below 10 na. Coincident gamma-ray and gamma-ray singles events were detected by the High Energy Resolution Array of 21 Compton-suppressed germanium detectors. The detector at zero degrees to the beam was removed to install a shielded external beam dump for the unscattered protons. Approximately 60 million coincident events were recorded. These events were then sorted off-line into a two dimensional matrix. Detector resolution was found to be 2.32 keV FWHM at 838.5 keV, with no significant decrease in resolution in the sum spectrum as compared to that of a single detector. Detector energy and efficiency calibrations were performed with standard sources placed at the target position. In addition to the energy signals, we generated timing information signals (TAC) between detectors. A subset of ten of the detectors were designated as start detectors and the stop signal was generated by a coincident event in any of the other ten detectors. The hardware gate of 100 ns established the maximum time difference between coincident events. The resolution of the TAC was ~10 ns.

Analysis and Results

Gates were placed on ~400 of the strongest transitions and coincidence relationships were established in the background subtracted gated spectra. Using these data, the previously established level scheme, and ^{176}Lu levels established with particle transfer experiments, we constructed the level scheme shown in Figs. 4-10. In these figures, the levels and decay transitions have been grouped into band structures. These groupings are supported by data in the literature and by the work of References 6 and 7. Those levels and transitions which did not fit into known bands are presented in Figures 8 and 9. In all, we have proposed 170 transitions between 85 levels. We have emphasized the transitions which feed and decay from the 838.5 keV level in Figure 10.

Many of these proposed placements confirm previous work. All proposed levels were checked for self-consistency with parallel and sequential decays and for γ-ray decay intensities. The relative intensities of all transitions from each level were confirmed to be independent of which populating transition was gated on. No attempt was made to determine the spins and parities of the transitions from the intrinsic angular distribution data, rather data from the literature[4, 6-15] were used to assign the spins and parities suggested in Figs. 4 through 10.

As can be seen in Figure 10, where we have highlighted the transitions from and to the level at 838.5 keV, this level decays with significant decay strengths to both the ground state and to the isomer. From the transitions shown in the figure we can infer the (J,π) of this level as being their 5^- or 6^-. This assignment agrees with the assignments of 5^- suggested in the literature and with the assignment deduced in References 6 and 7. For the specific transitions originating from this level we have measured the decay strengths, corrected for detector efficiency, but not for internal conversion. These are presented in Table I. The errors are estimates of only the statistical errors involved in extracting the peak areas. In addition, to corroborate the decays to and from the level at 838.5 keV, we generated the TAC spectra for all combinations of start detectors feeding and stop detectors decaying from this level. These TAC spectra all showed the same time relationships between all combinations of the feeding and exiting γ rays which increases our confidence in their placement.

From the placement of this level and its inferred spin we can calculate the photoexcitation rate as a function of temperature using the expression[16]:

$$\frac{\tau\,(I \Rightarrow I*)}{\tau_{sp}\,(I * \Rightarrow I)} = \frac{2J_I + 1}{2J_{I*} + 1}\left\{\exp\left(\frac{\Delta E}{kT}\right) - 1\right\} \qquad (1)$$

τ_{sp} is the spontaneous decay rate, J_I and J_{I*} are the spins of the states I and I* and ΔE is the energy difference between these two states. From our TAC data we could place an upper limit on the spontaneous decay lifetimes of the potentially mediating levels of $\tau_{sp} \leq 10$ ns, which is consistent with the observed resolution. The single particle Weisskopf estimate for the rates of these decays are substantially faster than this limit. Using the theoretical estimates for τ_{sp} we present the three curves in Fig. 11 corresponding to the population of the 838.5, 722.9, and 563.9 keV levels from the ground state. Assuming that the photoexcitation time is short in comparison to the mean-life of the isomer establishes the criterium for thermal equilibrium. We see in Fig. 11 that for temperatures greater than 3×10^8 K, the isomer and ground state will be in thermal equilibrium using the 838.5 keV level as the mediating level. From Figure 4., we might also expect that the band head at 722.9 keV would serve as a mediating level. Calculating the single particle transition strength for this level we find that the direct decay from the 722.9 to the ground state would be only ~3% of the 838.5 decay strength. Consequently, it is possible that we would not directly observe this decay with our coincident gamma-ray technique. However, even a 1% branch to the ground state would be adequate to equilibrate the ground state and isomer via this level. Evaluating eq. 1 assuming the moderating level is at 722.9 keV rather than 838.5 keV yields an estimate of the equilibration temperature of 2.6×10^8 K. A more careful examination of the level scheme yields several other levels which could act as mediating levels, the lowest one being at 563.9 keV. This level results in equilibration being reached at 2×10^8 K.

The resulting effective half-life of ^{176}Lu is an extremely sensitive function of the temperature. The effective beta-decay rate, λ_{eff}, for the nucleus is given by:

$$\lambda_{eff} = \frac{\sum_i g_i \lambda_i \exp(-E_i / kT)}{\sum_i g_i \exp(-E_i / kT)} , \qquad (2)$$

where $g_i = (2J_i + 1)$, λ_i is the beta decay rate of the state i, E_i is the excitation energy of state i, and the summation is extended over all states that are in thermal equilibrium. Assuming that none of the other states are involved besides the ground state and the isomer (we would not expect any of the other levels to have drastically larger β-decay rates) this expression simplifies to:

$$\lambda_{eff}(^{176}\mathrm{Lu}) = \frac{2.54 \times 10^{-10} + 4.93 \times 10^{3} \exp(-14.26/T_8)}{15 + 3 \exp(-14.26/T_8)} \ (\mathrm{year}^{-1}) , \qquad (3)$$

where T_8 is the temperature in units of 10^8 K. We have tabulated the solutions of this equation for the temperature range of $0<T_8<5$ in Table II. We compare our estimates to those of References 17 and 18 which differ from ours in several respects. Cosner and Truran[17] assumed that all levels up to 300 keV contribute to the decay rate. However, Cosner and Truran and Takahashi and Yokoi[18] both used an incorrect value for the isomer energy which was in the literature (127 keV as opposed to the value we report of 122.9 keV). Table II vividly illustrates how a relatively small change in s-process temperature can result in a major change in the decay constant for ^{176}Lu. This strong temperature sensitivity quite effectively rules out ^{176}Lu for use as an cosmochronometer.

