The Disintegration Scheme of Au^{199}

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The disintegration of 3.3-day Au^{199} has been investigated using beta-ray spectrometers and counting techniques. The beta-ray spectrum was observed to be complex. Determinations were made of the conversion electron and γ-ray intensities of three γ-transitions of 50, 159, and 209 kev. A consistent disintegration scheme is given.

INTRODUCTION

THE 3.3-day beta-activity of Au^{199} was discovered by McMillan, Kamen, and Ruben.[1] In 1948, Mandeville, Scherb, and Keighton[2] reported that the beta-spectrum was simple, with an upper energy end point of 380 kev. In 1949, Meem and Maienschein[3] concluded that the beta-spectrum was simple and had an end point of 320 kev. They also observed γ-rays of 140 and 214 kev. In 1949, Beach, Peacock, and Wilkinson[4] investigated the beta- and γ-ray emission of Au^{199}. They observed a simple beta-spectrum of 320-kev end point and γ-rays corresponding to energies of 24, 52, 76, 154, 206, and 230 kev. More recent work[5] has seemed to indicate only the existence of 50-, 159-, and 209-kev γ-rays. Because of this and the interest attached to the disintegration scheme of Au^{199} in evaluating the neutron capture cross section[6] of Au^{198}, the present investigation was undertaken.

SPECTROMETER MEASUREMENTS

A. Electron Spectrum

The combined beta- and conversion electron spectrum was observed with a 180° variable field spectrometer of approximately 1.4 percent resolution. The detector was a Geiger counter having an 0.12 mg/cm² Nylon window. The source was ~0.01 mg/cm² thick and was mounted on a conducting plastic backing of 0.025 mg/cm² thickness. Sources were prepared by chemically separating gold from platinum metal which had been irradiated by neutrons in the Oak Ridge pile. The spectrum, after correction for known factors, is shown in Figs. 1 and 2.

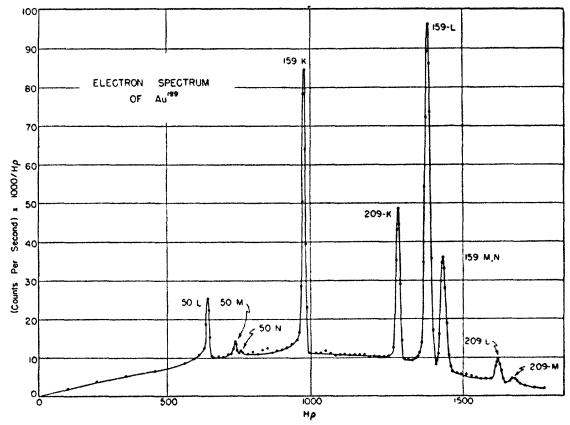

FIG. 1. Electron spectrum of Au^{199}.

Fermi plots of the beta-spectrum are shown in Figs. 3 and 4. Analysis of these plots, assuming as a first approximation allowed-shape spectra, indicates the existence of three components with upper energy end points of 250, 300, and 470 kev. Errors up to 25 kev would be possible for these energy values. The existence of the highest energy component is definite and the value of its upper energy limit is consistent with a transition to a level 159 kev below that reached by the main beta-ray component. The existence of the lowest energy component is less certain. Its existence is again supported by the value of its upper energy limit being 50 kev less than the main component.

The conversion lines corresponding to γ-transitions of 50, 159, and 209 kev confirm the experimental work of earlier observers.[4,5] The intensities of the lines were obtained by decomposing them from the beta-spectrum and then integrating the counting rates with respect to $H\rho$. These intensities are listed in Table I.

