Internal Conversion Coefficients in the Decay of Au^{199}

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The intensities of the γ rays and electron shell vacancies in the decay of Au^{199} have been measured by differential γ-ray spectrometry and 4π β-γ coincidence counting. These intensities, per Au^{199} disintegration, are 0.368 ± 0.011, 0.0774 ± 0.0022, and 0.0031 ± 0.0015 for the 0.158-, 0.209-, and 0.051-MeV γ rays, respectively, and 0.208 ± 0.014 and 0.176 ± 0.005 for the L- and K-shell vacancies, respectively. Internal conversion coefficients, obtained from these data and the relative conversion electron yields of Nall, Baird, and Haynes, are: in the K shell, 0.317 ± 0.013 and 0.768 ± 0.032 for the 0.158- and 0.209-MeV γ rays, respectively; and in the L shell, 0.464 ± 0.039, 0.139 ± 0.012, and 8.5 ± 4.1 for the 0.158-, 0.209-, and 0.051-MeV γ rays, respectively. These values are consistent with the internal conversion coefficients calculated by Rose. The intensities computed from the data for the 0.251-, 0.302-, and 0.460-MeV β-particle transitions are 0.189 ± 0.005, 0.680 ± 0.025, and 0.131 ± 0.025, respectively.

INTRODUCTION

THE decay of 3.15-day Au^{199} has been studied extensively by several workers.[1–5] In all of these investigations, only relative values of the electron and γ-ray intensities were determined and used in the elucidation of the decay scheme, which is shown in Fig. 1.[6] In particular, Nall, Baird, and Haynes (to be referred to as NBH) in a recent paper[4] presented a set of average values of the relative conversion electron intensities obtained from a comparison of their data with the earlier results of Cressman and Wilkinson[2] and of Bäckström *et al.*[3] It was then necessary for NBH to use theoretical internal conversion coefficients[7,8] with their conversion electron data in order to compute relative values of the γ-ray transitions in Au^{199}.

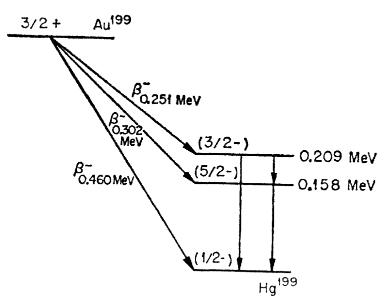

FIG. 1. The decay scheme of Au^{199}; see reference 6.

In the present investigation, the intensities of the γ rays and of the x rays arising from the internal conversion process in Au^{199} were measured on an absolute basis. By combining these data with the relative conversion electron intensities of NBH, one can then determine the internal conversion coefficients without reference to any theoretical compilation. It is then also possible to evaluate the total transition intensities in the decay of Au^{199}.

EXPERIMENTAL PROCEDURES

The Au^{199} activity was obtained from the decay of 30-min Pt^{199}, produced via the $Pt^{198}(n,\gamma)$ reaction. Natural Pt was used as the target material since no radioactive isotopes other than Au^{199} were formed by the decay of the Pt nuclides produced in the irradiation. The Pt target was dissolved in aqua regia, and the chloride concentration in the solution then adjusted to $8N$. The Au^{199} was obtained carrier free by repeatedly extracting it from the Pt solution with ethyl acetate.[9] The ethyl acetate solution was washed with $6N$ HCl several times. It was then heated to dryness, and the Au^{199} residue taken up in $3N$ HCl.

Essentially weightless samples were prepared by evaporating known aliquots of the Au^{199} solution on thin VYNS film backings; the disintegration rates of these samples were determined by the method of $4\pi\beta$-γ coincidence counting.[10] These samples were also assayed by differential gamma-ray spectrometry with a 3 in.$\times$3 in. NaI(Tl) detector so that the intensities per disintegration of the K x ray, 0.051-, 0.158-, and 0.209-MeV γ rays could be determined.[11] The value of the fluorescence yield, ω_K, used for the Hg K x ray was

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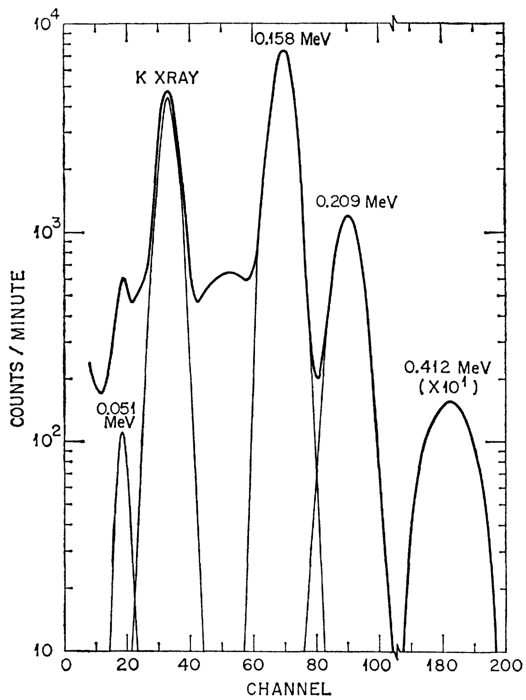

FIG. 2. The γ-ray spectrum of Au^{199}, measured with a 3 in.×3 in. NaI(Tl) detector, showing the Gaussian distributions fitted to the photopeaks. The 0.412-MeV γ ray from impurity Au^{198} is also shown.

