The Decay of Au^{197m}†

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The conversion electron spectra of Hg^{197m} and Au^{197m} have been re-examined. The 7.4-sec lifetime of Au^{197m} is found to be determined by a previously unresolved $E3$ transition of 130.2 kev energy, whose transition probability is 3×10^{-3} that expected on the Weisskopf estimate. The multipolarity of a "prompt" 77.4-kev transition in Au^{197} is found to be $M1+E2$ (the ratio of magnetic to electric transitions being 1.5). A consistent decay scheme is presented. Some systematics of L sub-shell conversion are discussed.

INTRODUCTION

PREVIOUS investigations[1] of the decay of Hg^{197m} have shown that the 23-hr level in Hg decays via a 164-kev $M4$ transition, followed by a 133-kev $E2$. It was also shown that the 23-hr level has a 3 percent K capture branch to a 7.4-sec level in Au^{197}. This level was thought to be depopulated by a 275-kev transition, possibly $M3$, followed by a 191-kev $M1$ and a 77-kev transition of low multipole order. There were 98.8 percent and 1.2 percent K capture branches from the 65-hr ground state of Hg to the 77- and 268-kev levels of Au, respectively.

However, certain discrepancies remained. First of all, the experimental K conversion coefficient (0.5) for the 275-kev transition was not in accord with the theoretical value[2] for an $M3$ (3.0). In addition, with this scheme, the 268-kev level had to have a spin of $5/2-$, thus making the 1.2 percent electron capture branch from the ground state of Hg^{197} inconsistent, since the levels in Hg are believed to be $i_{13/2}$, $f_{5/2}$, and $p_{1/2}$. It had been shown by Huber *et al.*[1] that the 191- and 77-kev transitions were in coincidence. Also, by a coincidence measurement between the supposed 191-K electron line (detected in a lens spectrometer) and the more energetic conversion electrons (those of the 275-kev γ-ray), the value of the supposed K conversion coefficient was determined to be 1.7. One purpose of this paper is to point out that the lifetime-determining transition is not the one of 275 kev, but a hitherto unresolved γ-ray transition of multipole order $E3$, and that in the experiment quoted above, the coincidence measurement was made between the L conversion electrons of the 130.2-kev $E3$ transition and the 275-kev transition conversion lines. The 191-K and 130-L electron lines differ by only 5 kev. In addition, we have determined the multipolarity of the 77-kev transition by a study of the L conversion ratios, and have redetermined the K conversion coefficient of the 275-kev transition. Energies of the electron lines have been measured with high resolution spectrographs with somewhat greater accuracy than before. A consistent decay scheme is presented. Some systematics of L conversion are discussed.

TABLE I. Conversion electron lines observed for γ-ray transitions in $Hg^{197m}-Au^{197m}$.

Transition energy (kev)	Conversion electrons observed
77.4	L_I, L_{II}, L_{III}, M_I, M_{II}, M_{III}, N
130.2	K, L_{II}, L_{III}, M_{II}
134.0	K, L_I, L_{II}, L_{III}, M_{II}, M_{III}, N
165.3	K, L_I, L_{II}, L_{III}, M_I, M_{III}, N
191.6	K, L_I, M
279.3	K, L_I

EXPERIMENTAL PROCEDURE AND RESULTS

The radioactive sources were prepared in the Princeton cyclotron by the $Au^{197}(p,n)Hg^{197}$ reaction. The radioactive Hg was distilled, in vacuum onto thin Au foils of proper dimensions ($\sim$2 cm$\times$0.3 mm), thus forming a source which was almost weightless. The low energy electron lines show some broadening, due probably to the diffusion of Hg into the Au. The electron lines were photographed in permanent magnet 180° spectrographs. Figure 1 is a reproduction of one of the spectrograms. This film had been deliberately overexposed on the lower energy end. Table I lists the conversion lines observed for each transition. Some of the energies differ slightly from the first published ones.

