Decay of Tl^{199} and Au^{199}*

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The decays of Tl^{199} to Hg^{199} and Au^{199} to Hg^{199} have been examined using gamma-ray coincidence spectrometry and angular correlation techniques. The level order as well as level spins of Hg^{199} deduced from this study support the recent work of Jung and Svedberg. The branching ratios of the electron capture transitions from Tl^{199} to six levels in Hg^{199} have also been determined. Particular emphasis was placed on obtaining the empirical parameters of importance to the measurement of the magnetic moment of the 158-keV first excited state in Hg^{199} as well as to the interpretation of the state as one member of a doublet resulting from coupling the $1/2^-$ ground state with the 2^+ core excitation. This interpretation, due to de-Shalit, implies the interrelations of some of the magnetic moments and lifetimes of the states involved. This study supports such an interpretation.

I. INTRODUCTION

In a recent paper[1] we reported on the measurement of the nuclear magnetic moment of the 158 keV, $5/2^-$, first excited state of Hg^{199} by the precession of an angular correlation by means of an external magnetic field. This state has been proposed,[2] and the measurement interpreted,[1] as the lower member of the doublet formed by the coupling of the $p_{1/2}$ ground state to the 2^+ core excitation; the other member being the 208 keV, $3/2^-$, second excited state. With this interpretation, it is possible to predict[2] the value of the magnetic moment of the $5/2^-$ state from the measured values of the ground-state nuclear magnetic moment and the $M1$ speed of the 50-keV gamma ray, "spin-flip," transition between members of the doublet. The measurements reported here are mainly concerned with those features of the level structure of Hg^{199} relevant to the measurement and prediction of the magnetic moment of the 158-keV state.

The level structure of Hg^{199} has been studied via the beta decay of Au^{199}, the electron capture of Tl^{199}, and the isomeric decay of the $13/2^+$, 526-keV state of Hg^{199}. Except for the last, no substantive gamma-ray spectroscopy has been reported. Moreover, the internal conversion measurements have led to varying results on multipolarity assignments and branching ratios. Recently, Jung and Svedberg[3] have reinvestigated the internal conversion spectrum of Hg^{199} following the decay of Tl^{199} and report the existence of a new level at 403 keV as well as new spin assignments for the 455- and 492-keV levels. The results presented here confirm the level structure and spin assignments deduced by these authors, though our branching ratios of gamma-ray transitions were found to be substantially different. The decay scheme, including the results of the present work, is presented in Fig. 1.

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[1] L. Grodzins, R. W. Bauer, and H. H. Wilson, Phys. Rev. **124**, 1897 (1961).

[2] A. de-Shalit, Phys. Rev. **122**, 1530 (1961).

[3] B. Jung and J. Svedberg, Nuclear Phys. **20**, 630 (1960).

This paper is written in three parts: The first concerns the decay of Tl^{199}, the second discusses the decay of Au^{199}, and the third represents a summary and conclusion with particular attention to the de-Shalit proposal. Section II describes the gamma-ray singles and coincidence spectra from Tl^{199} which determine the level ordering and branching ratios in Hg^{199}. The measurement of the mean life of the 158-keV state is then described and the partial lifetimes for the decay of the 208-keV state are deduced from this work together with resonance fluorescence data. Finally, the gamma-ray angular correlation studies are described. In Sec. III, the measurement of the internal conversion coefficient of the 50-keV gamma transition, which is strongly fed from the decay of Au^{199}, is described. The beta-decay branching ratios are deduced and compared with previous work.

The Tl^{199} sources were prepared by 27-MeV alpha bombardment of 0.1 mil gold foil. All sources were used *in situ*; no subsequent chemistry or heat treatment were performed on the foils. The resulting spectrum contained $\lesssim 1\%$ Hg^{200} and the source was useful for two days following bombardment. The Au^{199} sources were obtained from Oak Ridge National Laboratories as the beta decay product of Pt^{199}.

