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Isomeric Levels in ^{180}Lu and the Nucleosynthesis of $^{180}Ta^m$

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Abstract: Searches have been made for short-lived isomers in ^{180}Lu suggested by the preceding paper. Samples of ^{180}Lu were produced via the ^{180}Hf(n,p) reaction. Evidence for decay of ^{180}Lu isomer was sought by examining the decay curves of the known ^{180}Lu and $^{180}Hf^m$ delayed γ rays, and by searching for previously unidentified delayed γ rays. No evidence for a ^{180}Lu isomer was observed for half-lives between 10 and 300 seconds. The relevance of these observations for understanding the nucleosynthesis of $^{180}Ta^m$ is discussed.

I. Introduction

One of the continuing puzzles in nuclear astrophysics is the origin of the naturally-occurring $I^{\pi}=9^{-}$ isomer $^{180}Ta^{m}$, which appears as 0.012% of natural Ta[1]. Even this small abundance can not be explained with the conventional s- (slow) and r- (rapid) neutron-capture processes. The presence of the stable $^{176-180}Hf$ isotopes allows the standard s- process flow to bypass $^{180}Ta^{m}$. The β decays following r- process neutron-captures would terminate at the first stable A=180 product, ^{180}Hf. Recently, however, it has been suggested that $^{180}Ta^{m}$ may be synthesized via small β-decay branches off the main s- and r- process paths.[2,3] Beer and Ward [2] have proposed a model in which $^{180}Ta^{m}$ is reached by the β decay of $^{180}Hf^{m}$. An upper limit on the β-decay branch of the $I^{\pi}=8^{-}$ $^{180}Hf^{m}$ to $^{180}Ta^{m}$ has recently been established to be $\leq$0.35% and, within the context of this model, places a limit on the s-process contribution to the observed $^{180}Ta^{m}$ abundance of no more than 15%.[4] The r-process can account for some or all of the remaining $^{180}Ta^{m}$, if $^{180}Hf^{m}$ is also fed by a small fraction, f_m, of the β decay of ^{180}Lu.[2]

Two groups have undertaken experiments to look for this feeding with conflicting results. In the first experiment, Kellogg and Norman[5] activated Hf metal samples with fast neutrons, producing ^{180}Lu and $^{180}Hf^{m}$ via the (n,p) and (n,n') reactions, respectively. The Lu activity was chemically separated from the Hf in approximately 5 minutes. A search was made for evidence of an ingrowth of $^{180}Hf^{m}$ decay γ rays as a result of ^{180}Lu decay. An upper limit of $f_m \leq 0.026\%$ was set. A subsequent measurement by Eschner et al.[6] has reported a value of f_m=0.46±0.15%, which was obtained using an on-line mass separator following the bombardment of a W target with ^{136}Xe. As discussed in the preceeding paper of Kellogg and Norman, this discrepancy could be explained if there exists a high-spin, short-lived isomer of ^{180}Lu that β decays to ^{180}Hf^{m}. Such an isomer would have decayed before the chemistry of Kellogg and Norman was completed. This would explain their insensitivity to such a level, whereas the on-line mass separator with its much shorter time delay, could observe the decay of the ^{180}Lu^{m} and its effect on the ^{180}Hf^{m} population.

In an attempt to resolve the issue, we have undertaken a series of experiments to search for isomeric levels in ^{180}Lu. The time required by Kellogg and Norman to perform their radiochemistry establishes an approximate upper limit of the lifetime of ^{180}Lu^{m}, while the time required to extract ions from the online mass separator establishes the lower limit, if ^{180}Lu^{m} is to explain the discrepancy between these two experiments. As a result, we sought to investigate half-lives in the range of 10 s $\lesssim$ t $\lesssim$ 300 s.

The region of prolately-deformed nuclei surrounding ^{180}Lu is rich in examples of "K-isomers." In the known odd-odd isotopes of Lu and Ta, the couplings of single-particle Nilsson proton and neutron orbitals yield both high-spin (I=K=7-10) and low-spin (I=K=1-5) states in the first few hundred keV of excitation[1,7,8]. The $I^{\pi} = 3^{-}$ and 5^{+} states usually assigned to ^{180}Lu^{g} are seen at 0-and 6-keV excitation in ^{182}Ta.[1] Near the ground-state of ^{180}Lu we should expect the existence of the 1^{+} state seen in ^{178}Lu, ^{178}Ta, and ^{180}Ta as well as the 9^{-} state observed in ^{178}Lu and ^{180}Ta. In addition, the coupling of the proton in the $7/2^{+}$[404] orbital with the neutron in the $9/2^+[624]$ orbital produces an $I^{\pi}=8^+$ state which forms a Gallagher-Moszkowski doublet with the 1^+ state. This state is seen at 174 keV of excitation in ^{180}Ta [9]. Reports concerning isomers in ^{180}Lu are contradictory. [10-13] A recent calculation of the ^{180}Lu level scheme suggests that the $I^{\pi}=8^+$ and 9^- levels should be located within about 200 keV of the ground state[14] and are thus candidates for the high-spin isomer in ^{180}Lu. Fig. 1 illustrates the principle decay modes of the known 5.7-minute ^{180}Lu and possible decay modes of the proposed ^{180}Lu^m.