Recent measurements by Cork *et al.*[3] of the transition energies observed in a source of Hg^{197m} yield the following values: 134.3, 165.4, 191.4, and 77.6 kev. The transition energies observed[4] in a source of Pt^{197} (17.4 hr) are 191.2 and 77.4 kev.

All the L components are resolved in the higher intensity transitions, as are some of the M sub-shell

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[1] Huber, Humbel, Schneider, deShalit, and Zunti, Helv. Phys. Acta **XXIV**, 127 (1951); M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

[2] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[3] Cork, Martin, LeBlanc, and Branyan, Phys. Rev. **85**, 386 (1952).

[4] Cork, LeBlanc, Stumpf, and Nester, Phys. Rev. **86**, 415 (1952).

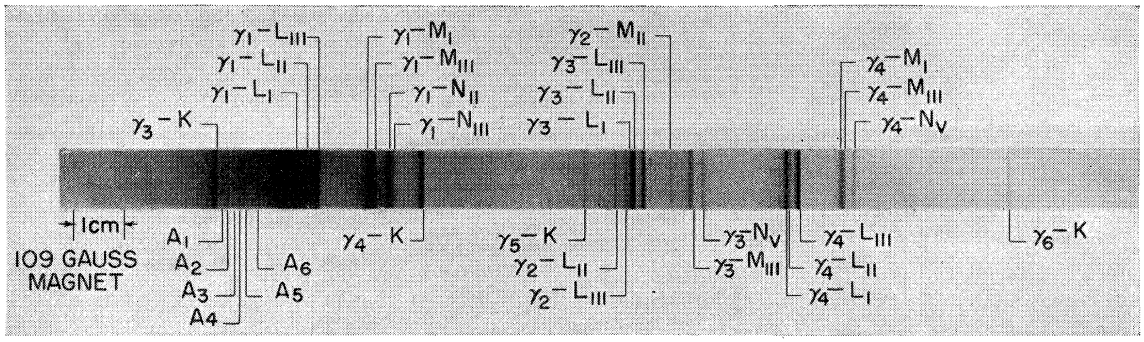

FIG. 1. Conversion electron spectrum of Hg^{197m}—Au^{197m}. The transition energies in kev are as follows: $\gamma_1=77.4$ (Au); $\gamma_2=130.2$ (Au); $\gamma_3=134.0$ (Hg); $\gamma_4=165.3$ (Hg); $\gamma_5=191.6$ (Au); $\gamma_6=279.3$ (Au). A_1 to A_6, which are the Auger lines (L_I, $L_I\rightarrow K_I$, ∞ etc.) arising from the internal conversion of K series radiations have been previously reported [J. W. Mihelich, Phys. Rev. **88**, 415 (1952)].

electrons. Of particular interest in this study are the K, L_{II} and L_{III}, and M lines of the 130.2-kev transition, and the three L lines of the 77.4-kev transition. Previous investigations were performed with spectrometers of insufficient resolving power to separate the comparatively weak conversion lines of the 130.2-kev transition from the conversion lines of the 134-kev transition. Evidence presented below indicates that the 130.2-kev transition is of $E3$ multipolarity and determines the lifetime of the 7.4-sec level.

Photometry of the spectrogram showed that the K/L ratio of the 130-kev transition was less than $\frac{1}{3}$ of the K/L ratio for the 134-kev $E2$ transition. The electrons were analyzed in a Siegbahn-type[5] spectrometer at Princeton in order to obtain more accurate intensity data. It was found that the $K:L:M$ ratio was 1:7.5:3.6, in good agreement with the K/L ratio expected for an $E3$ transition.