The angular correlation apparatus and fast-slow coincidence circuits were standard except for the use, when needed, of a NaI (unactivated) crystal cooled to liquid nitrogen temperature. This apparatus as well as the electromagnet used to determine the magnetic moment have been described by Stiening and Deutsch.[4]

II. THE DECAY OF Tl^{199}

A. Gamma-Ray Spectra

The gamma-ray spectrum obtained with a $1\frac{3}{4}$-in.-diam $\times$1-in.-thick NaI(Tl) crystal is shown in Fig. 2(a). The gamma-ray spectra obtained with a 3- $\times$3-in. NaI(Tl) crystal in coincidence with the photopeaks of the 158-, 208-, and 247-keV transitions are displayed in Figs. 2(b), 2(c), and 2(d); the decay scheme as

[4] R. Stiening and M. Deutsch, Phys. Rev. **121**, 1484 (1961).

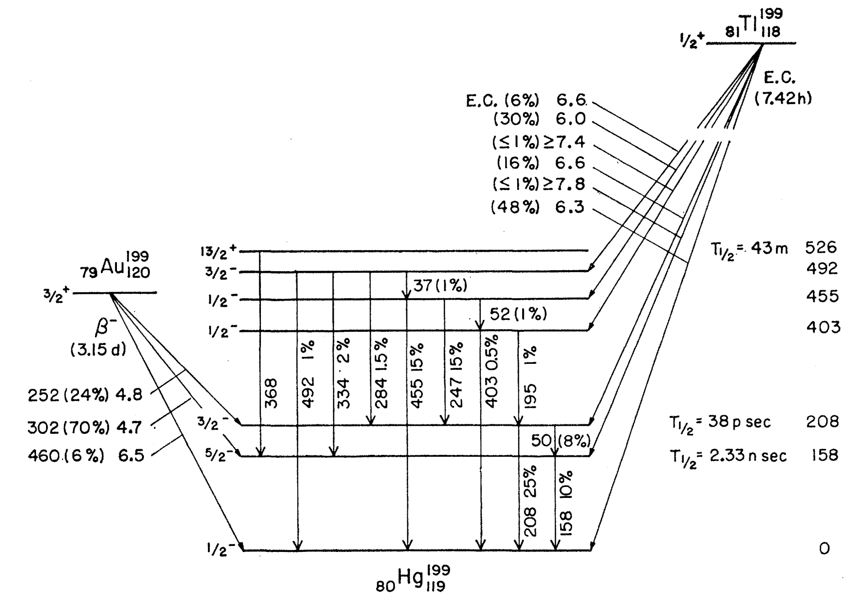

FIG. 1. Decay scheme of Tl^{199} and Au^{199}. The energy values are given in keV, the gamma intensities in percent of total disintegrations of Tl^{199}. For the beta and electron capture branches the intensities are given in percent of total disintegrations, the logft values for first-forbidden transitions. For the calculation of the logft values for the decay of Tl^{199} an excitation energy of 1500 keV was used, as determined by beta decay systematics. The half-lives of the beta unstable nuclei and excited states of Hg^{199} are included.

deduced from these multichannel analyzer spectra was further corroborated by coincidence studies using a XYZ recorder which depicts, qualitatively, the matrix of gamma-ray coincidences. The recorder used was similar in design to the coincidence sorter described by Grodzins.[5]

Although the disintegration energy of Tl^{199} is about 1.5 MeV as determined by beta decay systematics, the ratio of positron decay to electron capture is expected to be of the order of 10^{-3}. No search was made for annihilation radiation in this experiment.

The relative gamma-ray intensities deduced from the spectra of Fig. 2 are given in Table I. They have been normalized on the basis of 1000 disintegrations of Tl^{199}. The uncertainties in these intensities, based on internal consistency of the data, is about 10% for the strong transitions and 20% for the weak ones.

TABLE I. Gamma-ray intensities and multipolarities in Hg^{199}.