We can be guided in our expectations of the production of both low spin and high spin levels in ^{180}Lu by analogous work performed on ^{178}Lu[15]. Using the ^{178}Hf(n,p) reaction, it was found that the $I^{\pi}=9^-$ ^{178}Lu^m and the $I^{\pi}=1^+$ ^{178}Lu^g were produced with roughly equal cross sections. Furthermore, from the work of Kellogg and Norman[5], it is known that 8-15 MeV neutrons can bring in enough angular momentum to strongly excite the $I^{\pi}=8^-$ ^{180}Hf^m via the (n,n') reaction.

II. Experiment

Two separate experiments were performed to investigate the presence of isomeric levels in ^{180}Lu. In the first experiment, samples of approximately 15 mg of HfO_2, enriched to 93.8% in ^{180}Hf, were bombarded with fast neutrons. The $^{180}HfO_2$ samples were contained in small polyethylene botles and were wrapped with cadmium during activation. A thick, water-cooled beryllium target was bombarded with deuterons from the Lawrence Berkeley Laboratory 88-Inch Cyclotron, producing neutrons via the ^{9}Be(d,n) reaction. Separate activiations were performed at deuteron bombarding energies of 20,30,40,50 and 60 MeV. Each sample was activated for ~120 seconds. The beam was then turned off and the sample was quickly carried to a remote counting area where it was counted for up to 2500 seconds beginning about 90 seconds after the activation. Gamma-ray decays were observed using a shielded 110 cm^3 high-purity Ge detector. The energy and arrival time of each event was collected in event mode format. The time spectrum was divided into 512 time bins of 5 seconds each and the energy signals were digitized in 4096 channels with 0.5 keV/channel. In the first experiment, which was sensitive to longer time periods, we used a pulser at a known rate to correct for the deadtime of the data collection system. Examination of the data obtained in this experiment indicated that the yield of ^{180}Lu, relative to that of other activities, was maximized at a deuteron energy of 40 MeV.

In the second experiment, we used similar targets, but used a single deuteron energy of 40 MeV which produces a neutron energy spectrum with a peak at ~17 MeV.[16] A technique was developed to look for shorter isomeric lifetimes using an automated transfer system (rabbit) which transported the $^{180}HfO_2$ samples enclosed in small plastic vials (100mg of $^{180}HfO_2$/vial) from the irradiation site to the remote counting area, approximately 25 meters removed, in $\leq$ 3 seconds. The capsule was irradiated for 2 seconds and counted for 102 seconds. Again the data were collected in event mode format using 1.0 keV/channel, but using 0.2 seconds/time bin. In order to minimize the buildup of long-lived activities due to repeated irradiations, five separate samples were utilized. The initial counting rate when the capsule returned to the counting station was very high which resulted in very small livetimes for the data acquisition system. The principle component of this large background was a 7.1-second activity from ^{16}N formed by the $^{16}O(n,p)^{16}N$ reaction. In the rabbit experiment, γ rays observed from the decays of known nuclei were used as dead-time monitors. A relative measure of the deadtime can be obtained from each such γ ray by dividing the assumed exponential decay function by the observed time dependence.

III. Results

To determine whether isomeric levels are present in ^{180}Lu we analyzed the data using four methods. The first of these involved looking for the presence of previously unidentified γ rays in the irradiated $^{180}HfO_2$ sample. These new γ-ray lines could signal the existence of transitions in ^{180}Hf following the β decay of $^{180}Lu^m$ or transitions in ^{180}Lu by which $^{180}Lu^m$ decays to $^{180}Lu^g$. The event mode data from the first experiment were sorted into three γ-ray energy spectra corresponding to three different time bins while those from the rabbit run were sorted into five different spectra. Examples of γ-ray spectra obtained from both experiments are shown in Fig. 2. Comparison of the spectra allowed identification of activities with half-lives in the region of interest. An attempt was then made to identify all lines between 250 and 2000 keV to assess whether any new transitions could be identified. All unambiguous γ-ray lines between these energies were identified and no new transitions could be assigned to $^{180}Lu^m$. We were able to assess our sensitivity to this mode of identifcation of lines as a function of γ-ray energy by observing known $^{180}Lu^g$ transitions that possess well measured branching ratios at several energies in our spectra.