As mentioned above, in the coincidence experiment of Huber, etc., what was obtained was $\kappa_L^{130}=N_{e_L}^{130}/(N_{e_{\text{total}}}^{130}+N_\gamma^{130})$ rather than κ_K^{191} as was supposed. Since $\kappa_T^{130}=N_{e_{\text{total}}}/N_T\leqq 0.96$, we find that $\alpha_{\text{total}}^{130}\leqq 24$. Using the electron intensity ratios as determined in the counter spectrometer, we find that $\alpha_K^{130}\leqq 2.0$. Extrapolating the values of Rose *et al.*,[2] the theoretical values obtained are: $\beta_K^1=3.4$, $\alpha_K^1=0.14$, $\alpha_K^2=0.5$, and $\alpha_K^3=1.0$. The $L_{II}:L_{III}$ conversion is characteristic of that for electric γ-transitions.[6] The conversion coefficient and K/L ratio make the assignment of $E3$ reasonable.

The following reasoning also makes the assignment of $E3$ reasonable. The 65-hr K capture in Hg^{197} is most probably first forbidden, since it involves the changing of a neutron in one shell into a proton in a neighboring shell. Energy-lifetime considerations then make it likely that the 23-hr K capture transition is also first forbidden. Since the 65-hr and 23-hr levels in Hg^{197} have different parities, the isomeric and ground levels of Au^{197} should have opposite parities. Since the transition following the 7.4-sec isomeric transition in Au is $M1$ ($\Delta I=1$, No), as will be shown later, the isomeric transition must involve a change in parity. The L conversion indicates an electric transition, and only $E3$ is consistent with the 7.4-second half-life.

The $E3$ transition is of considerable interest, since it is the first observed odd-proton $E3$ transition which may occur between single particle levels. Previously observed $E3$'s have occurred between 7/2+ and $p_{1/2}$ levels, the presence of a 7/2+ level indicating a considerable deviation from a single particle model. In the case of Cd^{111}, the $h_{11/2}\rightarrow d_{5/2}$ transition involved an odd neutron.[7,8] This fact was thought to explain the slowness of the transition. In the case of $_{79}Au^{197}$, we should expect an odd proton $h_{11/2}\rightarrow d_{5/2}$ transition. The experimental value of the transition probability is 3×10^{-3} that expected on the Weisskopf estimate.[9] This may be explained by assuming that the "$d_{5/2}$" level is really not a single particle level, but is formed by the coupling of a number of nucleons.

Information was obtained regarding the multipolarity of the 279-kev transition. It is easily seen that

$$\alpha_K(279)=\alpha_L(134)\cdot\frac{N_K(279)}{N_L(134)}\cdot\frac{N_\gamma(134)}{N_\gamma(279)}.$$

$\alpha_L(134)$ and $N_K(279)/N_L(134)$ had previously been determined accurately.[1] Hence a measure of the ratio of the numbers of these two γ-rays will allow a determination of the K conversion coefficient of the 279-kev transition. This ratio was determined with a NaI scintillation spectrometer. The value of $\alpha_K(279)$ is ~ 0.27. The theoretical values are $\beta_K^1=0.5$, $\alpha_K^2=0.08$, and $\alpha_K^3=0.20$.[2] The experimental conversion coeffi-

[5] Thanks are due Dr. E. P. Tomlinson for kindly putting his spectrometer at our disposal.

[6] J. W. Mihelich, Phys. Rev. **87**, 646 (1952).

[7] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[8] A. W. Sunyar, Phys. Rev. **83**, 864 (1951); C. L. McGinnis, Phys. Rev. **83**, 651 (1951); and S. Johansson, Phys. Rev. **79**, 896 (1950).

[9] J. M. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

TABLE II. Conversion data on 77.4-kev transition in Au^{197}.

Sub-shell	Theoretical internal conversion coefficient $E2$	$M1$	Exp. rel. int.
L_I	~0.15	3.4	1.0
L_{II}	9.5	0.25	0.45
L_{III}	8.5	0.005	0.34

cient, the short lifetime, and the K/L ratio ($\geqq 6$) indicate a magnetic dipole.

