Radiochemical Study of Tl^{195}, Tl^{197}, and Tl^{198m}†

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A new neutron-deficient nuclide, Tl^{195}, has been produced by 20-Mev deuteron bombardment of Hg^{196}, and its identity and its half-life (1.2±0.1 hours) have been established by timed chemical separations of the Hg^{195} daughter. The assignment and half-life (2.8±0.2 hours) of Tl^{197} have been confirmed by timed chemical separations of its Hg^{197} daughter. The radiations of Tl^{197} and Tl^{198m} have been examined with a gamma scintillation spectrograph and a 180° beta-ray spectrograph. The presence of a number of new gamma rays associated with Tl^{198m} reveals an electron capture branch in the decay of this isomer; the data indicate a ratio of electron captures to isomeric transitions between 1 and 2.

INTRODUCTION

RECENT measurements of the yields of thallium, lead, bismuth, and polonium spallation products from bombardment of bismuth with 375- and 450-Mev protons[1] and of thallium and lead from bombardment of bismuth with 2.2-Bev protons[2] suggest that a significant fraction of the total spallation yields are made up of isotopes of these elements not yet known or identified. In the case of the thallium isotopes of masses 202 down to 198, a series for which measurements are reported in both investigations, the yields tended to increase with decreasing mass number, with indications that the yield trend had not reached maximum at 198, the lightest isotope of this series then known. The next two lighter isotopes, Tl^{197} and Tl^{196}, have recently been reported by other investigators.[3]

Since it appeared feasible to produce Tl^{197}, Tl^{196}, and Tl^{195} by charged-particle bombardment of gold and mercury at the Brookhaven 60-inch cyclotron, the present study was undertaken with the primary aim of characterizing these nuclides sufficiently for further spallation yield work.

This report describes, in a preliminary way, the identification of Tl^{195}, the confirmation of the half-life and some of the radiations ascribed to Tl^{197}, and some additions to previous information on the radiations and modes of decay of Tl^{198m}.

EXPERIMENTAL PROCEDURES

The gold targets were either 0.5-mil (~25 mg/cm²) foils of the rolled metal or ~1 mg/cm² foils of the metal evaporated onto aluminum. The mercury targets consisted of "mercury foils" deposited on 1-mil copper foil by electrochemical replacement. For thin deposits (as with small quantities of isotopically enriched mercury), a drop of the mercury solution in 3–6 normal nitric acid was pipetted onto the copper and stirred for a few minutes until most of the mercury was deposited. For heavier deposits (normal mercury), mercuric oxide was dusted onto the copper, moistened with 3 normal nitric acid, and rubbed with a stirring rod until the fresh amalgam surface appeared. The mercury-copper foils were used within a few hours after preparation, before the mercury had migrated down into the bulk of the copper, and when mounted carefully on a water-cooled target block and covered with a 0.5-mil aluminum protecting foil, could be exposed to up to 2μa/cm² of 20-Mev deuterons without apparent evaporation of the mercury.

The separation and purification of thallium from mercury, copper, and zinc was based on multiple extractions of Tl^{+++} into either from an aqueous phase made 1 normal in hydrobromic acid and containing the appropriate holdback carriers. In the preparation of thallium activity by bombardment of gold targets, the gold was removed from the thallium in some cases by reduction with sulfur dioxide or sodium bisulfite in acid, and in others by repeated precipitations of thallic hydroxide in strong sodium hydroxide.

Sample decays were followed on scintillation counters and on thin-window proportional counters. Gamma-ray energy measurements were made on NaI(Tl) scintillation counters with gray-wedge pulse-height analysis.[4] Electron energies were analyzed with a 180° beta-ray spectrograph which had an upper energy limit of ~1200 kev. With the sources used, the resolution was approximately 1%.