Our sensitivity to new γ-ray transitions is obviously a function of both the assumed half-life of $^{180}Lu^m$ and of the γ-ray energies. Assuming that the energies of any $^{180}Lu^m$ decay γ rays do not coincide with those produced by the decays of other nuclei identified in our samples, our sensitivities can be compared to the intensity of the 408-keV γ ray that is produced in 50% of the $^{180}Lu^g$ decays. The results of our searches for new γ rays in both experiments are summarized in Fig. 3.

The second method we employed consisted of examining the time history of particular transitions in ^{180}Hf, such as the 408-keV γ ray resulting from the decay of the 1608-keV level to the 1200-keV level. The β decay of $^{180}Lu^g$ has a 91% branching ratio to populate this state. If $^{180}Lu^m$ decayed electromagnetically to ^{180}Lu, the subsequent production of 408-keV γ rays would show an ingrowth and then the decay characteristic of the 5.7-minute $^{180}Lu^g$. The functional form of the rate of 408-keV γ rays would be:

$$\mathrm{Rate}(408) \propto N_g e^{-\lambda_g t}\left(e^{\lambda_g \Delta} - e^{-\lambda_g \Delta}\right) + N_m\left(\frac{\lambda_m}{\lambda_m-\lambda_g} e^{-\lambda_g t}\left(e^{\lambda_g \Delta} - e^{-\lambda_g \Delta}\right) - \frac{\lambda_g}{\lambda_m-\lambda_g} e^{-\lambda_m t}\left(e^{\lambda_m \Delta} - e^{-\lambda_m \Delta}\right)\right) \quad (1)$$

where λ_g (λ_m) is the decay rate for $^{180}Lu^g$ ($^{180}Lu^m$) and N_g (N_m) is the initial population of the ground state (isomer). The counting period is given by $t-\Delta$ to $t+\Delta$, where 2Δ is the bin width.

To enhance our statistics, we summed several γ rays produced in the decay of the 1608-keV level, specifically the 408, 1106, 1200 and the 1299-keV γ rays. We fit the background subtracted and livetime corrected spectra with this functional form, varying $t_{1/2}$ of the isomer between 10 and 300 seconds and obtained the populations of the hypothetical isomer and ground state levels that minimize χ_ν^2. The time spectra of the γ rays for both runs are shown in Fig. 4. Based upon analysis of these data we deduce a 1-σ upper limit on the relative $^{180}Lu^m$ production cross section as a function of the isomer lifetime, shown in Fig. 5a.

The third analysis we performed on our data was to examine it for signatures of $^{180}Lu^m$ β decay to the same levels as $^{180}Lu^g$ decays. In this case γ-ray decay curves would show the sum of two exponential components. We fit the same background and livetime corrected data with this new functional form and obtained the limits given in Fig. 5b.

Takahashi et al. reported a 2.5 minute β activity with a 3.3-MeV endpoint energy.[11] This was interpreted as the β decay of a low-spin isomer directly to the ground state of ^{180}Hf, but it has not been confirmed. If the spin of this isomer were 1^+, then it might also decay to the $I^\pi=2^+$ level at 93.3 keV in ^{180}Hf. We have sought evidence of this isomer by examining the decay curves of the 93-keV γ rays observed in our experiment. We have analyzed the time structure of the 93-keV γ rays in terms of a three component sum: a 5.7-minute component from the decay of the ground state of ^{180}Lu, a 5.5-hour component from the decay of $^{180}Hf^m$, and a 2.5-minute component from the decay of the proposed low-spin isomer in ^{180}Lu. We find no evidence for an isomeric level in ^{180}Lu that decays to the 93.3-keV state in ^{180}Hf, and can place an upper limit of 10% on the product of production cross section and decay branching ratio for such an isomer relative to that of the ground-state of

^{180}Lu.