Determination of the multipolarity of the 77.4-kev transition was made by an analysis of the intensities of the three L conversion lines. The $L_I:L_{II}:L_{III}$ ratio is 1.0:0.45:0.34. Comparison of these data with the theoretical prediction of Gellman, Griffith, and Stanley[10] will make possible an estimate of the multipolarity. The absence of a measurable lifetime and the L conversion coefficient of 2.5[1] indicates that the multipolarity is $E2$, $M1$, or a mixture of both. In Table II are listed the theoretical values of Gellman *et al.* for the individual L shells. It will be recalled that these calculations are done neglecting screening and are available for only a few values of Z. Considerable uncertainty is introduced by the rather extensive interpolation required. Probably the absolute values are in considerable error, but a comparison of relative values may be fairly good.

One finds that the experimental intensity observed may be explained by a mixture of magnetic γ-rays to electric γ-rays of 7.0, or a ratio of magnetic to electric transitions of 1.5. It may be noted that a comparison of the relative L-shell conversion is a very powerful tool for detecting or measuring proportion of mixtures. In the case of $M1$ and $E2$ mixtures, the $M1$ L_I internal conversion coefficient is ~20 times larger than the value of the L_I internal conversion coefficient for $E2$. On the other hand the L_{II} and L_{III} internal conversion coefficients for $E2$ are many times those for $M1$. Hence a small admixture of either will alter the L conversion to a considerable degree. Accurate determination of such mixtures await more accurate theoretical values of the L shell internal conversion coefficients.

Figure 2 shows our proposed decay scheme. Some comments are in order concerning the choice of level assignments. The 130-kev and 279-kev transitions are in coincidence, as stated above. It had been thought that the 279-kev was followed by the 77.4-kev transition. However, Dr. H. C. Martin of Los Alamos has kindly communicated to us the information that the threshold for producing the 7.4-sec Au isomer by fast neutron scattering in Au is less than 420 kev. Hence, it seems likely that the 279-kev transition goes to the ground state. It is then likely that the upper levels in Au^{197} are $h_{11/2}$, and $d_{5/2}$, as predicted by the shell model. The ground state has been determined as $d_{3/2}$.

In previous experiments,[1] a high coincidence rate between the 279-kev conversion electrons and other conversion electrons was measured. This coincidence was 30 percent greater than would be expected for a simple two step decay (130 and 279 kev). Hence the conclusion was drawn that these two transitions were also in coincidence with the 77.4-kev transition. In the light of the inelastic scattering data, these results have to be reinterpreted. Due to the geometry of the spectrometer, coincidences between the 279-K electron and the 130-L electrons are taken at 180°. It is possible that there is sufficient angular correlation to produce the high coincidence rate at 180°.

The 191-kev γ-ray is probably in a branch separate from the 279-kev γ-ray. The K/L ratio of 6 indicates it is an $M1$. The 191- and 77.4-kev transitions are in coincidence, and the ratio of the intensity of the two transitions is a constant with time, both activities exhibiting the same growth and decay.

No crossover transitions were definitely observed via conversion electrons. One doubtful electron line of 328 kev is present. Such a line could be the K conversion line of a 409-kev $M4$ $h_{11/2} \rightarrow d_{3/2}$ crossover. It is too faint to allow any accurate measurements. An electron line