B. Photoelectron Spectrum

This spectrum was measured using a lens spectrometer of approximately 1 percent transmission and 3

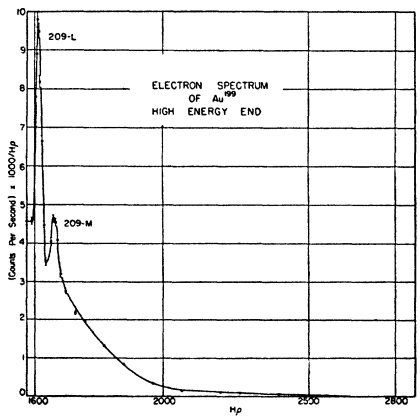

FIG. 2. Electron spectrum of Au^{199}: high energy end.

* Assisted in part by the joint program of the ONR and AEC.
[1] McMillan, Kamen, and Ruben, Phys. Rev. **52**, 375 (1937).
[2] Mandeville, Scherb, and Keighton, Phys. Rev. **74**, 601 (1948).
[3] J. L. Meem and F. Maienschein, Phys. Rev. **76**, 328 (1949).
[4] Beach, Peacock, and Wilkinson, Phys. Rev. **76**, 1585 (1949).
[5] R. D. Hill, Phys. Rev. **79**, 413 (1950).
[6] R. D. Hill and J. W. Mihelich, Phys. Rev. **79**, 275 (1950).

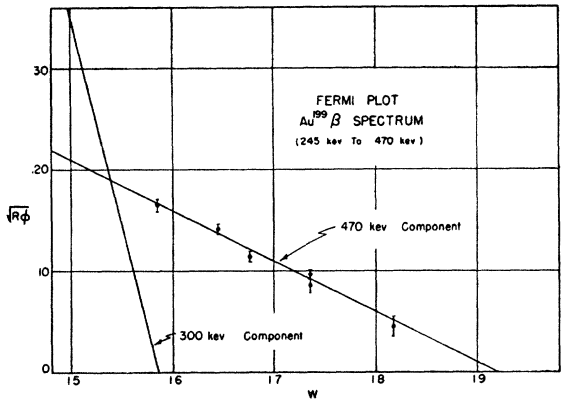

FIG. 3. Fermi plot of Au^{199} beta-spectrum (245 kev to 470 kev). [R is the normalized counting rate and $\phi=(\eta^2F)^{-1}$.]

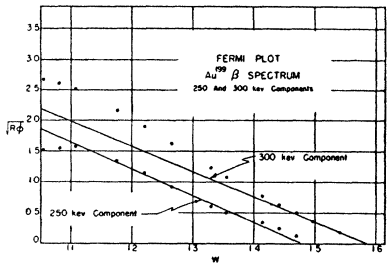

FIG. 4. Fermi plot of Au^{199} beta-spectrum (250- and 300-kev components). [R is the normalized counting rate and $\phi=(\eta^2F)^{-1}$.]

percent resolution. In order to resolve the photoelectron lines from the 159- and 209-kev γ-rays to the greatest extent possible a tin radiator was used. The spectrum obtained from a 15.7 mg/cm^2 tin radiator, with no counting rate corrections applied, is shown in Fig. 5. The lines marked K_α-L and K_β-L arise from the conversion of K x-rays of mercury in the L-shell of tin. The small peak at $H\rho=1370$ is due to a trace of source material deposited on the outside of the container. This peak is the strong 159-L conversion line, and corrections for the presence of the unshielded Au^{199} were made on the basis of its intensity.

The peak heights of the photoelectron lines were measured and corrected for known factors such as background, decay of source, and window absorption. The ratio of the peak heights of the K lines of the 159- and 209-kev γ-rays was determined to be 6.1. Using Richardson's thick target formula,[7] this ratio was then used to calculate the true ratio of the 159-kev γ-ray intensity to the 209-kev γ-ray intensity. The final value obtained was 4.2. Gamma-ray intensities are shown in Table I.

No trace of a 230-kev γ-ray, such as reported by Beach *et al.*[4] was found. The strong line interpreted by these workers as the K line of a 230-kev γ-ray can be attributed, for a lead radiator, to the L line of the 159-kev γ-ray.