Transition energy[a] (keV)	Gamma-ray intensities[b]	Intensities of internal conversion electrons[c]	Internal conversion coefficient α	Multipolarities
49.8	4		19[d]	$M1$
158.4	54	48.7	0.90	$E2$
195.2	9	3.3	0.37	$M1$
208.2	119	125.1	1.05	89% $M1$ +11% $E2$[e]
247.2	93	57.5	0.62	$M1$
284.0	13	10.2	0.78	$M1$
333.9	16	4.2	0.26	95% $M1$ +5% $E2$[e]
403.4	4	3.1	0.78	$M1$
455.1	136	18.3	0.13	$M1$
491.8	10	1.3	0.13	$M1$

[a] Energy values taken from reference 3.
[b] Based on 1000 disintegrations of Tl^{199}; estimated accuracy ±10% for strong transitions, ±20% for weak transitions.
[c] Measured by Jung and Svedberg, (see reference 3) normalized so that the 158.2-keV transition has the internal conversion coefficient corresponding to a pure $E2$.
[d] Measured; see Sec. III.
[e] See Sec. IID, Gamma ray-gamma ray angular correlations.

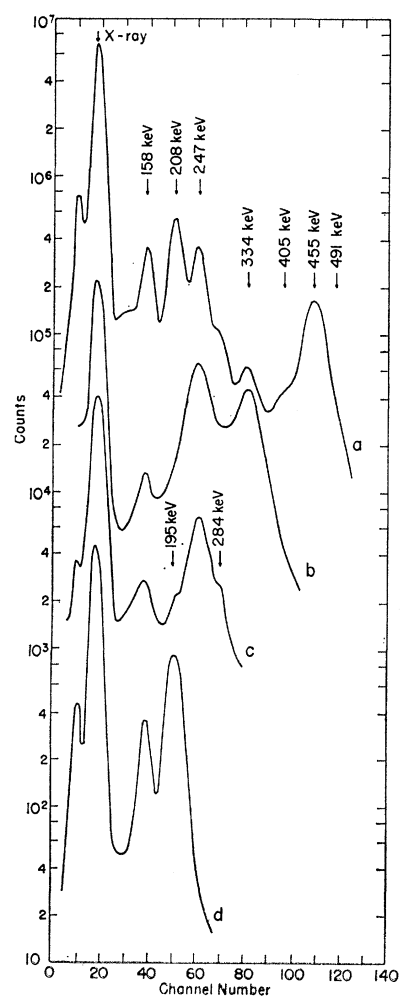

FIG. 2. Gamma-ray pulse height distributions from the decay of Tl^{199}. (a) Singles spectrum obtained with a $1\frac{3}{4}$-in. diam×1-in. thick NaI(Tl) crystal. (b) Spectrum in coincidence with 158-keV photopeak, (c) with 208-keV photopeak, (d) with 247-keV photopeak. The coincidence spectra were measured with a 3- ×3-in. NaI(Tl) crystal having slightly poorer resolution than the counter used for the singles spectrum.

[5] L. Grodzins, Rev. Sci. Instr. 26, 1208 (1955).

The relative intensities of the internal conversion electrons as determined by Jung and Svedberg[3] are given in column 3 of Table I. They have been normalized so that the 158.2-keV transition has the internal conversion coefficient corresponding to a theoretical $E2$ pole using Sliv's tables.[6] The inferred multipolarity of the transitions deduced from the columns 2 and 3 is then given in column 5 of Table I.

It will later be important to know the branching ratio of the 208- to 50-keV transitions. This ratio was deduced from the ratio of the intensities of the 208- to 158-keV gamma rays in coincidence with the 247-keV gamma ray; Fig. 2(d). The result is

$$N_\gamma(208)/N_\gamma(158)=3.07\pm0.10.$$

This ratio when corrected for the internal conversion of the 208- and 158-keV transitions yields the desired ratio

$$N_{208}/N_{50}=N_{208}/N_{158}$$
$$=(1+\alpha_{208})N_\gamma(208)/(1+\alpha_{158})N_\gamma(158)=3.24\pm0.12.$$

B. Mean Life of the 158 keV State

Bell and Graham[7] have measured the mean life of this state by delayed coincidence techniques using a Au^{199} source; their result is $\tau=3.4\pm0.3$ nsec. The state has also been observed by Coulomb excitation by Barloutaud *et al.*[8] and Davis *et al.*[9]; results for the nuclear mean life, after correcting for internal conversion, are 4.0 and 8.0 nsec, respectively.