A second analysis of the A=176 system is based on the formalism of Schramm and Wasserberg[19]. In this analysis the mean duration of nucleosynthesis, Δ_{max}, can be expressed as:

$$\Delta_{max} = \frac{1}{\lambda_{^{176}\mathrm{Lu}}} \ln \left[\frac{B \langle N_s \sigma \rangle_{176}}{N_{^{176}\mathrm{Lu}} \langle \sigma_{^{176}\mathrm{Lu}} \rangle} \right] \qquad (4)$$

where $\lambda_{^{176}\mathrm{Lu}}$ is the decay constant of ^{176}Lu, $\langle N_s \sigma \rangle_{176}$ is the product of the s-process abundance and the Maxwellian averaged neutron capture cross section evaluated at mass 176 and at s-process temperatures (T~23 keV). $N_{^{176}Lu}$ is the present day abundance of ^{176}Lu and $<\sigma_{^{176}Lu}>$ is the ~23 keV ^{176}Lu(n,γ)^{177}Lu cross section. Finally, B is the branching ratio for the formation of the ground state in the ^{175}Lu(n,γ)^{176}Lu reaction. The determination of B has been the subject of much experimental work in recent years[20-27], and the exact determination of the isomer and ground state capture cross sections critically affect the deduced parameter Δ_{max}. If we use the most recent values to evaluate the expression, presented in Table III, we find for that the argument of the logarithm in equation 4 is less than unity, which results in a negative value for the mean age. This can be interpreted as implying that there exists more ^{176}Lu today than that estimated from the systematics of the s process. This excess of Lu could be explained by a readjustment of the isomer and ground state fractions formed in the neutron capture reaction. By equilibrating the population, additional long lived ground state nuclei would be created, explaining the present-day "surplus"of ^{176}Lu.

The final topic we wish to address in this work is that of the use of ^{176}Lu as a stellar thermometer. In several other works[25,26] it has been suggested that if ^{176}Lu is in thermal equilibrium in stellar environments, then it would be possible to use the observed abundance of ^{176}Lu to deduce the stellar temperatures of the s process. However, to obtain the temperature profile during the s process will require a model dependent analysis. Under the assumptions that the s-process neutron density and temperature are uniform, one can more easily extra limits on the s-process temperature. Klay et al.[6,7] have done this and obtain results consistent with other determinations of the s-process temperature.

In conclusion, we have established the level scheme of ^{176}Lu and have placed 170 transitions between 85 levels. We have identified a specific level at 838.5 keV which decays to both the ground state and to the isomer. This level can then serve as an equilibration path between the two levels, and through photo-excitation alone guarantee that the two levels are in equilibrium for temperatures > 3×10^8 K. In addition to photo-excitation, the processes of Coulomb excitation, inelastic neutron scattering, and positron annihilation excitation will also contribute to the equilibration of the two levels and will reduce the temperature where the two levels achieve equilibration. Also, we would expect that the levels at 722.9 and 563.9 might serve as mediating levels and would significantly reduce the equilibration temperature. This equilibration of the ground state and isomer rules out the use of ^{176}Lu as an s-process chronometer. The extreme temperature sensitivity of the effective half life of ^{176}Lu also complicates efforts to deduce the s-process temperature profile.

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Figure 1. The ^{176}Lu^{g} decay spectrum observed from a sample of ^{nat}Lu. The principal peaks in ^{176}Lu are labelled by their energy in units of keV.

Figure 2. The s-process path in the vicinity of ^{176}Lu. The stability of ^{176}Yb and ^{176}Hf guarantees that ^{176}Lu can only be produced in the s process.

Figure 3. A partial level scheme of ^{176}Lu, showing the positions and decays of the ground state and isomer at 122.9 keV. The equilibration of the these two levels could be achieved by way of a level of intermediate spin, as illustrated in the figure.

Figure 4. The proposed level scheme of ^{176}Lu. We present the levels in band structures where possible, relying on earlier experiments to assign band heads and members to specific bands. The decay energies and the level energies are given in keV. The spins and parities are obtained from the literature. In this and the following six figures, those levels which are contained within the decay band are shown with solid lines, whereas the decays to levels in other bands (also seen in this experiment) are shown with dashed lines. In a) the band based upon p1/2^{+}[411] - n9/2^{+}[624] is presented, b) p1/2^{+}[411] - n7/2^{-}[514], c) p1/2^{+}[411] + n7/2^{-}[514], d) p1/2^{-}[541] + n7/2^{-}[514], e) p1/2^{-}[541] - n7/2^{-}[514], f) p5/2^{+}[402] + n9/2^{+}[624], and g) p5/2^{+} [402] + n7/2^{-} [514].

Figure 5. Additional band structures for ^{176}Lu. In a) the band based on p5/2^{+}[402] - n7/2^{-}[514] is presented and b) p7/2^{+}[404] - n3/2^{-}[512], c)

$p5/2^+[402] - n9/2^+[624]$, d) $p7/2^+[404] - n1/2^-[521]$, e) $p7/2^+[404] + n3/2^-[512]$, f) $p7/2^+[404] + n1/2^-[521]$, and g) $p7/2^+[404] + n5/2^-[512]$.

Figure 6. Additional band structures for ^{176}Lu. In a) the band based on $p7/2^+[404] - n1/2^-[510]$ is presented, b) $p7/2^+[404] + n1/2^-[510]$, c) $p7/2^+[404] + n9/2^+[624]$, d) $p7/2^+[404] - n5/2^-[512]$, and e) $p7/2^+[404] - n9/2^+[624]$.

Figure 7. Additional band structures for ^{176}Lu. In a) the band based on $p7/2^+[404] - n7/2^-[514]$ is presented, b) $p7/2^+[404] + n7/2^-[514]$, c) is a gamma-vibrational band, d) $p9/2^-[514] - n9/2^+[624]$, and e) $p9/2^-[514] - n7/2^-[514]$.

Figure 8. Additional band structures for ^{176}Lu. In a) the band based on $p9/2^-[514] - n1/2^-[510]$ is presented, b) $p9/2^-[514] + n1/2^-[510]$. The decays presented in c), d), e), f) and g) are not assigned to bands. The spins are parity are taken from the literature.

Figure 9. Additional transitions in ^{176}Lu. The scheme presented has approximate values of spin based upon the decay properties of the levels.

Figure 10. A subset of the proposed level scheme emphasizing the transitions originating from and populating the level at 838.5 keV.

Figure 11. The population time of the mediating level at 838.5 keV as a function of temperature for the ground state (solid curve). Assuming that the populating time is short (i.e. 1/10) in comparison to the meanlife (horizontal solid line) of the isomer establishes the

temperatures where the isomer and ground state will be in thermal equilibrium. Analogous curves for possible mediating levels at 722.9 (dashed) and 563.9 keV (dash-dot) are also presented.

Table I. The decay strengths for the five transitions from the level at 838.5 keV. These strengths have been corrected for the detector efficiency, but have not been corrected for internal conversion corrections. The errors shown in the third column are statistical (one sigma) errors

TABLE I

E_γ	I_γ	σ_{I_γ}
115.7 keV	3.1 %	± 0.5 %
181.2	4.9	± 0.4
203.5	7.9	± 0.5
274.6	13.9	± 0.8
838.3	70.2	± 2.9

Table II. Estimated effective β- decay half-lives for ^{176}Lu for a variety of temperatures between 0 and 5 x10^8 K.

