Half-lives of Au, Hg, and Pb isotopes from photoactivation

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The ground state half-lives of the gold isotope ^{196}Au, the mercury isotopes ^{195,197,203}Hg, the lead isotope ^{203}Pb, and the half-lives of two isomers in ^{199}Hg and ^{204}Pb have been measured with high precision using the photoactivation technique. The γ-ray activity was counted over several half-lives with high-purity germanium detectors. The measured half-lives are: $T_{1/2}(^{196}\mathrm{Au}) = 6.1669 \pm 0.0006$ d; $T_{1/2}(^{195}\mathrm{Hg}) = 10.53 \pm 0.03$ h; $T_{1/2}(^{197}\mathrm{Hg}) = 64.94 \pm 0.07$ h; $T_{1/2}(^{203}\mathrm{Hg}) = 46.6 \pm 0.1$ d; $T_{1/2}(^{204}\mathrm{Pb}) = 51.95 \pm 0.01$ h; $T_{1/2}(^{199m}\mathrm{Hg}) = 42.67 \pm 0.09$ min; $T_{1/2}(^{204m}\mathrm{Pb}) = 1.14 \pm 0.04$ h.

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The precise knowledge of half-lives is a prerequisite for the analysis of activation experiments. In a recent photoactivation experiment with a quasithermal photon spectrum at astrophysically relevant energies [1] (γ,n) cross sections have been analyzed which are relevant for the nucleosynthesis of the heavy neutron-deficient nuclei (e.g. Ref. [2]). For the running experiments on gold, mercury, and lead isotopes [3] the half-lives of the isotopes ^{196}Au, ^{195,197,203}Hg, and ^{203}Pb have to be known precisely to avoid systematic errors in the data analysis. This is especially relevant in the case when a relatively long time lies between the irradiation and the counting phase. Therefore we measured the half-lives of these isotopes by the decay curves of several γ-ray lines. Additionally, two half-lives of isomers in ^{199m}Hg and ^{204m}Pb have been determined.

In general, half-lives are known with relatively small uncertainties [4]. However, this is not the case for the neutron-deficient isotopes which can be produced by the (γ,n) reaction on the most neutron-deficient stable isotopes because these isotopes have typically very low abundances in the order of 0.01–1 %. For example, the half-life of ^{195}Hg is only known with an uncertainty of about 5%, and the two available experimental results are not consistent [4–6]. Additionally, it has been shown in our previous experiment [7] that the adopted values with their quoted small uncertainties in Ref. [4] are not always in agreement with new experimental results which used high resolution detection techniques to avoid systematic uncertainties from background activity.

The experiment was performed at the real photon facility [8] of the superconducting Darmstadt linear electron accelerator S-DALINAC [9]. Our samples of gold, mercury, and lead were irradiated with bremsstrahlung at an endpoint energy of 9.975 MeV for about 12 h. The samples consisted of 149 mg Au, 2616 mg HgS, and 12.91 g Pb in natural isotopic composition. The bremsstrahlung was produced by a continuous wave electron beam of about 25 μA impinging on a copper bremstarget. The samples were mounted directly behind the copper bremstarget, and they were activated simultaneously mainly by (γ,n) reactions and by (γ,γ') in the case of the two isomers. The activity of mercury and lead was counted immediately after the irradiation using two high-purity germanium (HPGe) detectors with efficiencies relative to a 3 in.×3 in. NaI(Tl) crystal of 30% (Hg) and 100% (Pb). After 270 h the mercury sample was replaced by the gold sample, because the activity of the relevant mercury isotopes has totally disappeared. A γ-ray spectrum is shown in Fig. 1.

The photon spectra of the Au, Hg, and Pb samples were accumulated for 834, 271, and 485 h with an automatic data saving every hour for the analysis of the decay curves. The absolute time scale was derived from the time standard distributed by the Physikalisch-Technische-Bundesanstalt, Braunschweig, Germany via a long-wave radio signal. The excellent resolution of the HPGe detectors made us able to analyze each γ line separately, reducing the disturbances of the other lines to a minimum.

The dead time was determined automatically by the data acquisition system by measuring the duration of the busy signal of the 100-MHz Wilkinson ADC (Silena 7411) which is true from the beginning of each conversion (input signal exceeds lower level discriminator) until the data acquisition system acknowledges the transfer of the digitized data. The dead time of the whole system is given by this period because it exceeds the signal width of the analog amplifier. The dead time given by the data acquisition system was checked in a previous experiment with a pulse generator at variable frequencies, and it was found that the automatically determined deadtime is reliable up to count rates of more than 10^4/s [7].