It is possible that $^{180}Lu^m$ will not decay to either known or unknown levels in ^{180}Hf above 1150-keV, but does decay directly to $^{180}Hf^m$. This isomer decays to $^{180}Hf^g$ by a series of γ rays and can be monitored by observing the 443-keV γ ray. We examined our data for ingrowth of the population of $^{180}Hf^m$ by analyzing background and livetime corrected 443-keV γ ray time spectra using the functional form of Equation 1. The limited statistics of this γ ray seriously limit our ability to estimate the $^{180}Lu^m$ population. If this were the only $^{180}Lu^m$ decay mode, our data permit us to only rule out equal $^{180}Lu^m$ and $^{180}Lu^g$ production cross sections.

IV. Discussion

In conclusion, we have produced ^{180}Lu via the $^{180}Hf(n,p)$ reaction using a wide range of neutron energies and analyzed the resulting γ-ray energy and time spectra for evidence of the existence of isomeric levels in ^{180}Lu. As discussed in the preceeding paper, a high-spin isomer might be expected to electromagnetically decay to $^{180}Lu^g$ as well as β decay to $^{180}Hf^m$. Signatures of both of these decay modes were sought using several different types of data analysis. We have failed to observe any evidence of long-lived states in ^{180}Lu. This argues strongly against an isomer in ^{180}Lu with $10 \lesssim t_{1/2} \lesssim 300$ seconds as a means to explain the discrepancy between the results of Kellogg and Norman[5] and those of Eschner et al.[6] and reduces the likelihood of an r-process contribution to the solar-system abundance of $^{180}Ta^m$.

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Figure Captions

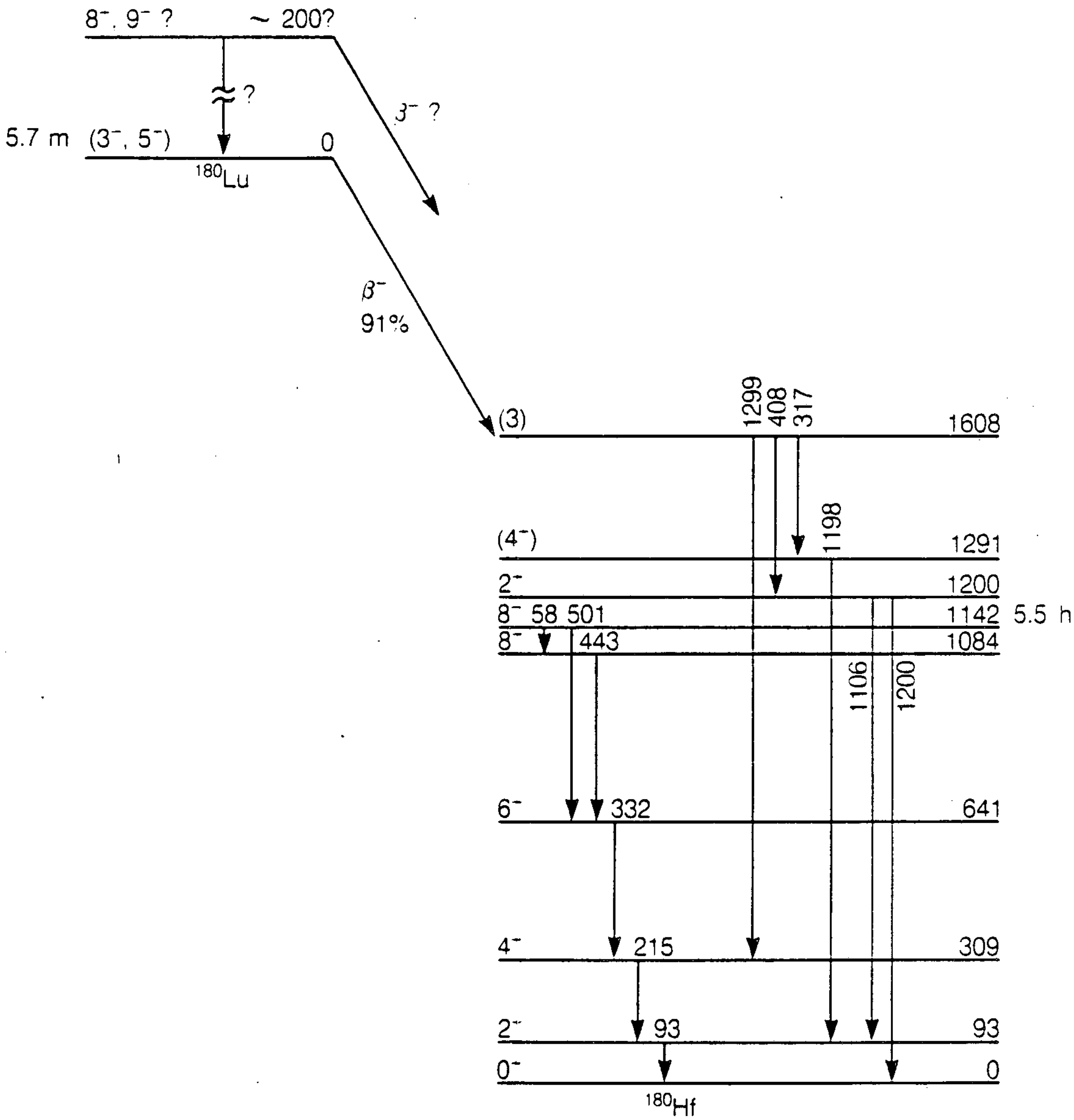
1. Partial level schemes of ^{180}Hf and ^{180}Lu, indicating some of the γ- and β-delayed, γ transitions used in this work.