TABLE II

Temperature (10^8 K)	$t_{1/2}(^{176}\mathrm{Lu})$ (years)		
	Ref 17	Ref 18	This work
0	-	-	4.08 x 10^{10}
1	5.070 x 10^3	5.28 x10^3	3.29 x 10^3
1.5	-	-	28.3
2	2.66	3.17	2.63
2.5	-	-	0.634
3	0.143	0.230	0.245
3.5	-	-	0.125
4	0.026	0.053	0.0750
4.5	-	-	0.0506
5	0.0082	0.019	0.0370

Table III. The various parameters needed to calculate Δ_{max} using Equation 4.

Table III.

$\lambda_{^{176}Lu}$	=	2.45×10^{-11} yr^{-1}	Ref. 2
$<N_s\sigma>_{176}$	=	8 mb (Si=10^6)	Ref 28
$N_{^{176}Lu}$	=	0.001035 (Si=10^6)	Ref 29
$<\sigma_{^{176}Lu}>$	=	1537 mb	Ref 30
B	=	0.11 ± 0.04	Ref 31
(based on σ_{tot}=1203±10 mb, σ_{iso}=1036±2),			Ref 30

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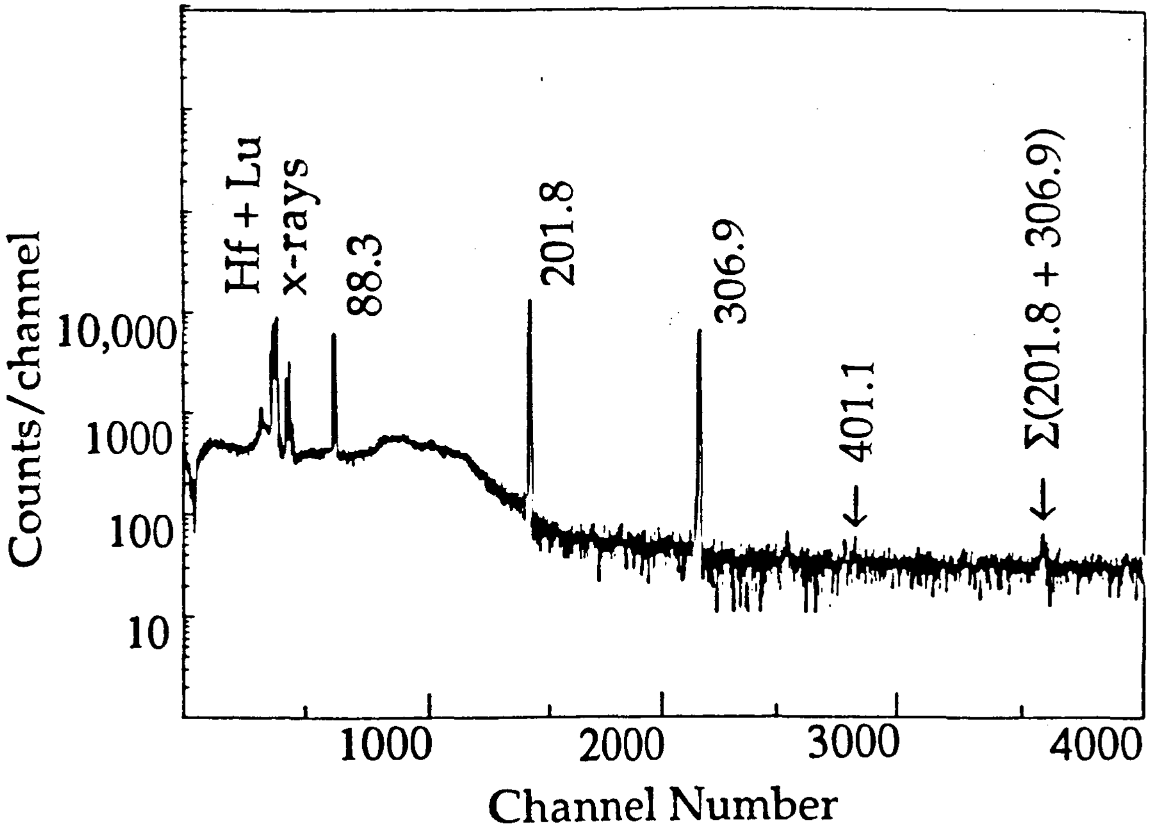