Since the knowledge of this mean life is crucial to the measurement of the magnetic moment of the state, we have redetermined it. The technique used has been described by Stiening and Deutsch.[4] The two detectors were sodium iodide—one, 3 in. ×3 in. was thallium activated; the other, 1-in. diam ×1/4-in. thick was unactivated. The latter scintillator, maintained at liquid nitrogen temperature, exhibits a decay time approximately 10 times shorter than the thallium activated detector. The NaI crystal was coupled to a quartz-faced RCA 14-stage photomultiplier with S-20 response cathode by means of a 4-in.-long quartz light pipe in an arrangement very similar to that described by Beghian, *et al.*[10] A conventional fast-slow coincidence circuit was used in conjunction with a time to pulse height converter and multichannel analyzer. The lifetime of the 158-keV state was determined both for the 334–158 keV and the 247–158 keV transitions. The results were identical. One such time-to-pulse-height spectrum is shown in Fig. 3; the chance rate has been subtracted. Also shown on this figure is the time spectrum using the 247–208-keV cascade, whose mean life (see below) is 5.5×10^{-11} sec. Similar prompt coincidence spectra were also obtained for 350–160-keV coincidences resulting from the scattering of 511-keV gamma rays from one counter to the other. From such runs we obtain the mean life of the state $\tau(158)=3.35\pm0.12$ nsec. The result is in agreement with Bell's measurement within the errors quoted, but is much lower than the value obtained by Coulomb excitation by Davis *et al.*[9]

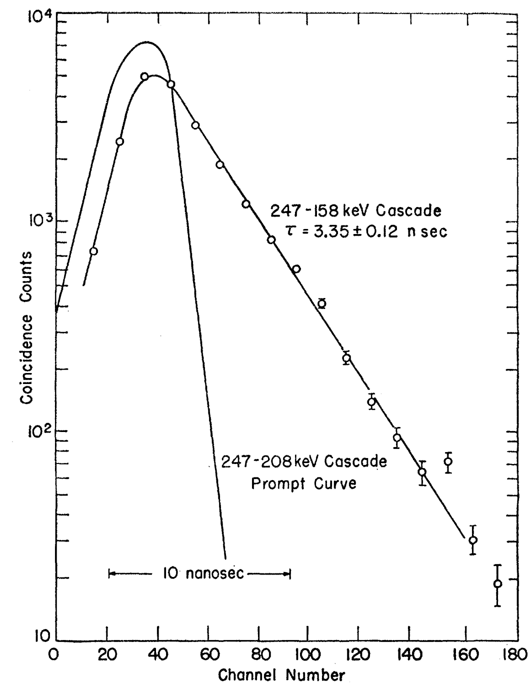

FIG. 3. Decay curve of the 158-keV state in Hg^{199} observed with the time-to-pulse-height converter using the unactivated NaI crystal for the detection of the 158-keV gamma ray. The 247–208 keV cascade illustrates the time resolution for prompt coincidences between gamma rays of about the same energy.

C. Partial Width of the 208 keV State

The lifetime of the 208-keV state has been measured by resonance fluorescence, by Coulomb excitation, and by delayed coincidence.

Graham *et al.*[11] have performed the only direct measurement of the mean life of this state, by means of the delayed coincidence technique using a Au^{199} source. Their result is $\tau=100\pm60$ psec.