2. Gamma-ray spectra observed following the bombardment of $^{180}HfO_2$ with fast neutrons generated by the ^{9}Be(d,n) reaction using 40-MeV deuterons. Peaks labeled with only an energy are attributed to the decay of $^{180}Lu^{g}$. a) Spectrum observed form 11 transport cycles of the rabbit system in a 5.2-second interval beginning 53 seconds after the end of the activation. b) Spectrum observed from a hand-carried sample in a 60-second interval beginning 110 seconds after the end of the activation.

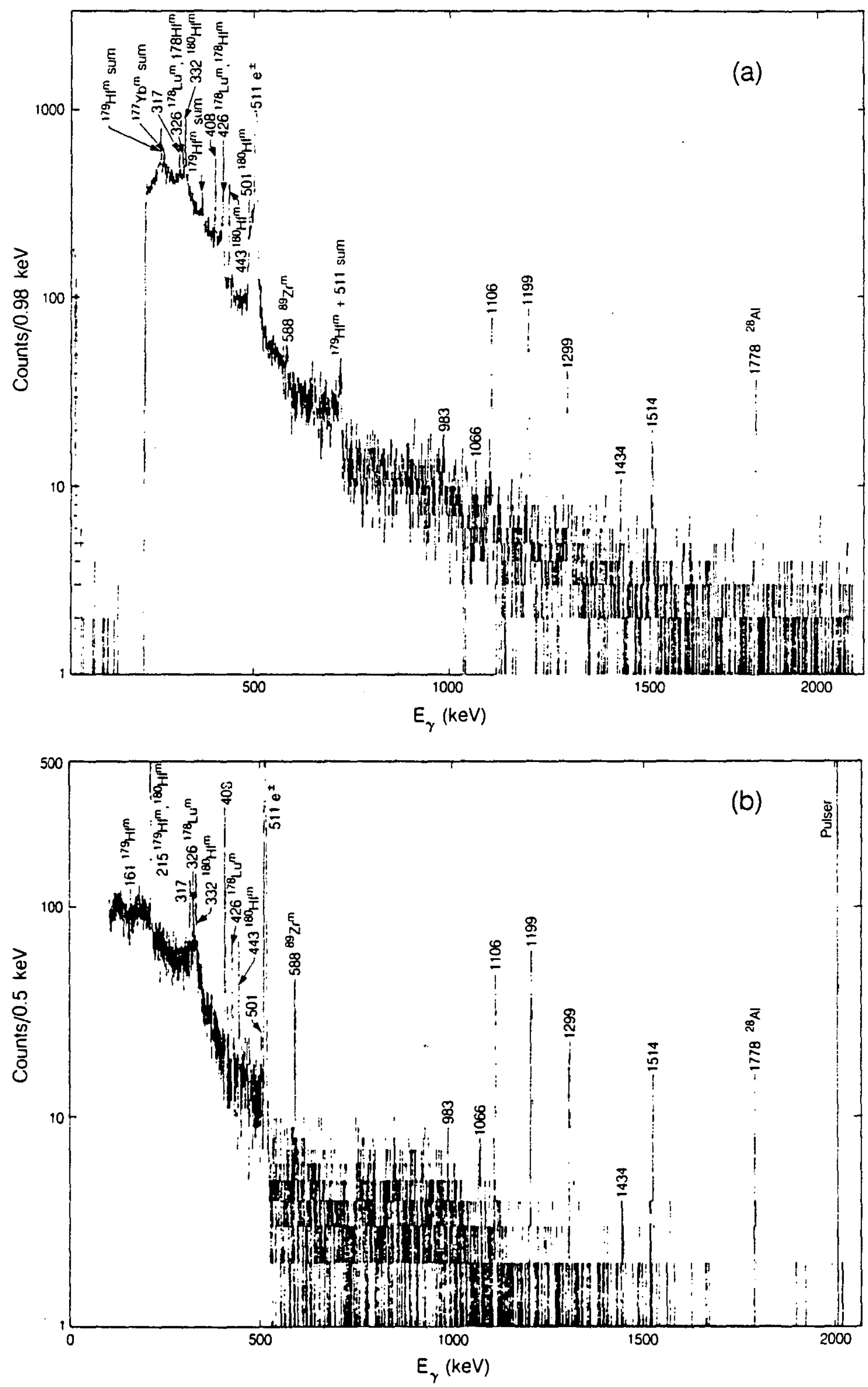
3. Sensitivity limits on observing new γ rays. The upper limits of the intensity of new γ rays from $^{180}Lu^{m}$ expressed as the ratio to the intensity of the 408-keV γ ray are shown as a function of the $^{180}Lu^{m}$ half-life. The solid (dashed) curve is for γ-ray energies less (greater) than 500 keV.