XBL 905-1757

Figure 1

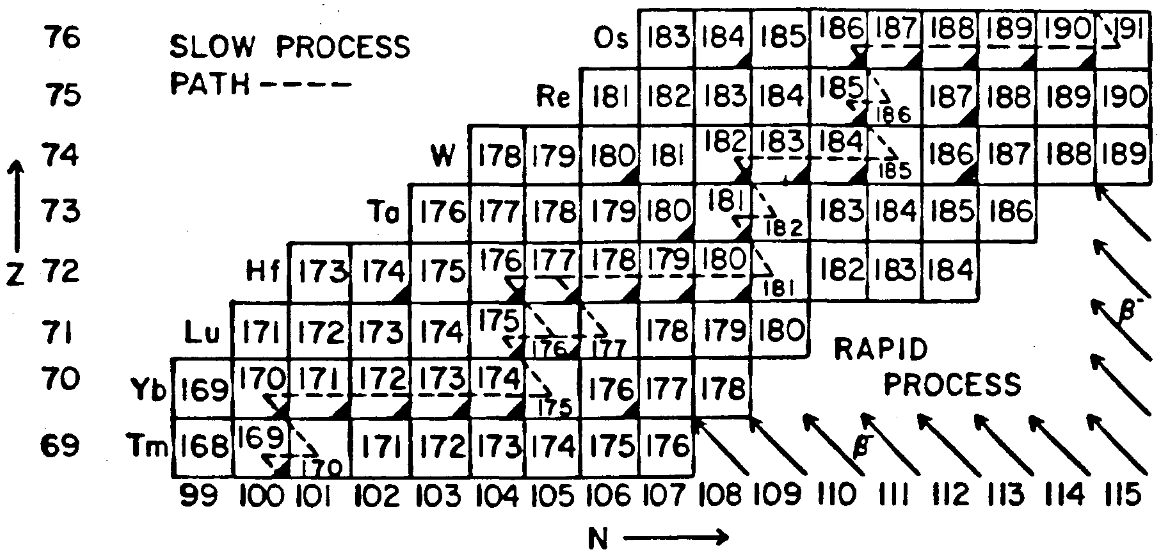

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Figure 2

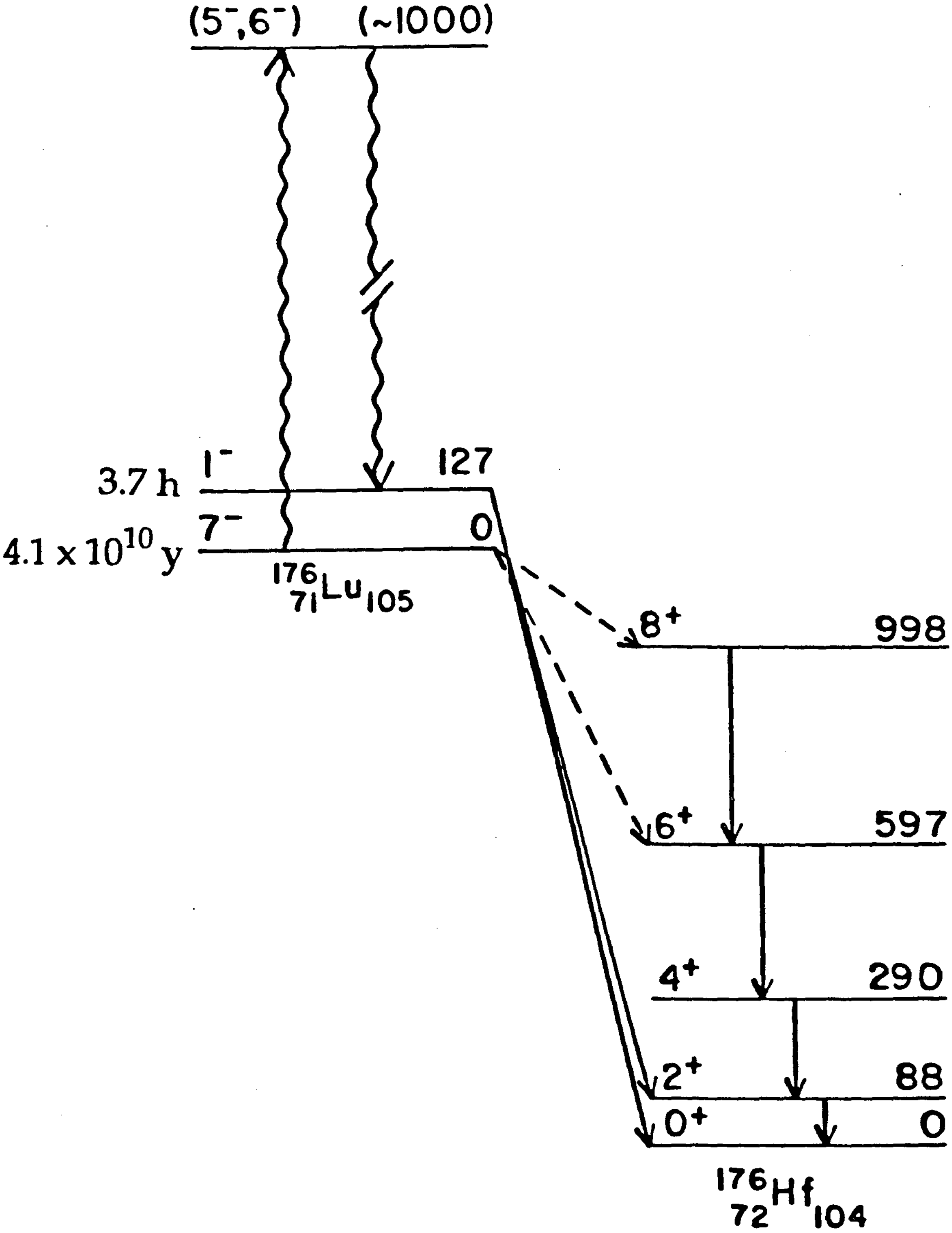
(5⁻,6⁻)
(~1000)
3.7 h
1⁻
127
7⁻
0
4.1 x 10¹⁰ y
¹⁷⁶₇₁Lu₁₀₅
8⁺
998
6⁺
597
4⁺
290
2⁺
88
0⁺
0
¹⁷⁶₇₂Hf₁₀₄
XBL 892-628