IDENTIFICATION OF Tl^{195} AND Tl^{197}

Tl^{195}

Electromagnetically enriched[5] Hg^{196} was bombarded for 30 minutes with 20-Mev deuterons, the thallium activity was separated, and a series of 8 mercury milkings was carried out on the thallium source. Analysis of the

† Research carried out under the auspices of the U. S. Atomic Energy Commission.
* On temporary assignment from the Los Alamos Scientific Laboratory, Los Alamos, New Mexico.
[1] W. E. Bennett, Phys. Rev. 94, 997 (1954).
[2] Sugarman, Duffield, Friedlander, and Miller, Phys. Rev. 95, 1704 (1954).
[3] Andersson, Arbman, Bergström, and Wapstra, Phil. Mag. 46, 70 (1955).
[4] Chase, Bernstein, and Schardt, Phys. Rev. 90, 353 (1953).
[5] Mass analysis: 196, 1.5%; 198, 7.5%; 199, 9.0%; 200, 13.2%; 201, 7.9%; 202, 25.2%; 204, 35.8%. The electromagnetically enriched mercury isotopes were borrowed from Dr. M. Goldhaber, who obtained them from the Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

decay curves of the first mercury samples gave components of half-life 9–10 hours (Hg^{195}) and 65 hours (Hg^{197}). No evidence was found for the 38-hour Hg^{195m}, this isomer, if present, was estimated to constitute less than 20% of the total Hg^{195} which had grown in, a result consistent with the low spin one would expect for the ground state of Tl^{195}. The decay of the Tl^{195} and Tl^{197}, as obtained from the activities of their mercury daughters, is plotted in Fig. 1. The Tl^{195} half-life was found to be 1.2 ± 0.1 hours.

The gamma-ray spectrum of Tl^{195} was not observed. Due to the limited enrichment of mass 196 in the mercury target material, Tl^{195} could not be produced in sufficient quantity to be distinguishable from the overwhelming background of Tl^{198m} and Tl^{198} also produced during bombardment.

Tl^{197}

From measurements of the relative amounts of the 65-hour components in the above-mentioned series of mercury sources, and from similar experiments with thallium obtained from 39-Mev alpha bombardment of gold, the Tl^{197} half-life was found to be 2.8 ± 0.2 hours. The target reactions were $Hg^{198}(d,3n)$ and $Au(\alpha,4n)$, respectively. The half-life agrees with the value cited by Andersson *et al.*,[3] who obtained their Tl^{197} from thallium bombarded with protons from a synchrocyclotron.

From Tl^{197}, as from Tl^{195}, only the ground-state mercury daughter was observed to grow in. An analysis of the decay data on mercury grown from thallium extracted from alpha-bombarded gold, where no Hg^{195} could have been produced to complicate the system, leads to an estimate of 5% as an upper limit for the fraction of Tl^{197} decaying to Hg^{197m}.

In alpha bombardments of gold, the Hg^{197} may be produced either directly by $Au(\alpha,p3n)$ or by $Au(\alpha,4n)$ followed by Tl^{197} decay. The low-energy ends of the excitation functions for these two reactions were measured by a stacked-foil experiment. The experiment involved two bombardments, in each of which five 0.5-mil gold foils were stacked and exposed to a beam of 39-Mev alpha particles impinging normal to the plane of the foils. The foils from the first bombardment were allowed to stand until the Tl^{197} had decayed to Hg^{197} and were then analyzed for mercury activity; the foils from the second bombardment were dissolved and cleaned of mercury activity immediately, then were allowed to stand until the remaining Tl^{197} had decayed and finally were analyzed and counted like the first set. The results, calculated in terms of the two reactions by which the Hg^{197} was produced, are plotted in Fig. 2.

RADIATION MEASUREMENTS

Electron spectra were measured on three thallium sources: the first was separated from gold bombarded with the full-energy (39-Mev) alpha beam, the second from gold covered with sufficient aluminum absorber to drop the maximum alpha energy to 34 Mev, and the third from mercury-copper bombarded with 20-Mev deuterons. The three series of electron spectrograms