ABSORPTION MEASUREMENTS

Using scintillation counters with 2-mm thick sodium iodide crystals, γ-γ-coincidences were observed. The resolving time of the coincidence circuit was 1.2×10^{-7} second. Both counters were shielded with enough aluminum so as to prevent the passage of 500-kev electrons.

An absorption curve of the γ-rays in coincidence was obtained by introducing copper absorbers in front of one counter. The curve showed that a soft γ-ray component of $\sim$50 kev was in coincidence with an x-ray of $\sim$70 kev and a γ-ray of $\sim$160 kev. Copper absorbers placed in front of both counters showed that there was no appreciable coincidence rate between γ-rays $\sim$200 kev.

The intensity of the 50-kev γ-ray relative to the 159-kev γ-ray was estimated by comparing coincidence and singles rates. With no absorber, the coincidence rate was 0.059 count per second, as compared with a singles rate of 276 counts per second. Using the solid angle and efficiency factors of the crystals, it was then estimated that the 159-kev γ-ray was $\sim$120 times more intense than the 50-kev γ-ray.

This result assumes that the lifetime of the level intermediate between the 50- and 159-kev γ-transitions is shortlived compared with the resolving time of the coincidence circuit. This point was verified by showing that the γ-γ-coincidences were as prompt, within the limits of our experiment, as the β-γ-coincidences from Au^{198}. Coincidences between beta- and γ-rays were also investigated, but no significant results of their interrelationship were obtained.

DISCUSSION

A. Energy Level Diagram

Considering the three main lines of evidence: γ-transition energy values, component beta-ray energy values, and γ-γ-coincidence observations, the most probable Hg^{199} level scheme involved in the decay of Au^{199} is one consisting of two excited levels of 159 and

TABLE I. Radiation intensities.

Radiation	(250+300)β	470β	50γ	159γ	209γ	50-L	50-M
Intensity	96.2	3.8	0.4	47.9	11.4	2.46	0.5
Radiation	50-N	159-K	159-L	159-M, N	209-K	209-L	209-M, N
Intensity	0.15	9.0	15.0	5.2	6.14	1.14	0.3

[7] H. O. W. Richardson, Proc. Phys. Soc. (London) **63A**, 234 (1950).

209 kev above the ground state. This scheme is shown in Fig. 6. It is found to lead to a consistent interpretation of the intensities of all transitions and also to assignments of spin values for the levels which are consistent with interval conversion and radiation theory.

B. Radiation Intensities

The observed radiation intensities are shown in Table I. The intensities have been normalized so that the total beta-ray intensity is 100. The γ-ray and conversion electron intensities have been related to each other and to the beta-ray intensities by assuming that the 159- and 209-kev total transition intensities equal the sum of the 250- and 300-kev beta-intensities, i.e., on the basis of the level diagram in Fig. 6.

C. Conversion Coefficients

The conversion coefficients calculated from the above intensity data are shown in Table II. Limits of errors involved in these coefficients were arrived at after

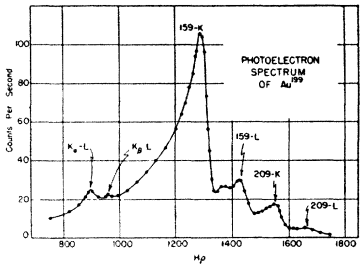

FIG. 5. Photoelectron spectrum of Au^{199}.

trying a number of decompositions of beta-spectra and conversion lines, as well as after considering the inaccuracies arising from the observations alone. The different upper and lower limits arose primarily from the decomposition process. For example, the lowest intensity limits of the lines were obtained when the continuous beta-ray spectrum was drawn as far as feasible up into the bases of the lines.