4. a) The integrated yield of γ rays from the decay of the 1608-keV level obtained using the rabbit. The curve is the result of a fit of a single exponential with a 5.7-minute half-life to these data. b) The yield of 408-keV γ rays from the hand-carried sample at E_d= 40 MeV. The curve is the result of a fit of a single exponential with a 5.7-minute half-life to these data.

5. a) Upper limits on the product of $^{180}Lu^{m}$ production cross section and decay branching ratio, B, relative to the production cross section for $^{180}Lu^{g}$. The solid curve (dashed) indicates the limits obtained by fitting the data from the rabbit (hand-carried) samples with Equation 1. b) The same upper limits as in a), except fitting the data with the sum of two exponential decay functions. The solid curve (dashed) represents the limits obtained by fitting the data from the rabbit (hand-carried) samples.



XBL 865-10785



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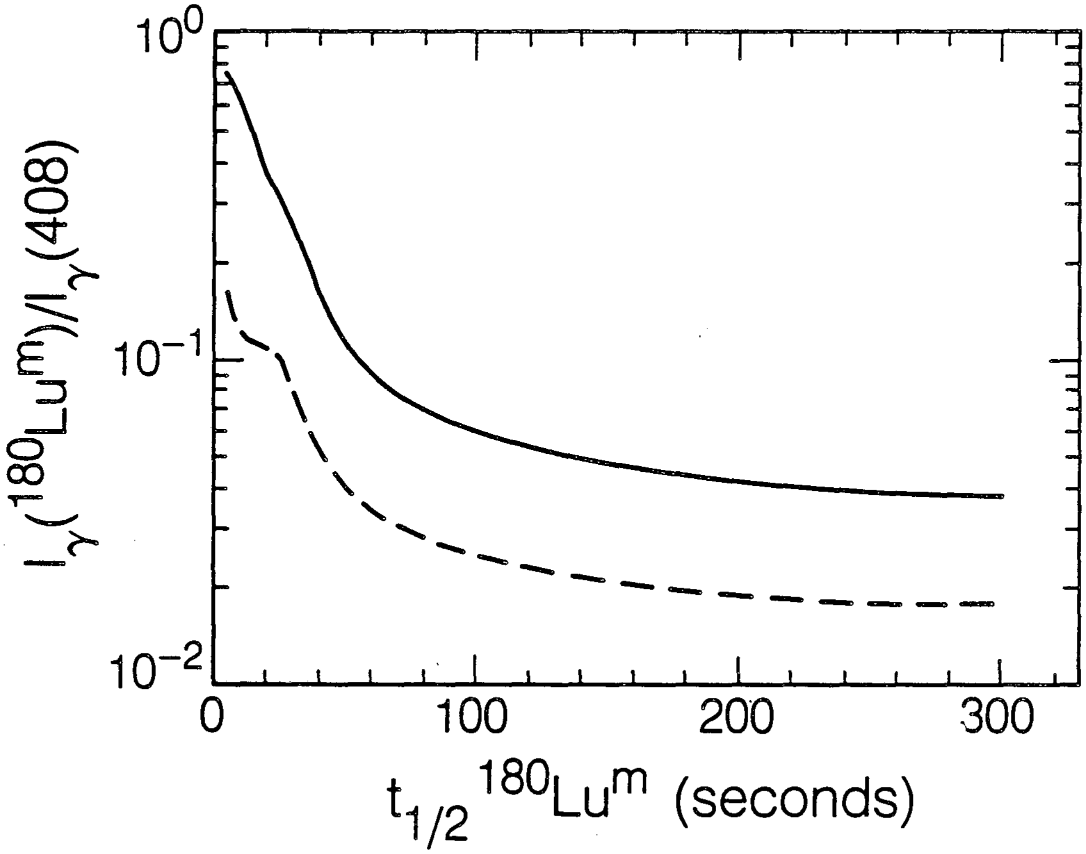
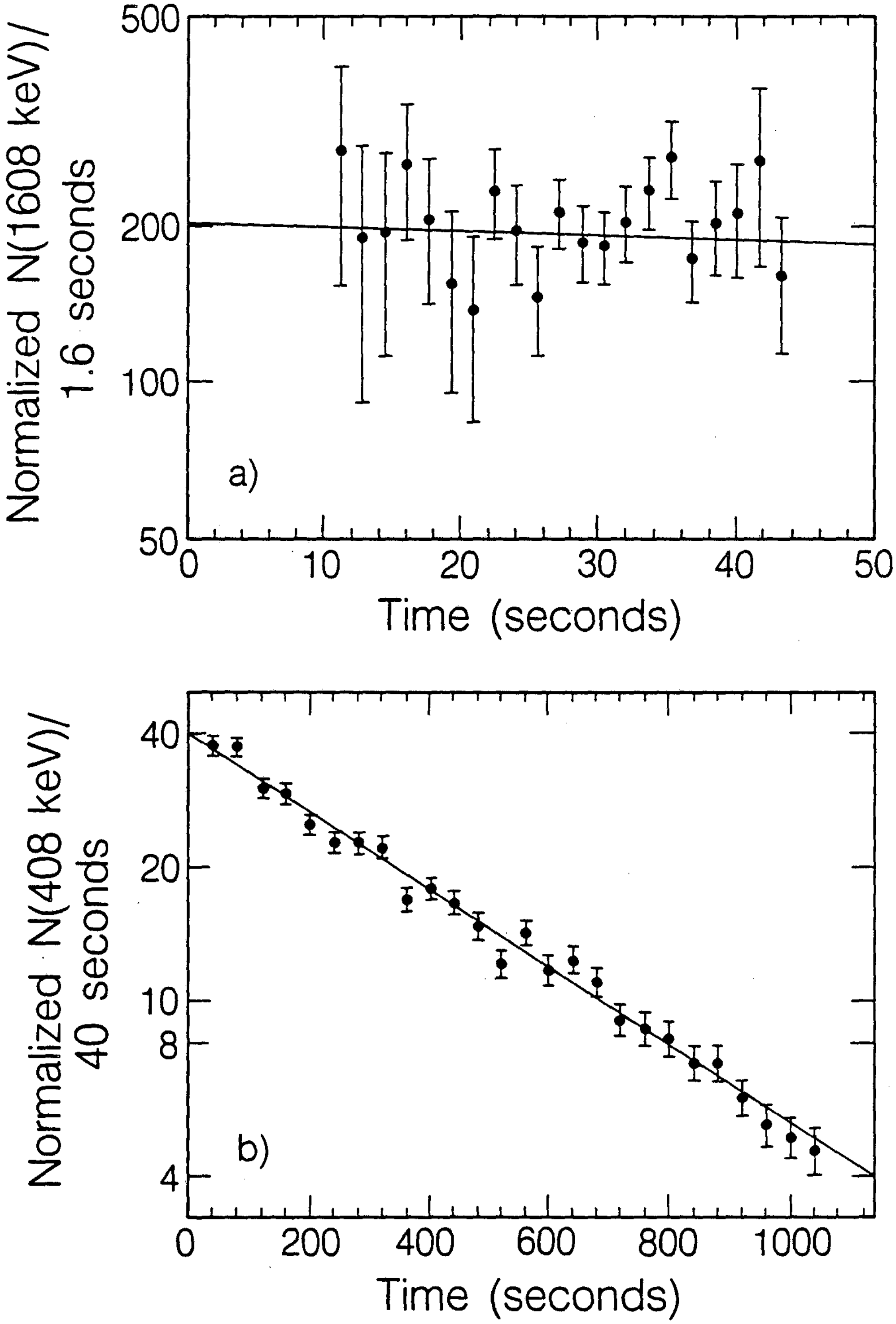