The Coulomb excitation data[8,9] again show a discrepancy among themselves. When corrected for the gamma-ray mixing and branching ratios and internal

[6] L. A. Sliv and I. M. Band, *Coefficients of Internal Conversion of Gamma Radiation* (Academy of Sciences, U.S.S.R. 1956, 1958).
[7] R. E. Bell, R. C. Graham, and H. E. Petch, Phys. Rev. **84**, 380 (1951).
[8] R. Barloutaud, T. Grejebine, and M. Riou, Compt. rend. **242**, 1284 (1956).
[9] R. H. Davis, A. S. Divatia, D. A. Lind, and R. D. Moffat, Phys. Rev. **103**, 1801 (1956).
[10] L. E. Beghian, G. H. R. Kegel, and R. P. Scharenberg, Rev. Sci. Instr. **29**, 753 (1958).
[11] R. Graham, R. E. Bell, L. Yaffe, and J. Geiger, Phys. Rev. **99**, 1646 (1955).

conversion coefficients, the results for the nuclear mean life are of the order of 150 and 270 psec, respectively.

The most accurate data are those obtained by resonance fluorescence.[12,13] With the ratio of total width to partial 208-keV gamma width, $\Gamma/\Gamma_{\gamma 208}=(1+\alpha_{208})\times(1+N_{50}/N_{208})=2.65\pm0.25$, the partial gamma mean life τ_γ as determined by Metzger[12] is 120 ± 40 psec; as determined by Knapp[13] 143 ± 20 psec. This gives for the mean life of the level including conversion $\tau(\text{Metzger})=47\pm20$ psec, $\tau(\text{Knapp})=55\pm13$ psec. We adopt the value $\tau_{208}=55\pm13$ psec. The relative mean lifetimes, including internal conversion, are then $\tau_{50}(M1)=234\pm55$ psec, $\tau_{208}(M1)=73\pm17$psec, $\tau_{208}(E2)=595\pm140$ psec. Using the internal conversion coefficients given in Table I, the partial gamma-ray mean lives are $\tau_{\gamma 50}(M1)=4.46\pm1.0$ nsec, $\tau_{\gamma 208}(M1)=0.15\pm0.03$ nsec, $\tau_{\gamma 208}(E2)=1.22\pm0.27$ nsec.

D. Gamma Ray-Gamma Ray Angular Correlation

The angular correlations were obtained with two NaI(Tl) crystals, one a 3 in. ×3 in., the other a 2-×2-in. scintillator. The fast-slow coincidence circuit was the same as above but with a fixed resolving time of 35 nsec.

The gamma-ray spectra in coincidence with the 158-keV transition at 90°, 135°, and 180° are shown in Fig. 4. The 334–158-keV angular correlation is seen to

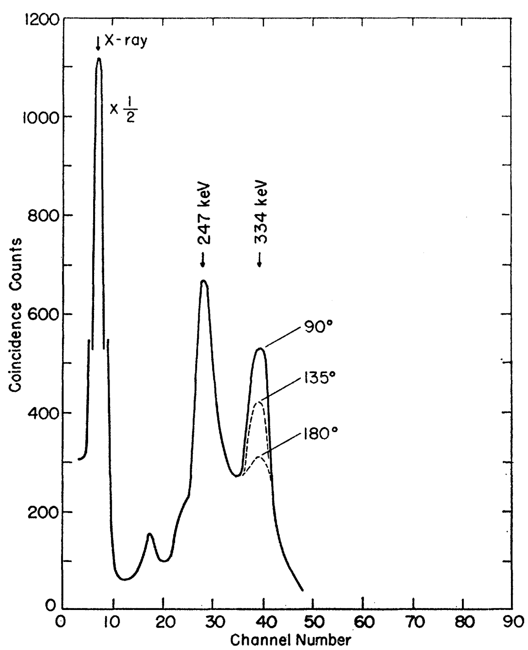

FIG. 4. Angular correlation of the coincidence spectrum gated by the 158-keV photopeak. Note the strong correlation for the 334–158 keV cascade, used in the measurement of the magnetic moment of the intermediate state.

[12] F. R. Metzger, J. Franklin Inst. **261**, 219 (1956).
[13] V. Knapp, Proc. Phys. Soc. (London) **A70**, 142 (1957).