Figure 3

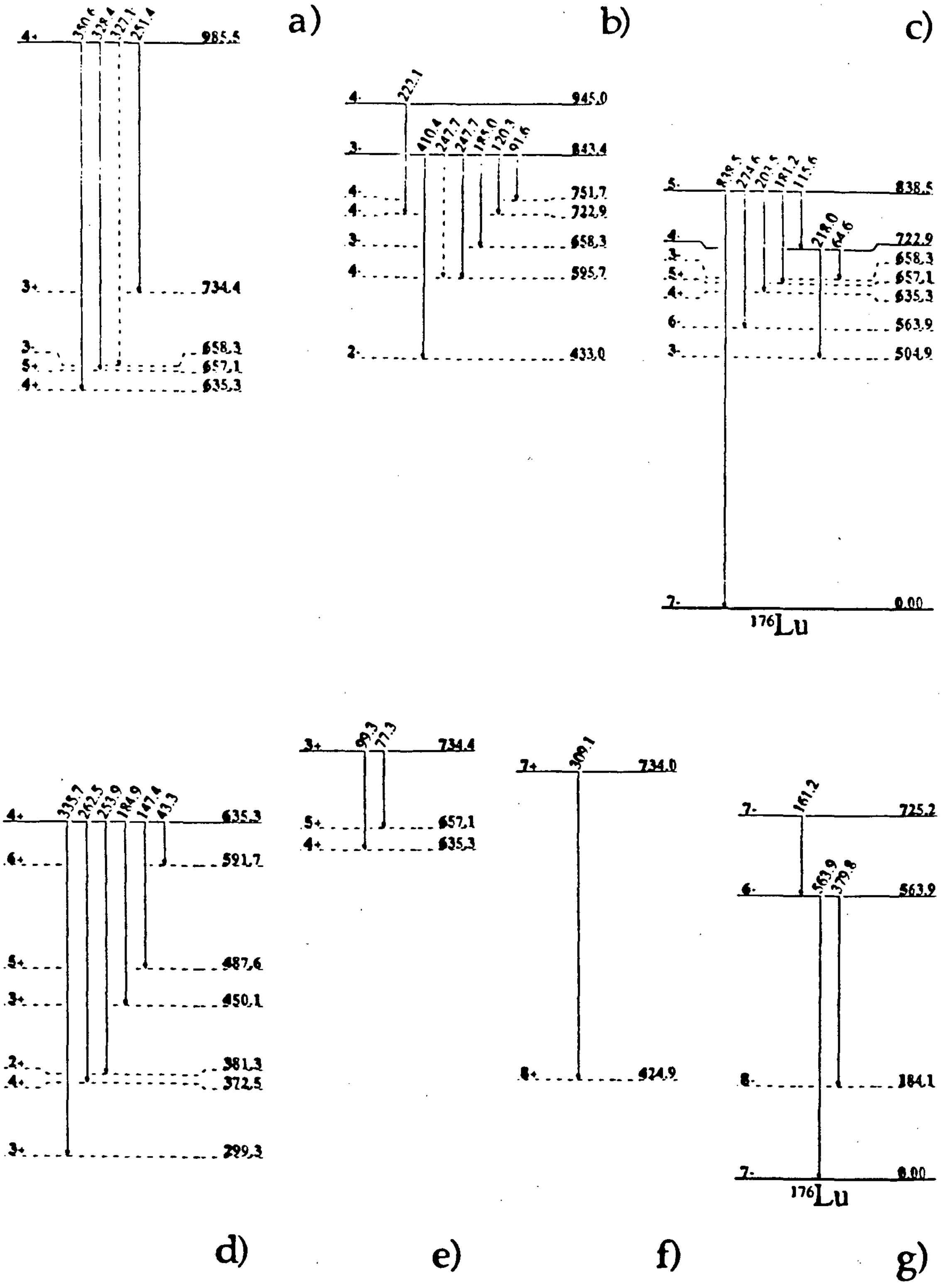

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Figure 4

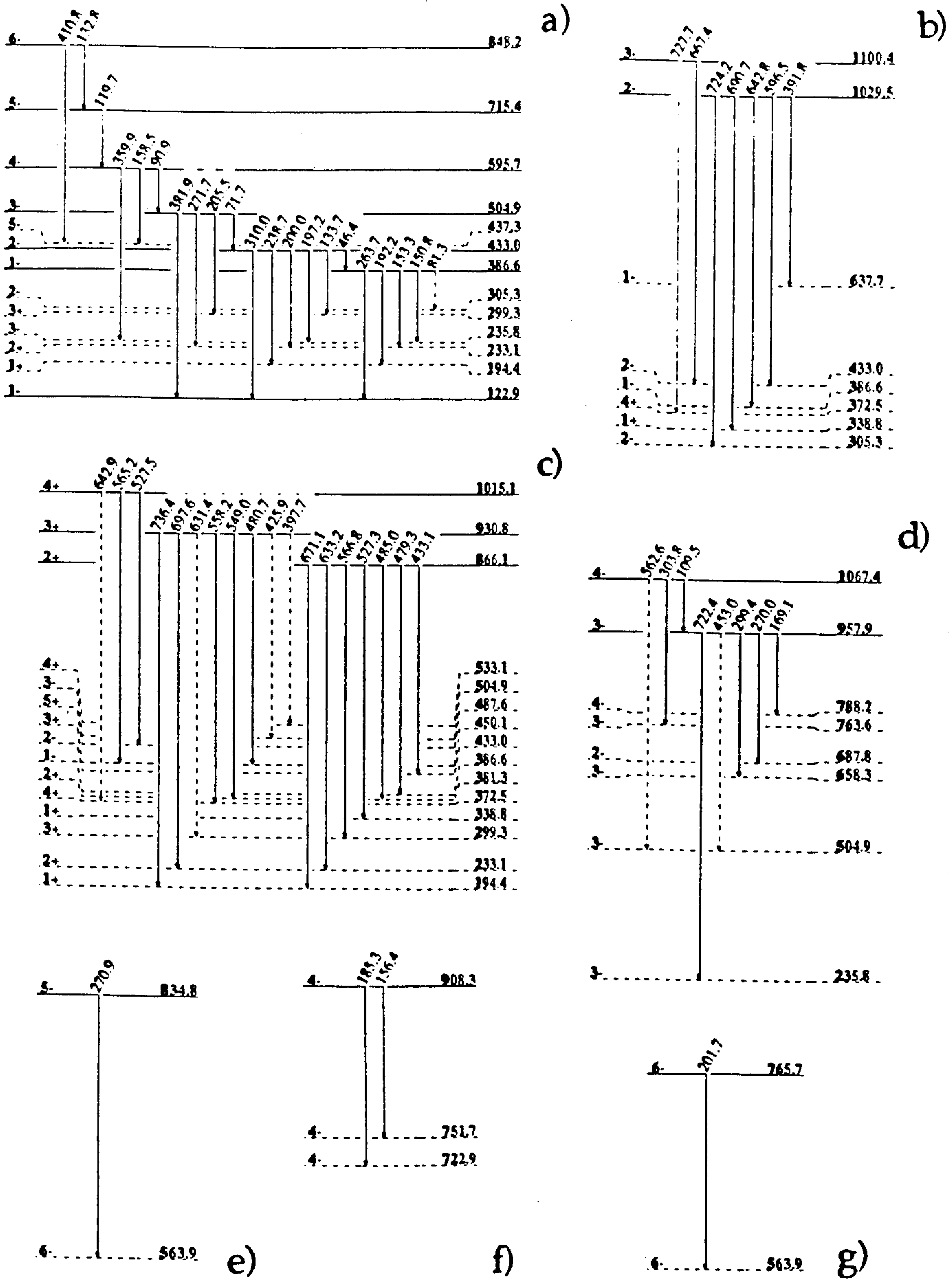

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Figure 5