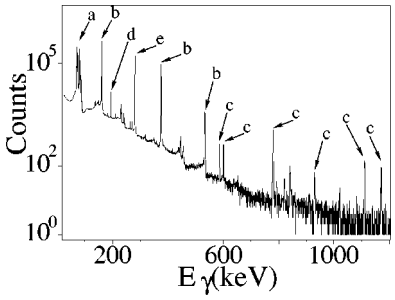

FIG. 1. Photon energy spectrum of the HPGe detector of the mercury sample after irradiation. (a) x ray, (b) lines and sum lines from ^{199m}Hg, (c) lines from ^{195}Hg, (d) lines from ^{197}Hg, (e) lines from ^{203}Hg. Minor branches are not labeled; all lines have been identified.

TABLE I. Comparison of our results (weighted average of all analyzed decay curves) with the adopted values from Ref. [4]. For each analyzed decay curve we list the γ-ray energy, the reduced χ^2/F, and the half-life $T_{1/2}$. Additionally we compare the weighted average of our results to the adopted values.

Isotope	E_γ (keV)	χ^2/F	$T_{1/2}$ [a]	$T_{1/2}$ [b]
^{195}Hg	w. average:		10.53±0.03 h	9.9±0.5 h
	585.1	0.81	10.62±0.08 h	
	599.7	1.14	10.57±0.08 h	
	779.8	1.09	10.49±0.04 h	
	930.9	1.05	10.74±0.33 h	
	1111.0	1.42	10.54±0.13 h	
	1172.4	0.93	10.66±0.14 h	
^{197}Hg	191.4	1.73	64.94±0.07 h	64.14±0.05 h
^{203}Hg	279.2	1.07	46.6±0.1 d	46.612±0.018 d
^{199m}Hg	w. average:		42.67±0.09 min	42.6±0.2 min
	158.3	2.75	42.68±0.11 min	
	374.1	1.19	42.65±0.19 min	
^{203}Pb	w. average:		51.95±0.01 h	51.873±0.009 h
	279.2	1.12	51.94±0.01 h	
	401.3	1.04	52.01±0.04 h	
	680.5	1.08	52.02±0.06 h	
^{204m}Pb	w. average:		1.14±0.04 h	1.120±0.005 h
	374	0.20	1.11±0.10 h	
	899	0.26	1.14±0.05 h	
	912	0.28	1.17±0.10 h	
^{196}Au	w. average:		6.1669±0.0006 d	6.183±0.010 d
	333	1.08	6.1686±0.0014 d	
	356	1.23	6.1666±0.0007 d	
	688	1.00	6.1627±0.0048 d	
	1091	1.08	6.163±0.047 d	
	1446	1.17	6.3±1.9 d	

[a]This experiment.
[b]Adopted values from Ref. [4].

The γ-ray spectra were analyzed with the program package TV [10]. Different regions of interest for the peak areas and for the background calculation were chosen to check the consistency of the data analysis procedure, and the resulting uncertainties were typically in the order of less than 0.1%. Where several decay curves could be observed using only peaks with sufficient statistics, it turned out that the measured half-lives from the different γ rays were internally consistent and a weighted average was calculated. The reduced χ^2 per degree of freedom F is close to unity for all analyzed decay curves with good statistics. A summary of the results is given in Table I.

In the following we present the results from our photoactivation experiment and discuss the differences to previous measurements. Note the significantly reduced uncertainties in several cases.

^{195}Hg. Because of the short half-life of about 10 h, we only could follow the decay from 37 h (930.9-keV line) up to 80 h (779.8-keV line). Nevertheless, the statistics were good enough to measure the half-life with high precision. Hence the published values [5,6] of 9.5±0.5 h and 11.5±1.0 h could be upgraded to the accurate value of 10.53±0.03 h. Two decay curves are shown in Fig. 2.

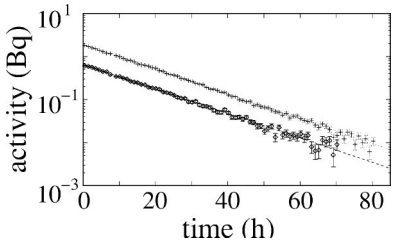

FIG. 2. Decay curves of ^{195}Hg at E_γ=779.3 keV (upper line) and at 585.1 keV (lower line).

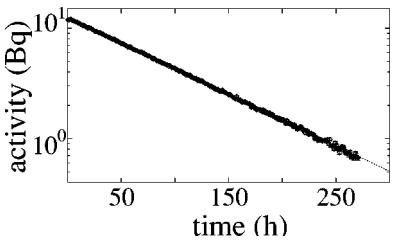

FIG. 3. Decay curve of ^{197}Hg at $E_\gamma = 191.4$ keV.

^{197}Hg. The known value of 64.14 ± 0.05 h from literature [4,11] could not be confirmed. We annotate that we were able to exclude all potentially systematic errors due to the excellent energy resolution of the HPGe detector. Our result of 64.94 ± 0.07 h is based on the analysis of 270 measured spectra corresponding to more than four half-lives. The decay curve is shown in Fig. 3. Other measured values vary between 64 and 66.4 h [12–14].