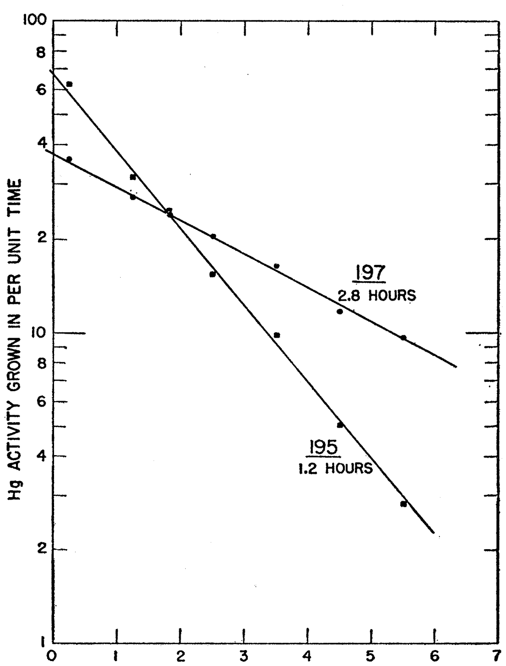

FIG. 1. Decay of Tl^{195} and Tl^{197}, measured in terms of mercury daughter activities in successive milkings from the thallium source.

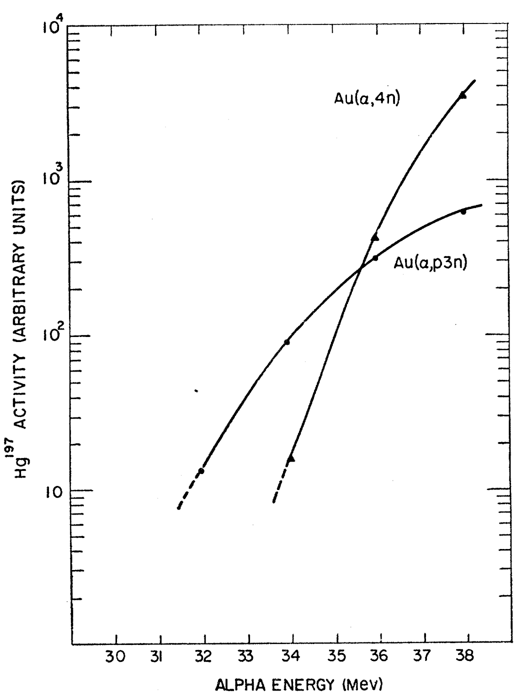

FIG. 2. Excitation functions of reactions leading to Hg^{197}.

obtained from these sources will be referred to as Au^{39}, Au^{34}, and Hg^{20}, respectively. The choice of alpha energy for the gold bombardments was based on the excitation function shown in Fig. 2; only the higher-energy bombardment should have produced significant amounts of Tl^{197}. The third source, although it contained a larger variety of thallium activities, had a more favorable Tl^{197}/Tl^{198} ratio than the first. Due to the necessity of limiting the deuteron beam strength on the mercury-copper target, this source was not as strong as those from the gold target, and measurement of conversion lines was limited to the region $\lesssim 550$ kev. Half-life estimates of most of the lines were made by visual comparison of their intensities in successive exposures; the first three exposures in each of the first two runs were taken for periods of 160 minutes, and the remainder at intervals up to 24 hours.

The gamma spectra of each of the gold-plus-alpha sources and of the thallium sources prepared by deuteron bombardment of enriched Hg^{196} and Hg^{198} were examined with a scintillation spectrometer.

Energy calibrations were based on gamma energies reported by Bergström *et al.*[6] for the isotopes Tl^{198} through Tl^{202}.

RESULTS

Tl^{197}

The report by Andersson *et al.*[3] assigns to the decay of Tl^{197} gamma transitions of energy 134, 152, 174, 434, and 611 kev, plus others, which they list as uncertain, of 269, 583, 588, and 637 kev, all measured from their conversion lines. Four of these transitions were recognized in the present work. The K and L lines of a 152.6 ± 0.5-kev gamma of half-life $\sim$3 hours were found in the spectrogram series Au^{39} and Hg^{20}, and are thus clearly assignable to Tl^{197}. A very weak electron line observed at 350 kev corresponds to the K line of the 434-kev gamma, but its low intensity precluded a half-life estimate. Also found were K, L, and probably M lines of relatively strong transitions of energies 586 and 635 kev, both with K–L energy differences characteristic of conversion in mercury and both of half-life $\sim$2 hours. However, the presence of these lines in approximately equal intensities in the spectrogram series Au^{39} and Au^{34} indicates that these transitions, which presumably correspond to the 583- or 588- and 637-kev gamma rays previously reported, belong to Tl^{198m} rather than Tl^{197}.