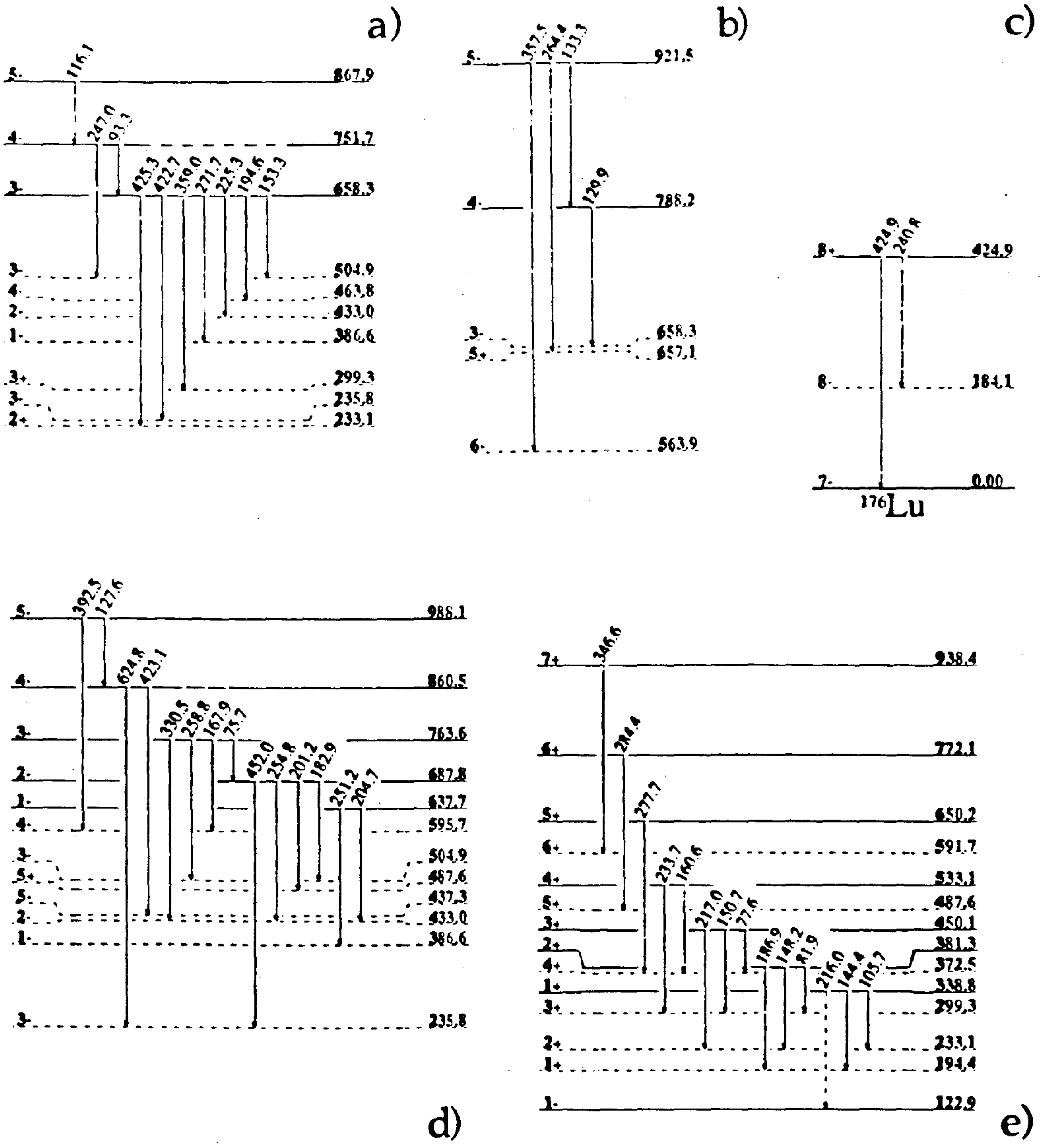

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Figure 6

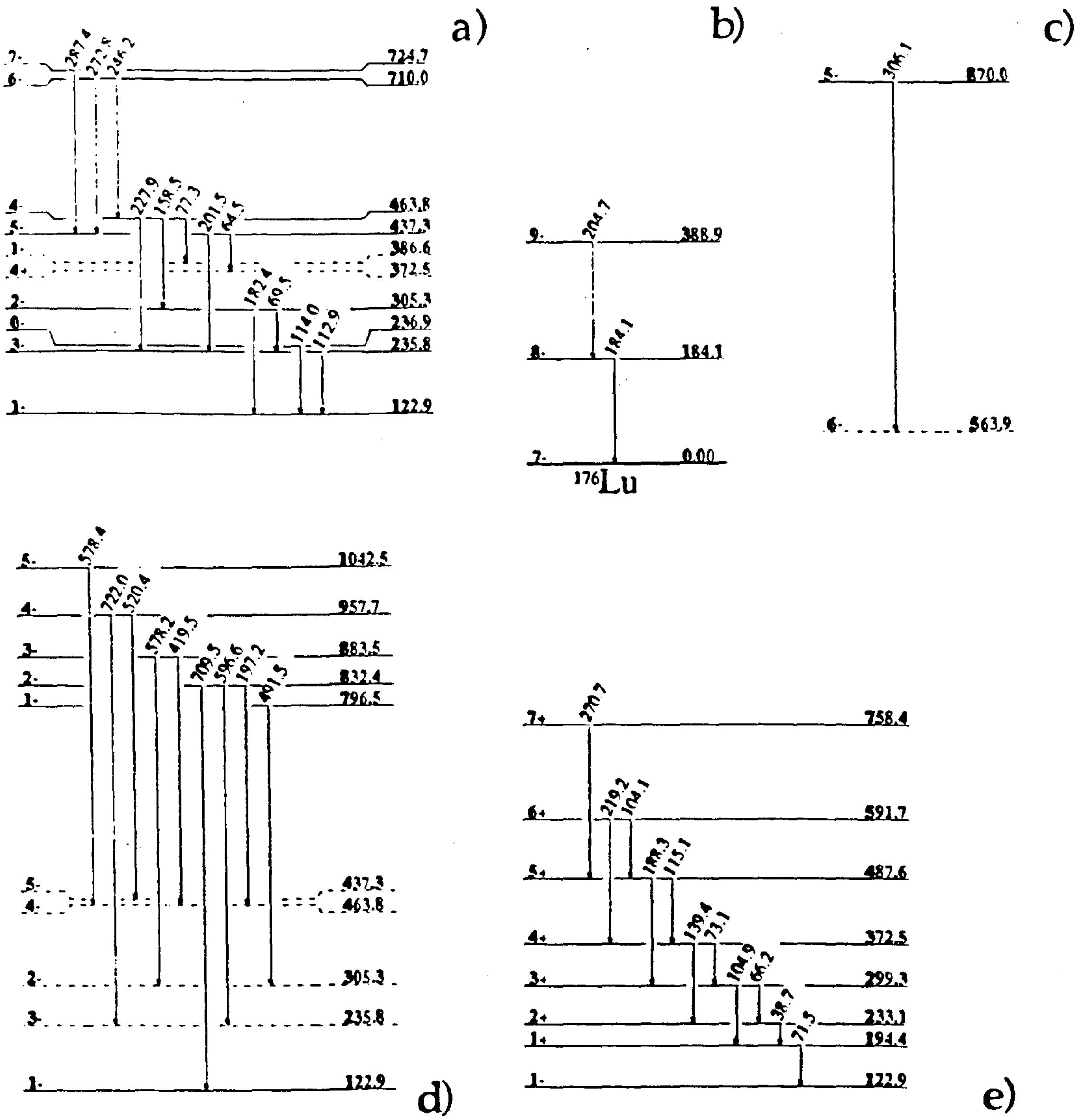

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Figure 7