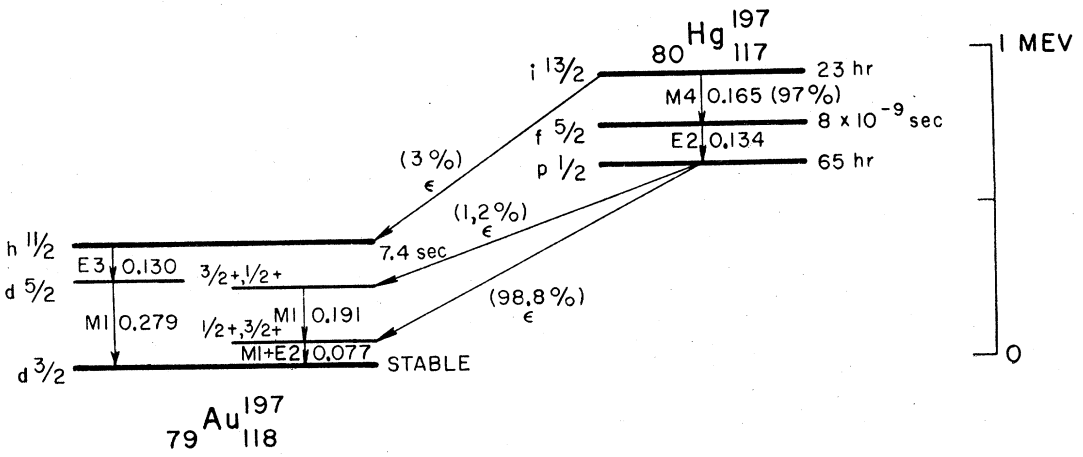

FIG. 2. Proposed decay scheme for $Hg^{197m}-Au^{197m}$.

[10] Gellman, Griffith, and Stanley, Phys. Rev. 85, 944 (1952).

(intensity roughly that of the 191-M line) of 185.8 kev is observed. It cannot be assigned to any expected transition.

The spins assigned to the 77.4- and 269-kev levels are consistent with the available data. Tentatively, we designate the two levels as $\frac{1}{2}+$ or $\frac{3}{2}+$ and $\frac{3}{2}+$ or $\frac{1}{2}+$, respectively. The $\frac{1}{2}+$ assignment for the spin of the 77.4-kev level is preferred since the 201.9-kev transition from the 279-kev ($d_{5/2}$) level to the 77.4-kev level is not observed.

Some comments may be made regarding the systematics of L shell conversion. This is the first $E3$ isomeric transition for which adequate resolution of the L lines has been obtained. The establishing of the systematics of L conversion for a transition of this multipole order, energy, and Z is invaluable for deciding multipole orders of other transitions. For the $E3$ isomeric transition of 130 kev in Au, the ($L_{\mathrm{I}}:L_{\mathrm{II}}:L_{\mathrm{III}}$) ratio is ($<0.1:\sim 2.5:1$). Taking the K-shell internal conversion coefficient as 2.0 and the K/L ratio as 0.133, the empirical values of the absolute L-shell internal conversion coefficients are ($L_{\mathrm{I}}:<0.4$; $L_{\mathrm{II}}:10.4$; $L_{\mathrm{III}}:4.2$). Rutledge, Cork, and Burson[11] have reported that for certain transitions, designated by them as $E3$, the L conversion is predominantly L_{I}. These transitions occur in Se($Z=34$). In order to establish that the conversion is in the L_{I} shell, one has to measure the $K-L$ energy difference well enough to distinguish between $K-L_{\mathrm{I}}=9.99$ kev and $K-L_{\mathrm{II}}=10.18$ kev. However, it would not be impossible that for this low Z, $E3$ L conversion occurred in the L_{I} shell. It has been pointed out[6] that for the $E2$ transition of 247 kev in Cd^{111m}, the L conversion is probably mostly in the L_{I} shell, as borne out by the calculations of Gellman *et al.*

For the 166-kev $M4$ in Hg, the L ratio is $1:<0.1:1.5$. The theoretical estimate of Tralli and Lowen[12] would predict the ratio to be $1:<0.5:\sim 2.2$.

The 134-kev $E2$ has an L ratio of 0.04:1.1:1 as compared to the theoretical value (obtained by interpolation of the data of Gellman *et al.*) of 0.1:1.4:1. Owing to the absence of theoretical data and the paucity of experimental data on $E3$ transitions, one cannot yet distinguish between $E2$ and $E3$ transitions on the basis of L ratios alone.

We are indebted to Dr. M. Goldhaber for his continued interest in this problem.

[11] Rutledge, Cork, and Burson, Phys. Rev. **86**, 775 (1952).