The presence of the L, M, and N lines of the 77-kev gamma ray and the K-line of the 191-kev gamma ray of Hg^{197} in the long-exposure spectrograms of the Au^{39} and Hg^{20} series confirmed the earlier presence of the parent Tl^{197} in these sources. From a comparison of the intensity of the K line of the 153-kev transition in the first Hg^{20} spectrogram with the intensity of the $L_I L_{II}$ doublet of the 77-kev transition in a later spectrogram of the series, together with calculations involving L-shell conversion coefficients[7,8] and growth and decay times of the Hg^{197} and Tl^{197}, it is estimated that each Tl^{197} disintegration gives $\sim$0.17 K electrons from the 153-kev transition. If this transition is magnetic dipole, as reported,[3] it may then be calculated that $\sim$27% of the Tl^{197} disintegrations go through the 153-kev level.

Of the thallium sources examined by scintillation spectrometry, the one prepared by 20-Mev deuteron bombardment of enriched (79%) Hg^{198} had the best Tl^{197}/Tl^{198} ratio. Gamma scintillation spectrograms of this source showed, in addition to the Tl^{198m}/Tl^{198} spectrum, a peak at 152–155 kev, the shorter-lived component of which is presumed to belong to Tl^{197}. In general, the strength and complexity of the gamma background from Tl^{198m} and Tl^{198} were such as to overwhelm any weaker Tl^{197} gamma rays.

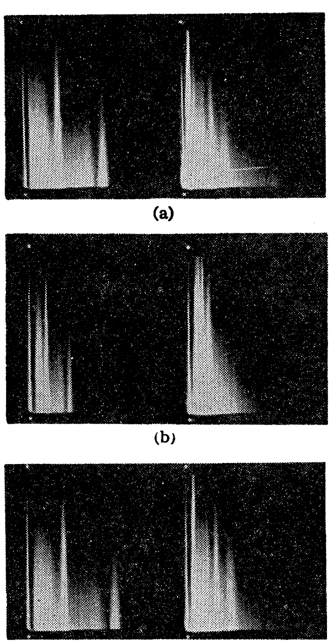

FIG. 3. Gray-wedge spectrograms of thallium sources. (Na^{22} standards are shown on left.) (a) Source Au^{39}, 4.5 hours after bombardment; (b) Same as (a), lower gain; (c) Source Hg^{20}, 4.25 hours after bombardment.

Tl^{198m}

The electron lines found in the spectrograms of the series Au^{39} and Au^{34} or in Au^{34} alone, i.e., restricted to thallium isotopes of mass 198 or higher, were classified according to the following half-lives: $\sim$2 hours (Tl^{198m}), $\sim$6 hours (Tl^{198} and Tl^{199}), and $\sim$27 hours (Tl^{200}). In the 2-hour group there were found, besides the lines of the known 260.7- and 282.4-kev gamma transitions, a considerable number of lines of lower intensity. The strongest of the latter corresponded to gamma rays of

[6] Bergström, Hill, and DePasquali, Phys. Rev. **92**, 918 (1955).

[7] Huber, Humbel, Schneider, de-Shalit, and Zunti, Helv. Phys. Acta **24**, 127 (1951).

[8] J. W. Mihelich and A. de-Shalit, Phys. Rev. **91**, 78 (1953).