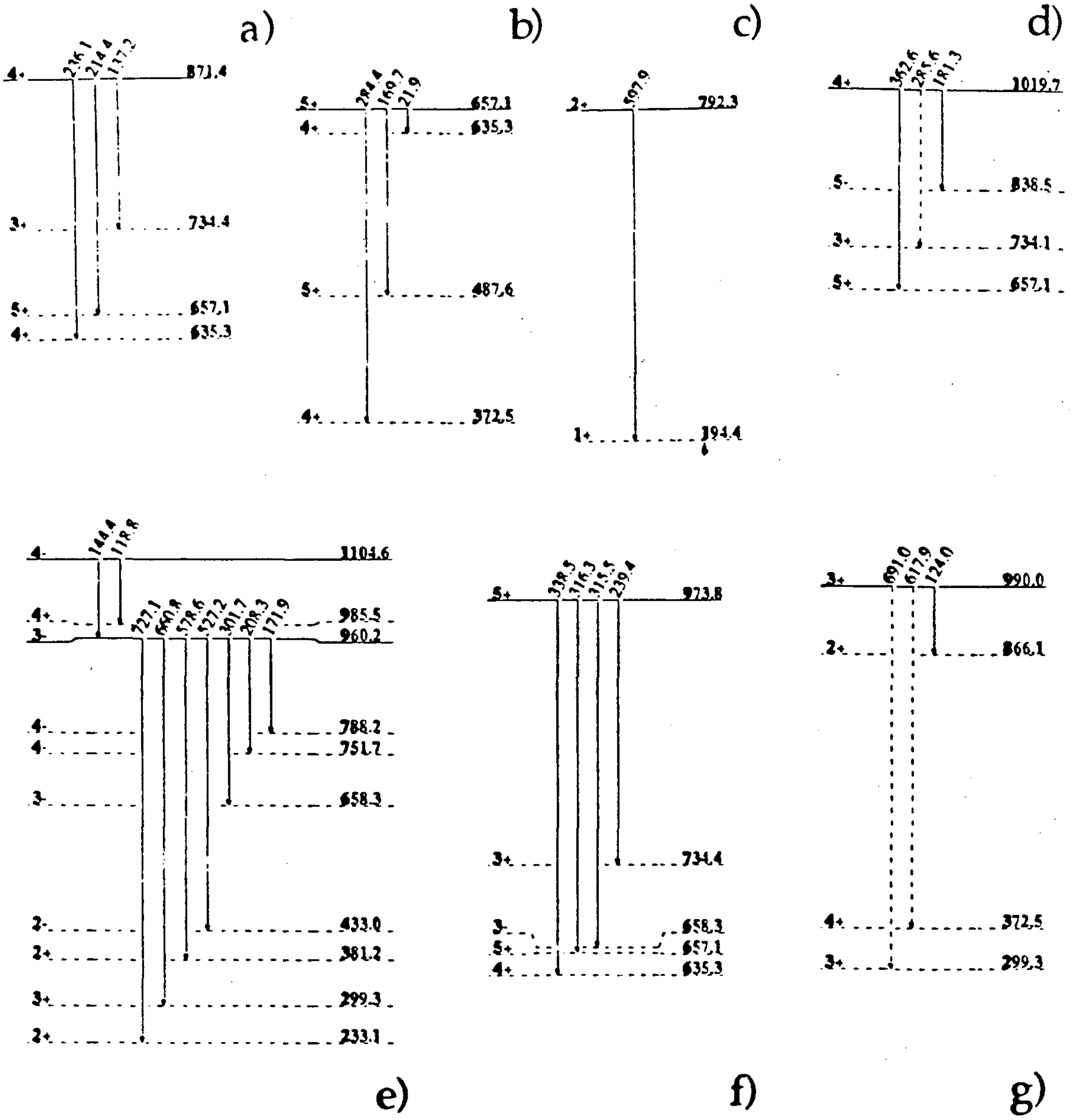

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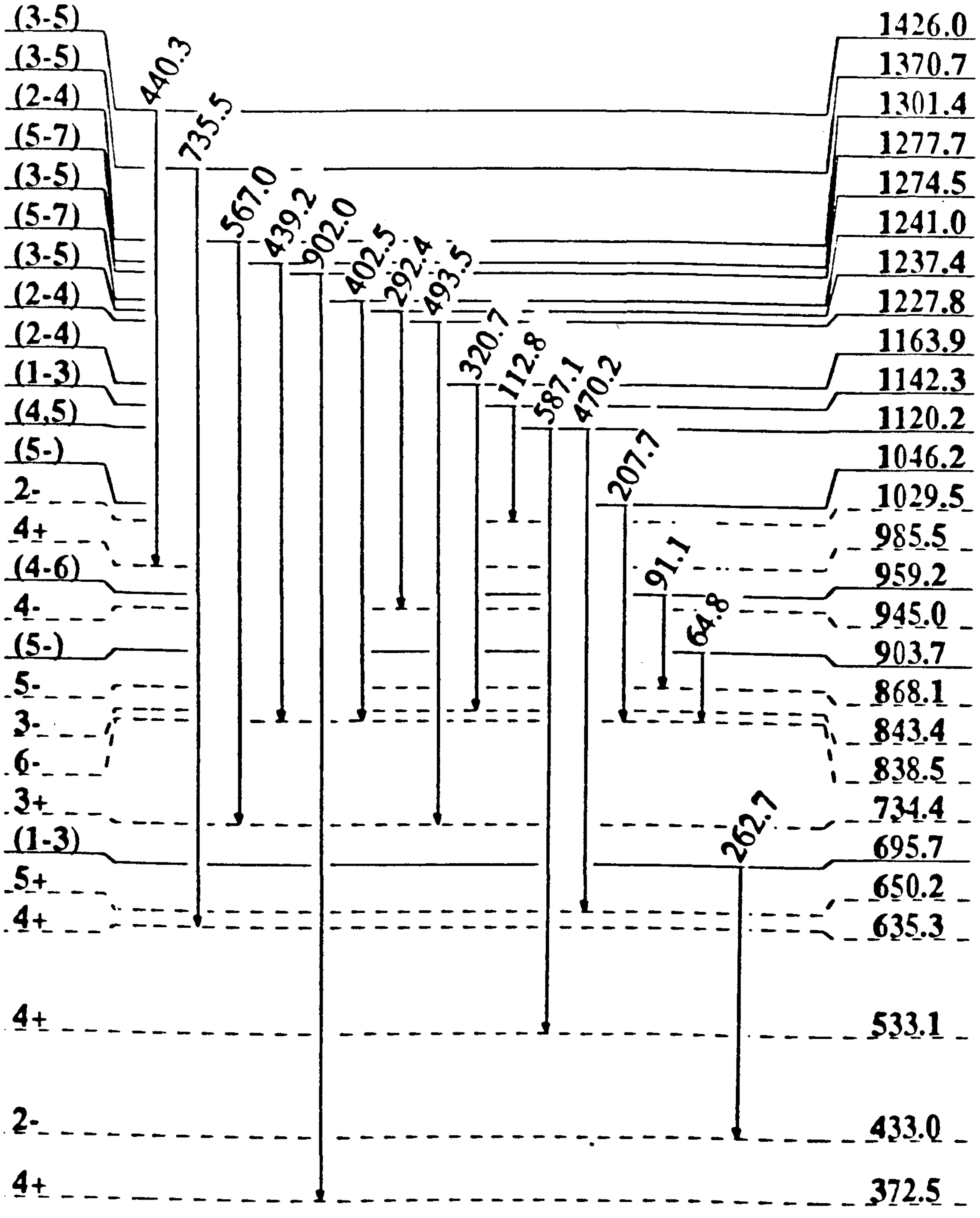
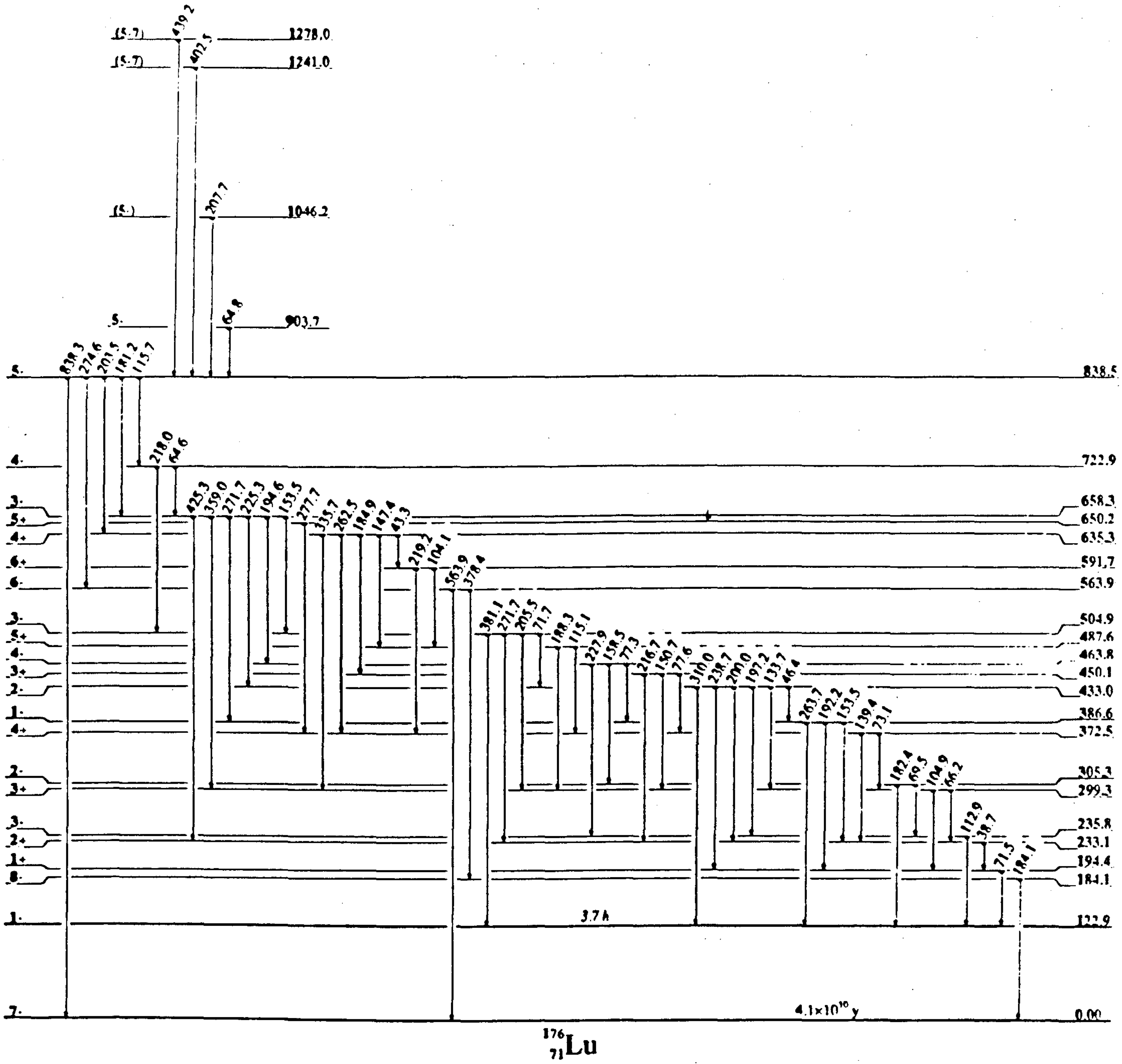
Figure 8