[12] N. Tralli and I. S. Lowen, Phys. Rev. **76**, 1541 (1949).

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Nuclear Magnetic Resonance in Thallium Compounds*

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A precision, resonance absorption measurement has been made of the ratio of the magnetic moments of Tl^{205} and Tl^{203}. The experimental ratio is $\mu^{205}/\mu^{203}=1.009838\pm 0.000001$. This result, when compared with the value observed by Berman for the ratio of hyperfine structure separations in the $^2P_{\frac{1}{2}}$ state, $\Delta\nu^{205}/\Delta\nu^{203}=1.00974\pm 0.00003$, establishes the reality of effects attributable to the finite nuclear size.

Shifts of about 0.2 percent have been found in the thallium resonance position in Tl^{+3} compared to Tl^{+1} compounds. The resonance shifts in aqueous solutions varied linearly with the anion concentration. Complex formation and interionic electronic exchange interactions, respectively, account qualitatively for the observed effects.

INTRODUCTION

THESE experiments were initiated to obtain data for comparison with theoretical values for the nuclear magnetic shielding. Thallium is one of the very few elements with two different, simple oxidation states, Tl^{+1} and Tl^{+3}, for which nuclear magnetic resonances might be observed and any differences in position compared with theory.[1,2] However, we have found the nuclear magnetic shielding to depend not only upon the oxidation state of the thallium but also upon the anions present in solution. Moreover, the magnetic shielding changes remarkably in solutions of different concentration. Here, results are given for several compounds and concentrations.

In addition, we have made a precision determination of the ratio of the Tl^{205} and Tl^{203} magnetic moments, because this ratio is important in analyzing hyperfine structure anomalies. Berman[3] reported recently a precise value for the ratio of the hyperfine structure separations of the $^2P_{\frac{1}{2}}$ ground states of Tl^{205} and Tl^{203} by the atomic beam magnetic resonance method. This ratio differs by 9 to 12 parts in 10^5 from values determined for the ratio of the magnetic moments by nuclear magnetic resonance methods.[4–7] But the combined

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† U. S. Atomic Energy Commission Predoctoral Fellow.
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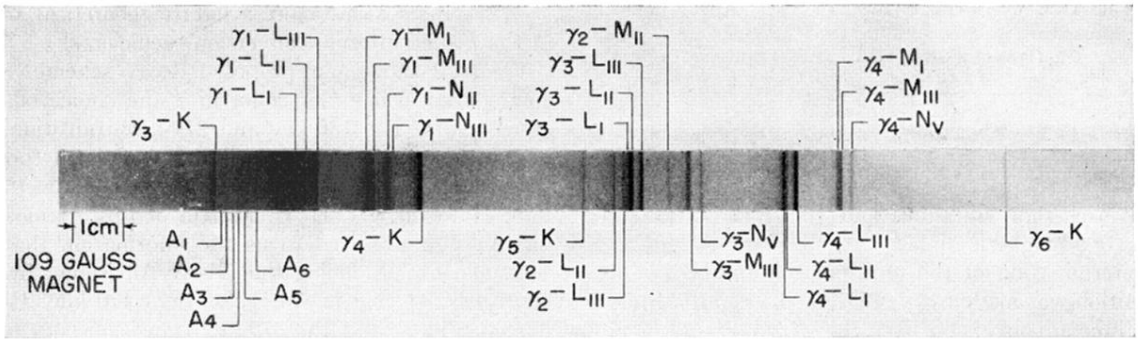
[2] W. C. Dickinson, Phys. Rev. **81**, 717 (1951).
[3] A. Berman, Phys. Rev. **86**, 1005 (1952).
[4] W. G. Proctor, Phys. Rev. **75**, 522 (1949).
[5] H. L. Poss, Phys. Rev. **75**, 600 (1949).
[6] R. E. Sheriff and D. Williams, Phys. Rev. **82**, 651 (1951).



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