635, 586, and 442 kev, all converted in mercury. The 635-kev transition had a K/L ratio of $\sim$6 and its K electrons were estimated to be roughly 0.08 as abundant as those of the 260.7-kev transition. For the 586- and 442-kev transitions, the corresponding numbers were $K/L\sim 8$ and $N_K(586)/N_K(260.7)\approx 0.03$, and $K/L\sim 8$ and $N_K(442)/N_K(260.7)\approx 0.02$, respectively. In addition, a moderately strong line at 436 kev can probably be assigned as the K line of a 519-kev gamma ray; the L_{I} or L_{II} conversion electrons, which would have had an energy near 504 kev, could not be distinguished above the background of the stronger 503-kev K conversion electrons of the 586-kev gamma ray. Other electron lines at 292 (doublet ?), 308, 339, 415, 458, 467, 642, and possibly 753 and 816 kev were too weak for classification but appear to belong to the 2-hour group. A number of the lines found here are probably identical with some of the weak lines listed as unclassified in the $Tl^{198-202}$ study of Bergstrom *et al.*

An examination of the gray-wedge scintillation spectrograms, examples of which are reproduced in Fig. 3, showed prominent photo-peaks corresponding to gamma rays of 155, 225, 282, 412, and 590–645 kev, plus weaker peaks at 510, $\sim$1075, $\sim$1230 and $\sim$1440 kev; the 282- and 412-kev gammas were used as internal energy standards. The 155-kev peak had decay components slightly shorter-lived than and slightly longer-lived than the 412-kev peak; the former is probably from a combination of the 153-kev gamma of Tl^{197} and the 158-kev gamma of Tl^{199}. The 225-kev photopeak may be identified with a 227-kev gamma ray deduced from the electron spectrograms and assigned to Tl^{198}, although the 208- and 247-kev gammas of Tl^{199} are also possible contributors. The 590–645-kev peak was too complex for unequivocal analysis. Its principal components decayed at the same rate as the 282-kev peak, and presumably correspond to the 586- and 635-kev gamma rays of Tl^{198m}.

The observation that the 442-, 586-, and 635-kev gamma rays are converted in mercury shows that Tl^{198m} decays in part by electron capture. Although the ratio of electron capture to isomeric transition in Tl^{198m} could not be determined without a knowledge of the decay schemes of Tl^{198m} and Tl^{198}, its approximate magnitude was deduced from gamma-scintillation measurements.

From the gray-wedge scintillation spectrograms of other $Tl^{198m}-Tl^{198}$ sources, each member of the 590–645 peak (here considered as only a doublet) was observed to be $\sim$1.1 times as abundant as the 282-kev gamma rays. Taking now for the 282-kev transition a total conversion coefficient of 0.27 from the data of Passell *et al.*,[9] and for the 635-kev transition a total conversion coefficient of 0.016 (supposing it to be $E2$), one finds that the 635- and 282-kev transitions occur in the ratio $\sim$0.9:1. The choice of $M1$ or $M2$ for the higher energy gamma ray would raise the ratio only slightly. Since the 635- and 586-kev gamma rays are the strongest gamma rays observed to be associated with the electron capture branch of Tl^{198m}, one may conjecture that the E.C./I.T. ratio for this isomer lies somewhere between about 1 and 2, depending on whether the two transitions represent a cascade or alternative decay paths. In either case, there remains the problem of disposal of angular momentum in the electron capture branch. If the Tl^{198m} has a spin of $\geqslant 8$, as postulated by Passell *et al.*, the excited state of Hg^{198} to which it decays must have a spin of at least 7; putting the 635-kev gamma ray in cascade with the 412-kev gamma ray and assigning to the former a multipolarity of 2 (probably the largest value consistent with its K conversion intensity and K/L ratio), one must still account for 3 units of angular momentum. In view of the apparent absence of unaccounted-for strong conversion lines in the electron spectrum, it appears that

[9] Passell, Michel, and Bergström, Phys. Rev. **95**, 999 (1954).

TABLE I. Internal conversion electron lines of Tl^{197}, Tl^{198m}, and Tl^{198}.