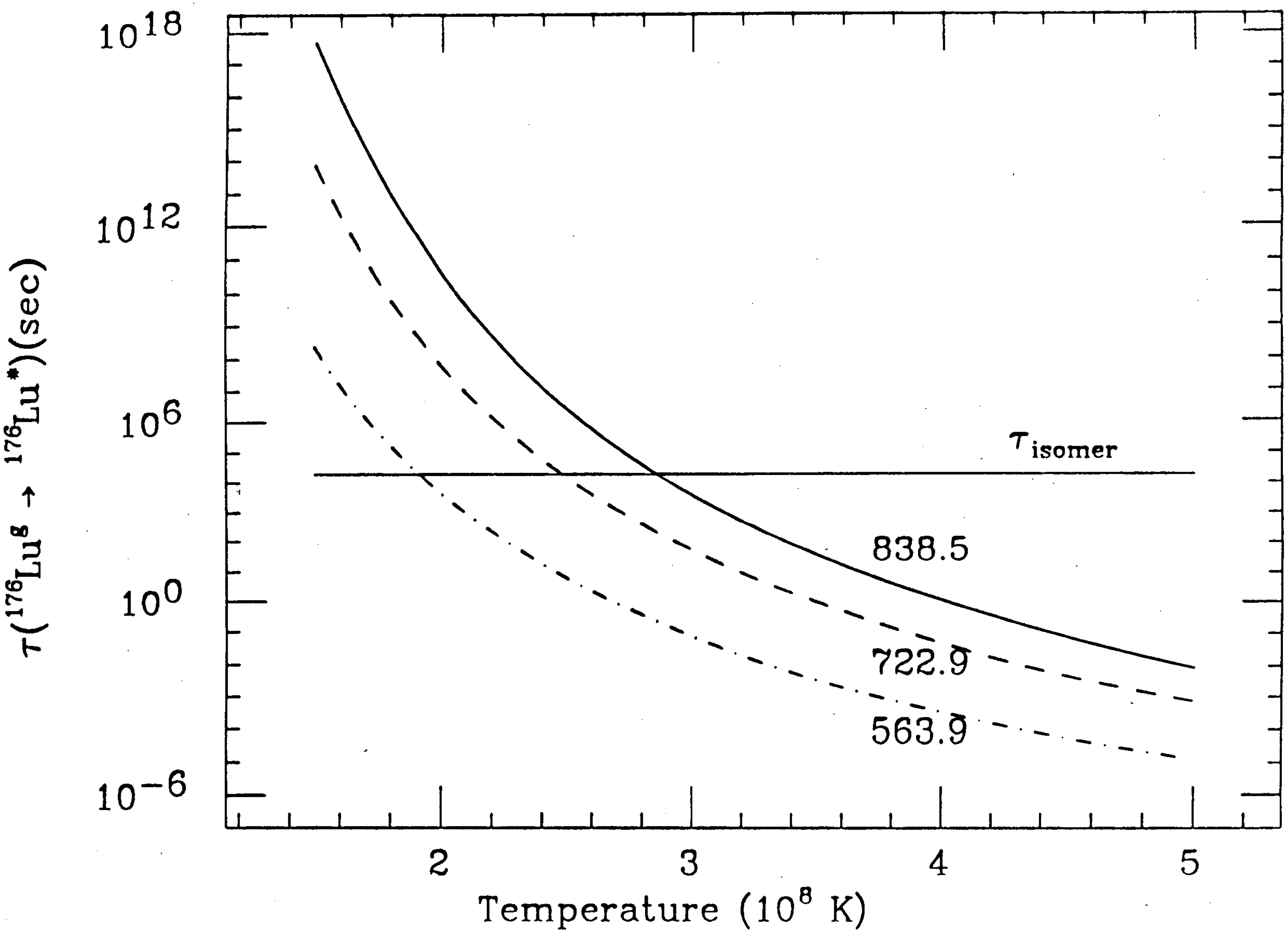
XBL 905-1758

Figure 9



XBL 909-3024

Figure 10



XBL 909-2969

Figure 11

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