Although the divergence of the experimental conversion coefficients from the theoretical value is admittedly large, the experimental values favor the electric 2^2 assignment for the 159-kev transition, and, to be in line with this, an assignment of either 2^1 magnetic or 2^4 electric for the 209-kev transition. These assignments are also supported by a consideration of the N_K/N_L ratios for these two transitions. Although the present state of the theory does not seem to warrant an accurate comparison with experimental N_K/N_L ratios, it would appear that a value of 0.6 for the 159-kev transition is in better agreement with one of $\sim$2 for electric 2^2 than with one of $\sim$8 for electric 2^1. And for the 209-kev transition, the experimental value of $N_K/N_L=5.4$ agrees better with a value $\sim$12 for

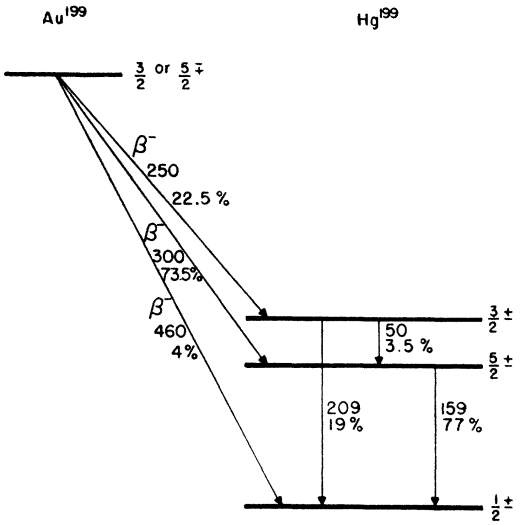

FIG. 6. Proposed disintegration scheme of Au^{199}.

magnetic 2^1 than with 0.7 or less for either electric 2^3 or 2^4.

D. Spin and Parity Assignments

The ground-state spin of Hg^{199} is known[8] to be 1/2. Thus considering the multipolarities assigned to the transitions, the 159-kev excited level must have spin 5/2, and the 209-kev excited level must have spin 3/2, both levels being of the same parity as the ground level.

On this basis, the 50-kev transition is magnetic 2^1. If the L conversion coefficient is increased from the experimental value of 6 to $\sim$10, so as to be in line with the probable values of the other K conversion coefficients, it would not seem to be inconsistent with one of $\sim$10 for the theoretical[9] L_I conversion coefficient.

The experimental branching ratio of the 209- to 50-kev γ-ray transitions is 28. The theoretical ratio, based on a $(2l+1)$ energy dependence, is 73, which is in reasonable agreement with experiment.

The logft values of 6.3 and 6.0 for the 250 and 300-kev beta-components indicate either allowed or first-forbidden transitions. The values of log$ft=7.8$ or

TABLE II. Conversion coefficients.

Transition (kev)	50	159	209
K-conversion coeff. (expt.)	—	$0.19+0.15$ -0.06	$0.54+0.8$ -0.19
K-conversion coeff. (theor.)	—	0.11, 2^1 el[a] 0.30, 2^2 el[a]	1.15, 2^1 mag[a] 0.42, 2^3 el[b] 1.18, 2^4 el[b]
L-conversion coeff. (expt.)	6	0.31	0.10

[a] J. R. Reitz, Phys. Rev. **77**, 10 (1950).
[b] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **76**, 184(A) (1949); **83**, 79 (1951).

[8] Schuler *et al.*, Z. Physik **72**, 423 (1931); **74**, 631 (1932).
[9] Gellman, Griffith, and Stanley, Phys. Rev. **80**, 866 (1950).

of $\log[(W_0^2-1)ft]=8.5$ for the 470-kev beta-component indicate either first or second forbidden, or possibly first forbidden, $\Delta I=\pm 2$. Since allowed and second-forbidden transitions involve no parity change, whereas first-forbidden transitions do, and since the parity changes of all three beta-components have to be the same, the parity of the Au^{199} ground level is probably opposite to that of the Hg^{199} levels involved. The spin of the Au^{199} level is therefore probably 3/2, but the possibility of 5/2 cannot be ruled out.