Electron energy (kev)	Intensity estimate	Assignment		Remarks
$T_{\frac{1}{2}}\sim 2$ hr:				
(816)	very weak			
(753)	very weak			
642	very weak			
633	very weak	$635-M$	Tl^{198m}	
621	medium weak	$635-L_{\mathrm{I,II}}$	Tl^{198m}	
585	very weak			
572	weak	$586-L_{\mathrm{I,II}}$	Tl^{198m}	
552	medium	$635-K$	Tl^{198m}	$K/L\sim 6$. Conv. Hg
503	medium	$586-K$	Tl^{198m}	$K/L\sim 8$. Conv. Hg
467	very weak			
458	weak	$543-K$(?)		
436	medium			
428	weak	$442-L$	Tl^{198m}	
358.5	medium weak	$442-K$	Tl^{198m}	$K/L\sim 8$. Conv. Hg
339	weak			
308	weak			
292	very weak			
279	medium weak	$282.4-M$	Tl^{198m}	
267.1[a]	medium	$282.4-L_{\mathrm{I}}$	Tl^{198m}	
260	weak	$260.7-N$	Tl^{198m}	
257.5	strong	$260.7-M$	Tl^{198m}	
248.1[a]	very strong	$260.7-L_{\mathrm{III}}$	Tl^{198m}	
245.4[a]	very strong	$260.7-L_{\mathrm{I}}$	Tl^{198m}	
(221)	very weak			
196.9[a]	very strong	$282.4-K$	Tl^{198m}	
175.2[a]	very strong	$260.7-K$	Tl^{198m}	
$T_{\frac{1}{2}}\sim 3$ hr:				
342	weak	$425-K$(?)	Tl^{196}(?)	
(350)	very weak	$433-K$(?)	Tl^{197}(?)	
138.1	weak	$152.6-L_{\mathrm{I,II}}$	Tl^{197}	
69.6	strong	$152.7-K$	Tl^{197}	
$T_{\frac{1}{2}}\sim 6$ hr:[b]				
1115	weak	$1198-K$	Tl^{198}	
(924)	very weak			
(715)	very weak			
661	weak	$675-L$	Tl^{198}	
592	weak	$675-K$	Tl^{198}	
(512)	very weak			
408	medium	$411.7-M$	Tl^{198}	
397.5[c]	medium	$411.7-L_{\mathrm{II}}$	Tl^{198}	
396.9[c]	medium	$411.7-L_{\mathrm{I}}$	Tl^{198}	
328.6[c]	medium strong	$411.7-K$	Tl^{198}	
211	weak	$226-L$(?)	Tl^{198}(?)	
200	weak	$283-K$	Tl^{198}	
143.6	medium	$226.7-K$(?)	Tl^{198}(?)	
132	weak			
111	medium weak	$194-K$	Tl^{198}, Tl^{198m}	Has short-lived component

[a] These lines were used as energy standards, computed from gamma transition energies reported by Bergström *et al.*, reference 6.
[b] Conversion lines corresponding to gamma rays of energies 491, 455, 333, 247, 208, and 158 kev, assigned to Tl^{199} by Bergström *et al.*, have been omitted from this list.
[c] These lines were used as energy standards, computed from the gamma transition energy reported by A. Hedgran and D. Lind, Arkiv Fysik **5**, 177 (1952).

either the 586-, 635-, and 412-kev gamma rays are in cascade, or the missing transition has an energy in the region ⩽45 kev or ⩾1250 kev, outside the range of the electron spectrograph.

Tl^{198}

The only conversion lines in the 5- to 7-hour category not already reported by Bergström *et al.*, were observed at 143.6, 211, 715, 924, and 1115 kev. The first two appear to be K and L conversion lines of a 226.7-kev gamma transition. Of the remainder, which are presumably K conversion lines, the 1115-kev line was the most intense; from a rough comparison of line densities, it was estimated that the 1115-kev electrons are about half as abundant as the K conversion electrons of the well-known 675-kev gamma ray. The complex high-energy gamma spectrum observed in the scintillation measurements is assigned in part to the decay of Tl^{198}. Of the 1075-, 1230-, and 1440-kev peaks previously mentioned, the first may be identified as the known 1086-kev gamma ray of Tl^{198} and the second probably corresponds to the gamma ray associated with the 1115-kev conversion electrons.

The beta-spectrograph data are summarized in Table I.