^{203}Hg. ^{203}Hg is a standard calibration source, and its half-life is well known. The adopted value [4] of 46.612 ± 0.018 d could be verified (46.6 ± 0.1 d) within our uncertainties. Due to the long half-life of ^{203}Hg it was not the aim of this experiment to improve this value, but the conformity verifies our other measurements.

^{199m}Hg. The decay curve (Fig. 4) of the $J^\pi = 13/2^+$, $E_x = 532$-keV isomer in ^{199}Hg was measured over a time interval of 8 h in the case of the 158-keV line, and of 9 h for the 374-keV line. The previous results in literature show some inconsistencies [4]. They vary from 42.6 ± 0.2 min [15] to 44.4 ± 0.5 min [16] (see also Refs. [17–19]). Our result of 42.67 ± 0.09 min is identical to the adopted value [15,4] within the errors. However, the uncertainty could be reduced by a factor of 2.

^{203}Pb. The decay of this isotope we could follow over 450 h, shown in Fig. 5. The analysis was made with the help of three lines. Because of high intensity and the small distance between the sample and the detector, sum lines occurred in the first spectra. Therefore we had to exclude the first spectra from the analysis. The measured half-life of 51.95 ± 0.01 h could not confirm the adopted value 51.873 ± 0.009 h [4] measured by Hoppes *et al.* [20] with comparable precision. But it agrees with results of former works [21–24] which range from 51.88 to 52.10 h.

^{204m}Pb. We could follow the decay curves of the $J^\pi = 9^-$, $E_x = 2186$-keV isomer in ^{204}Pb over 3 h in the case of the 374-keV line, and over 7 h in the case of the 899-keV line. The analysis of the 912-keV line succeeded for 4 h, but

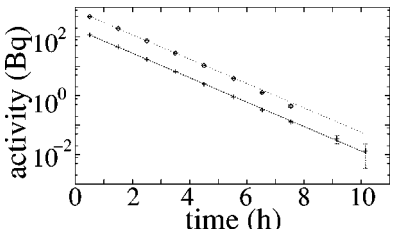

FIG. 4. Decay curve of ^{199m}Hg at $E_\gamma = 158.3$ keV (upper line) and at 374.1 keV (lower line).

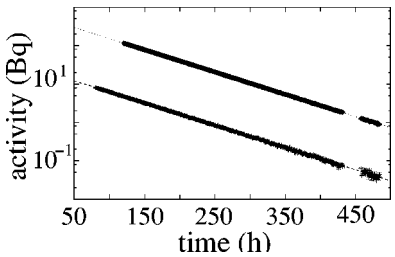

FIG. 5. Decay curve of ^{203}Pb at $E_\gamma = 279.3$ keV (upper line) and at 401.3 keV (lower line). The missing data points are again the consequence of a general failure of electric power in a thunderstorm over Darmstadt which seems to appear in all long time measurements [7].

there was a difficulty. This line overlaps with a background line. Thus we had to calculate the background with the last 100 spectra retrieved. Our value of 1.14 ± 0.04 h shows good consistency with the literature value [4] of 1.120 ± 0.005 h.

^{196}Au. High precision was achieved by the long observance of 834 h. Our evaluated half-life of 6.1669 ± 0.0006 d is in conformity with literature [4] measured by Ref. [25]. We have reduced the uncertainty of the half-life by more than one order of magnitude. Two decay curves of this isotope are show in Fig. 6.

In this experiment the half-lives of ^{195}Hg, ^{197}Hg, ^{203}Hg, ^{199m}Hg, ^{203}Pb, ^{204m}Pb, and ^{196}Au have been measured with high precision. The adopted values [4] could be significantly improved for ^{195}Hg, ^{199m}Hg, and ^{196}Au. For ^{197}Hg and ^{203}Pb our results have similar uncertainties but they are in slight disagreement with the adopted values [4]. The adopted half-lives have been confirmed for the standard γ source ^{203}Hg and for the short-lived isomer ^{204m}Pb. The improved half-lives will be needed for the analysis of the running activation experiment [3], especially in the case of ^{195}Hg, where the half-life was measured for the first time with high precision.

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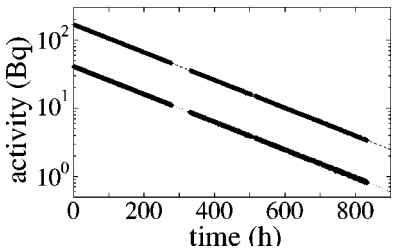

FIG. 6. Decay curve of ^{196}Au at $E_\gamma = 333$ keV (lower line) and at 356 keV (upper line) with the same gap in the data (see Fig. 5).

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