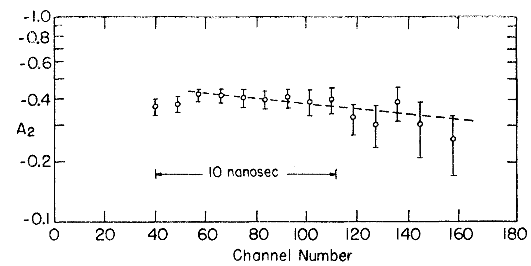

FIG. 5. Time dependence of the angular correlation of the 334–158-keV gamma cascade in Hg^{199}. The dashed line, showing 5% attenuation per mean life, is a least-squares fit to the data.

be quite large. It is given by the expansion

$$W(\theta)=1-(0.40\pm0.02)P_2(\cos\theta)+(0.01\pm0.02)P_4(\cos\theta).$$

The uncertainty is due mainly to the uncertain subtraction of the 247-keV photopeak.

The angular correlation of the 247–158-keV transition is zero within the accuracy of the experiment. The observed correlation is given by

$$W(\theta)=1+(0.009\pm0.012)P_2(\cos\theta)+(0.003\pm0.012)P_4(\cos\theta).$$

These results were obtained with a solid source upon which no chemistry was performed so that it is necessary to substantiate that there exists no appreciable attenuation of the correlation due to extra-nuclear perturbation during several mean lives of the intermediate state. This was determined by carrying out the angular correlation as a function of time on the 334–158-keV gamma cascade. In this case the cold NaI (unactivated) crystal was used as described previously. The plot of A_2 (the coefficient of $P_2(\cos\theta)$) vs time is shown in Fig. 5. The attenuation observed over a period of about 5 mean lives is found to be small (about 5% per mean life). Assuming exponential attenuation the unperturbed 334–158-keV correlation is then

$$W(\theta)=1-(0.42\pm0.02)P_2(\cos\theta)+(0.01\pm0.02)P_4(\cos\theta).$$

The angular correlation of the 334–158-keV cascade uniquely determines the spin of the 492-keV state to be 3/2. Its parity is known to be negative from its internal conversion. This result is in agreement with the work of Jung and Svedberg.[3] We also obtain, because the correlation is so large, $\delta^2=0.05\pm0.01$, uniquely, giving a 5% $E2$ admixture to the 334-keV gamma transition. The amplitude mixing ratio is, therefore, $\delta=-0.22\pm0.02$.

The angular correlation of the 247–158-keV cascade which occurs through an intermediate unobserved 50-keV transition does not uniquely determine either a spin or a mixing ratio. The result, however, is consistent with a spin and multipolarity sequence of 1/2(1)3/2(1)5/2(2)1/2.

The angular correlations of the 284–208-, 247–208-, and 195–208-keV gamma cascades are found to exhibit

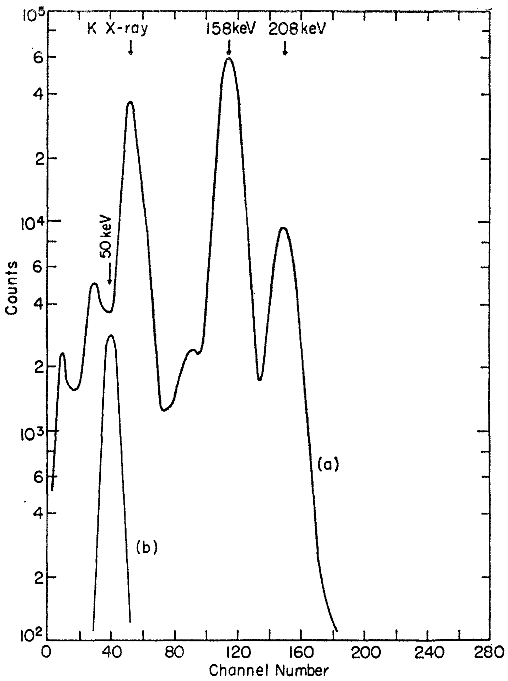

FIG. 6. Gamma-ray pulse-height distribution from the decay of Au^{199}. (a) Singles spectrum observed with a 1 3/4-in. diam $\times$1 in. thick NaI(Tl) crystal. (b) Spectrum in coincidence with 158-keV photopeak, observed with a 2 in. $\times$2 in. NaI(Tl) crystal.