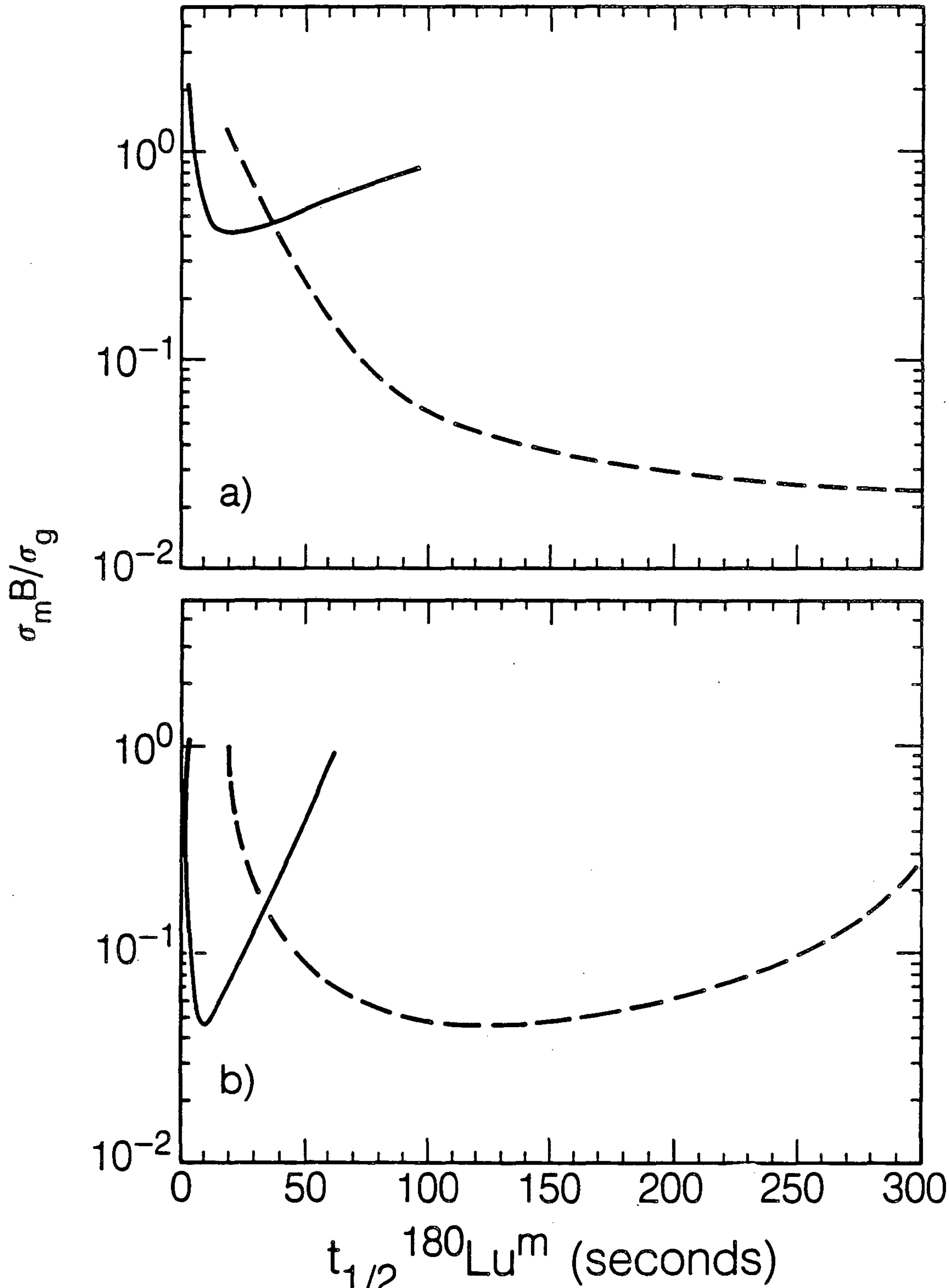

10^0
10^{-1}
10^{-2}
$I_\gamma(^{180}Lu^m)/I_\gamma(408)$
0
100
200
300
$t_{1/2}\ ^{180}Lu^m$ (seconds)

XBL 865-9187


Normalized N(1608 keV)/
1.6 seconds
500
200
100
50
a)
0
10
20
30
40
50
Time (seconds)
Normalized N(408 keV)/
40 seconds
40
20
10
8
4
b)
0
200
400
600
800
1000
Time (seconds)

XBL 865-9181



10^0
10^{-1}
10^{-2}
a)
$\sigma_m B/\sigma_g$
b)
0
50
100
150
200
250
300
$t_{1/2}$ $^{180}Lu^m$ (seconds)

XBL 865-9180

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