ACKNOWLEDGMENTS

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Slow Neutron Resonances in Rhenium*

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The BNL crystal spectrometer has been used to investigate the total cross section of rhenium for neutrons of energy 1 ev to 13 ev. Reasonances were detected at 2.156, 4.416, 5.90, 7.2, 11.1, 11.9, and 12.8 ev. Two resonances were analyzed to obtain the parameters of the Breit-Wigner single-level formula. The radiation widths measured in this experiment are in agreement with the general trend in radiation widths near $A=185$. A new resonance was observed at 11.9 ev.

INTRODUCTION

BECAUSE of interest in the dependence of radiation widths of neutron resonances on atomic weight of target,[1–5] the resonances of rhenium have been remeasured in order to determine the dependence on atomic number in the minimum preceding the peak at $A=100$.[3,5] For the resonances in the energy interval, 1 ev to 12 ev, which have been analyzed in this experiment, the resolution is of order 10% of the observed widths.

Many of the recent measurements of radiation widths, including those of rhenium, have experimental error of order 20%. With the resolution of the BNL crystal spectrometer, it was possible to reduce the experimental uncertainty to a few percent. However, only two resonances could be analyzed in this energy region and they occur in different isotopes. The results therefore are not average values as are some of those that appear in the compilations of references 1 and 5. On the other hand, in the isotopes in which more than one resonance is amenable to analysis, the deviation from their mean is smaller than the experimental error in the individual measurements.[1,3] An exception to this statement is found in europium and indium,[2] where the measurements suggest two distinct values for Γ_γ.

EXPERIMENTAL DESCRIPTION

Powdered metallic rhenium of high purity was dissolved in D_2O by treatment with concentrated hydrogen

TABLE I. Resonance parameter for Re.

Isotope	185	187
E_0(ev)	2.156±0.004	4.416±0.008
$\sigma_0\Gamma$(ev barns)	(7.25±0.07)×10^2	69.8±0.7
σ_0(barns)	(1.23±0.02)×10^4	(1.56±0.05)×10^3
Γ(ev)	0.0590±0.0006	0.045±0.001
Γ_γ(ev)	0.0557±0.0006	0.045±0.001
$g\Gamma_n$(ev)	0.00330±0.00005	0.00032±0.00001
$\sigma_0\Gamma^2$(ev^2 barns)	42.8±1.2[a]	
$\sigma_0\Gamma^2$(ev^2 barns)	41.6±1.2[b]	

[a] Central analysis.
[b] Wing analysis.

* Research performed under contract with the U. S. Atomic Energy Commission.

[1] D. J. Hughes and J. A. Harvey, Nature 173, 942 (1954).
[2] H. H. Landon and V. L. Sailor, Phys. Rev. 98, 1267 (1955).
[3] J. S. Levin and D. J. Hughes, Phys. Rev. 98, 1161(A) (1955).
[4] A. Stolovy and J. A. Harvey, Phys. Rev. 99, 611(A) (1955).
[5] J. S. Levin, thesis, Cornell University, 1955 (unpublished).

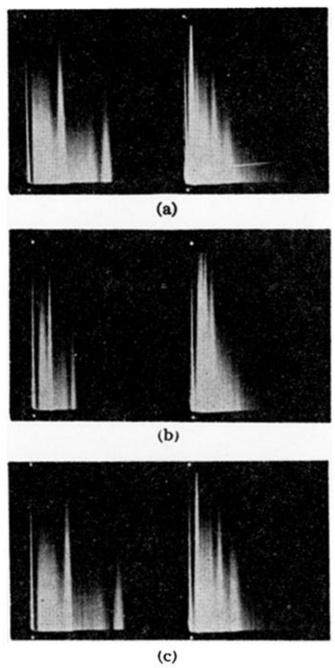

FIG. 3. Gray-wedge spectrograms of thallium sources. (Na^{22} standards are shown on left.) (a) Source Au^{39}, 4.5 hours after bombardment; (b) Same as (a), lower gain; (c) Source Hg^{20}, 4.25 hours after bombardment.