no correlations within statistical accuracy of the experiment. The result for the 247–208-keV correlation is $A_2=0.00\pm0.01$ and $A_4=0.00\pm0.01$. Such a result cannot be due to a perturbation in the intermediate 208-keV state, since we have already shown that the much longer lived 158-keV state does not show a perturbation. The observed angular correlations are consistent with an amplitude mixing ratio $\delta(E2/M1)=+0.30\pm0.05$, corresponding to about an 11% $E2$ admixture, for the 208-keV gamma transition. The angular correlations will then be zero irrespective of the multipolarities of the feeding 284-, 247-, and 195-keV transitions.

III. THE DECAY OF Au^{199}

The beta decay of Au^{199} takes place to the ground and first two excited states of Hg^{199}. [The branching ratio to the 403-keV level would be expected to be small ($\approx10^{-6}$) and was not observed.] The branching ratio, R, of the beta decay to the 208-keV vs the 158-keV state was determined from the ratio of 208- to 158-keV gamma rays observed in a 1 3/4-in. diam $\times$1 in. thick NaI(Tl); see Fig. 6. The result is

$$R=0.30\pm0.03.$$

This agrees with the branching ratio obtained by Haynes and Achor[14] and by Bäckström *et al.*[15] We have not measured the intensity of the beta branch to the ground state. We have assumed Haynes' value, which is in agreement with earlier measurements by de-Shalit *et al.*,[16] to be correct for the ground state transition and have placed it in Fig. 1.

Figure 6(b) shows the coincidence spectrum observed with a 2- $\times$2-in. NaI(Tl) detector gated by the 158-keV photopeak. The 50-keV transition, not observed in the singles spectrum, is shown to be the only gamma ray in coincidence with the 158-keV gamma transition.

We are interested in obtaining the partial gamma-ray lifetime for the 50-keV transition. This requires a knowledge of the 50-keV internal conversion coefficient. To determine this coefficient, α_{50}, the 50-keV gamma–158-keV gamma coincidence rate N_{50-158} was measured in a known geometry, and compared with the 158-keV singles rate. We get

$$N_{158}/N_{50-158}=(\epsilon_{50}\Omega_{50})^{-1}(1+\alpha_{50})(N_{208}/N_{50}+1)\times[1/R+(N_{208}/N_{50}+1)^{-1}],$$

where ϵ_{50} and Ω_{50} are the efficiency and the solid angle, respectively, of the detector for 50-keV gamma rays. The branching ratio for de-excitation, including internal conversion, is $N_{208}/N_{50}=3.24\pm0.12$, as given in Sec. IIA, and the branching ratio of the beta decay of Au^{199} to the 158- and 208-keV states is $R=0.30\pm0.03$, as given above in this section. Consistent results were obtained when the solid angle Ω_{50} was fixed by a collimator in front of the 50-keV counter which masked the edges of the counter. The result is

$$1+\alpha_{50}=19\pm1$$

compared to a theoretical $M1$ coefficient of 17.

IV. SUMMARY

The decay scheme of Hg^{199} following the decays of Tl^{199} and Au^{199} has been examined. Particular attention was paid to those parameters of importance to the measurement of the magnetic moment of the 158-keV state as well as its interpretation as one member of a doublet resulting from coupling the $p_{1/2}$ ground state with the 2^+ core.

Those parameters important to the magnetic moment measurement of the $5/2^-$ state were the mean life which was determined to be

$$\tau=(3.35\pm0.12)\times10^{-9}\text{ sec},$$

the coefficients of the angular correlation of the gamma-gamma cascade proceeding through this state, and the correlation attenuation coefficient which was found to be 5% per mean life. The result of the magnetic moment measurement, using the method of precessing the 334–158-keV angular correlation in a transverse

[14] S. K. Haynes and W. T. Achor, J. phys. radium **16**, 635 (1955).
[15] G. Bäckström, O. Bergman, and J. Burde, Nuclear Phys. **7**, 263 (1958).
[16] A. de-Shalit, O. Huber, and H. Schneider, Helv. Phys. Acta **25**, 279 (1952).

magnetic field of 26 KG, was reported to be $+1.03\pm0.08$ nm (see our previous paper, reference 1).