E. Final Conclusions

Hole[10] has found that the decay of Hg^{199m} contains a 159-kev γ-transition. From the half-life of 43 minutes, it would appear that the isomeric transition of 370 kev is a 2^4 magnetic transition to the 159-kev excited level. This would identify the spin of the isomeric level as 13/2 and the parity as opposite to that of the lower levels.

Another point concerns the neutron capture cross section of Au^{198}. If the intensity of the 159-K line is accepted as being present to the extent of 9 percent of the Au^{199} decays, then the cross section is estimated to be 3.8×10^4 barns.

In conclusion, it is significant to point out that if the ground state of Hg^{199} is $p_{1/2}$, as is suggested by Mayer's shell structure model,[11] then the 159-, 209-, and 529-kev excited levels are $f_{5/2}$, $p_{3/2}$, and $i_{13/2}$, respectively, all levels being consistently predicted by the model.

We are indebted to Dr. F. R. Metzger for the use of scintillation counting equipment and for his interest in the problem.

[10] N. Hole, Arkiv. f. Mat. Astron. Fys. **36A**, No. 9 (1948).

[11] M. G. Mayer, Phys. Rev. **78**, 16 (1950).

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Neutron Diffraction by the Gases N_2, CF_4, and CH_4*

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Neutron diffraction patterns for the gases nitrogen, carbon tetrafluoride, and methane are given for a neutron wavelength of 1.063A. It is found that the coherent scattering cross sections of nitrogen and fluorine are nearly equal to the total cross sections, and that the C—F bond length in carbon tetrafluoride is 1.33A. The error in a semiclassical form of the differential scattering cross section is discussed.

I. INTRODUCTION

NEUTRON diffraction by gases may be used to determine nuclear scattering cross sections and details of molecular structure. In the present paper diffraction patterns are given for nitrogen, carbon tetrafluoride, and methane. From these the bond lengths in carbon tetrafluoride, and the ratio of coherent to total scattering for nitrogen and fluorine are derived.

The present results, together with the patterns for deuterium,[1] oxygen and carbon dioxide,[2] are the only published diffraction patterns for gases. The deuterium measurements were analyzed according to the complete quantum-mechanical formulation.[3–5] Such an approach cannot be used for every gas because in many cases the necessary formulas have not been derived or because of the large amount of computation required even when the formulas are known. This theoretical complexity and the experimental requirement of large neutron fluxes are probably the reasons why the subject has not been developed previously.

The proof that a semiclassical approximation is often applicable[2,4] greatly simplifies the equations and removes one of the obstacles to structure determinations. The error in this approximation is examined below in the light of experimental and theoretical results for nitrogen, and an elementary application is made to the measurement of bond lengths in carbon tetrafluoride.

II. THEORY

In the quantum-mechanical treatment of the scattering of monoenergetic neutrons by molecules of a gas[3,4] a differential cross section is calculated for each possible molecular transition. The formulas given in reference (3) are for deuterium in the center-of-mass system, but Spiers[5,1] has derived the transformation to the laboratory system for a maxwellian distribution of molecular velocities. Similar formulas could be applied to nitrogen because 99.2 percent of the molecules are $N^{14}N^{14}$, the same type of molecule as D_2, but the greater mass and size of the nitrogen molecule would increase the amount of computation for the

* The results published in this paper were given at the Harwell Nuclear Physics Conference, Oxford, September, 1950, and are summarized in the Report on that Conference—Report A.E.R.E. G/M 68, p. 105 (September, 1950), unpublished.

† Now with Isotope Products Ltd., Oakville, Ontario.

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[3] M. Hamermesh and J. Schwinger, Phys. Rev. **69**, 145 (1946).

[4] J. A. Spiers, Phys. Rev. **75**, 1765 (1949).

[5] J. A. Spiers, National Research Council (Canada) Report CRT-417 (April, 1949). (N.R.C. No. 1940), unpublished.