The de-excitation branching ratios for the 208-keV level were measured. Using Knapp's value[18] for the partial gamma-ray mean life of this state,

$$\tau_\gamma = (14.3\pm2)\times10^{-11}\text{ sec},$$

we obtain a total mean life of the state, including conversion, of

$$\tau = (5.5\pm1.3)\times10^{-11}\text{ sec}$$

and a partial gamma-ray mean life for the 50-keV ($M1$) transition of

$$\tau_\gamma(50) = (4.46\pm1.0)\times10^{-9}\text{ sec}.$$

The latter value allows the prediction of the magnetic moment of the $5/2^-$ state since the ground-state magnetic moment is known. Using Eq. (1) of reference 1, we calculate a value for the magnetic moment of the 2^+ core excitation as

$$\mu_c = +0.33_{-0.25}^{+0.17}\text{ nm}.$$

According to the particle-core interpretation the magnetic moment of the $5/2^-$ state should be

$$\mu_{5/2} = +0.84_{-0.25}^{+0.17}\text{ nm}.$$

which is the sum of μ_c and the magnetic moment of the ground state μ_p. For the calculation, the ground-state moment, $\mu_p=0.504$ nm, was used as measured by Cagnac and Brossel,[17] including a diamagnetic correction of 0.96%. Figure 7 gives a graphical representation of these calculations. The solid curve is de-Shalit's prediction of the magnetic moment $\mu_{5/2}$ as a function of the partial gamma-ray mean life $\tau_\gamma(50)$. The experimentally determined values for $\mu_{5/2}$ and $\tau_\gamma(50)$, including the estimated errors, are shown in the figure. The agreement between the theoretical relation and the directly measured values is good. While the experimental uncertainties are rather large, particularly for the partial gamma-ray mean life for the 50-keV gamma transition, the agreement supports the core-particle coupling interpretation of the first and second excited states of Hg^{199}.

[17] B. Cagnac and J. Brossel, Compt. rend. **249**, 77 (1959).

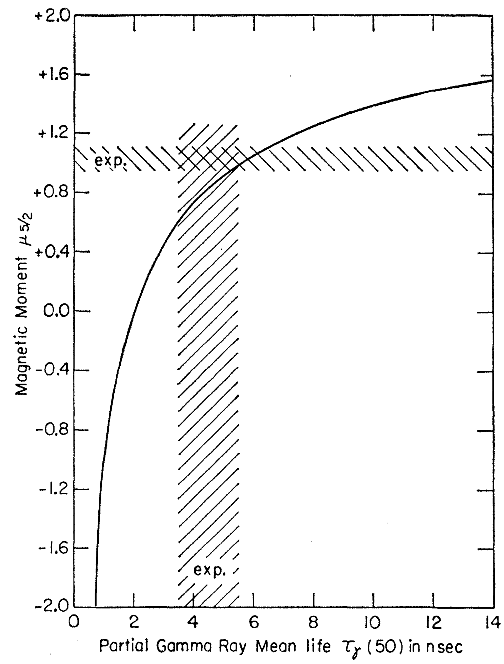

FIG. 7. The magnetic moment $\mu_{5/2}$ for the 158-keV excited state of Hg^{199} plotted vs the partial gamma-ray mean live $\tau_\gamma(50)$ of the 208-keV excited state. The solid curve is predicted by the core-particle coupling interpretation. It was calculated using the spin assignments as given in Fig. 1 and the magnetic moment of the Hg^{199} ground state as $+0.504$ nm. The experimentally determined values for $\mu_{5/2}$ and $\tau_\gamma(50)$, including the estimated errors, are shown in the figure.

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