0.952 ± 0.003.[4] The escape peak correction to the K x ray was taken to be 0.92,[12] while the escape peak losses for the 0.158- and 0.209-MeV gammas were less than 0.01. The L-x-ray yield was similarly determined by assaying the samples with a 1.5-in.-diam NaI(Tl) crystal, 3.4 mm thick; the Be window on the crystal was 0.137 g/cm^2 thick. The average fluorescence yield for the L shell, $\bar{\omega}_L$, was taken to be 0.391 ± 0.027; this value is the mean of the values reported by NBH, 0.410 ± 0.04, and by Haynes and Achor,[13] 0.371 ± 0.035.

To determine the counting rates of the x and γ rays of Au^{199}, the various photopeaks were fitted to Gaussian distributions, after corrections for the Compton distributions of the higher energy gammas were made. To facilitate this process, spectra of monoenergetic gamma rays from divers radioactive isotopes were obtained. Thus, with appropriate changes in high voltage and amplifier gain in the detector system, Cr^{51} and Hg^{203} were used to record a spectrum for the 0.209-MeV γ ray: Ce^{141} and Sc^{47}, for the 0.158-MeV γ ray; Hg^{197}, for the K x ray; and the x ray of Sr^{85}, for the Au^{199} L x ray. Comparison of the resulting net K x ray (0.072 MeV) and escape peak (0.044 MeV) of Au^{199} with those of Hg^{197} yielded a very small and approximate contribution from the Au^{199} 0.051-MeV γ ray.

[12] P. R. Bell, in *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. V, p. 155.

[13] S. K. Haynes and W. T. Achor, J. Phys. Radium **16**, 635 (1955).

The Au^{199} spectra, with the resolved Gaussian photopeaks, are shown in Figs. 2 and 3. In order to ascertain that the L- and K-x-ray yields were solely those of Au^{199}, and not due to Au^{198} produced from possible Au^{197} contamination of the Pt target, the 0.412-MeV gamma of Au^{198} was sought. As shown in Fig. 2, the intensity of this γ ray was very low; the Au^{198}/Au^{199} ratio was found to be 0.3%.

RESULTS AND DISCUSSION

The observed yields of γ rays and electron shell vacancies, in units of intensity per disintegration of Au^{199}, are listed in Table I. These quantities are the average values of data accumulated from six Au^{199} samples of different activity levels. The stated errors arise from statistical uncertainties due to the radioactive decay process, and to estimated errors encountered in the determination of the areas under the various photopeaks. In addition, the large uncertainty in the L-shell fluorescence yield contributes to the error listed for the

TABLE I. Intensities per disintegration of gamma rays and electron shell vacancies.

Photon or electron hole	Experimental results
L shell	0.208 ± 0.014
K shell	0.176 ± 0.005
0.158-MeV γ	0.368 ± 0.011
0.209-MeV γ	0.0774 ± 0.0022
0.051-MeV γ	0.0031 ± 0.0015

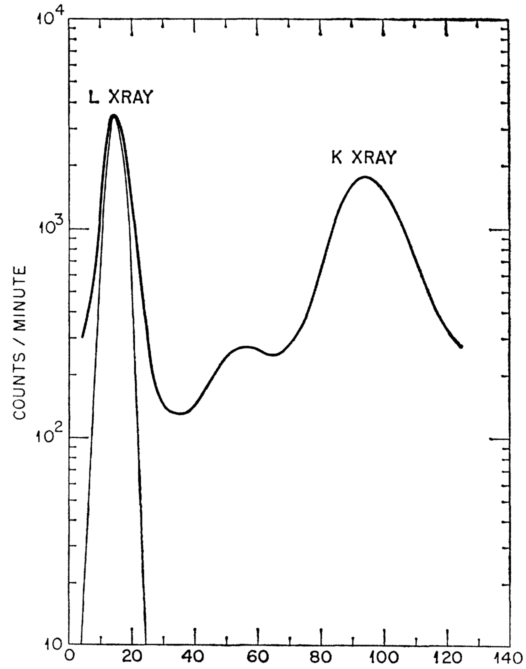

FIG. 3. The γ-ray spectrum of Au^{199}, measured with a 1.5-in.-diam NaI(Tl) crystal, 3.4 mm thick, showing the Gaussian distribution fitted to the L-x-ray photopeak.

L-shell vacancy. The ratio of 0.158 to 0.209 gammas, equal to 4.75 ± 0.19, agrees with the value of 4.52 ± 0.23 found by Cressman and Wilkinson.[2]

One may express the experimentally determined electron shell vacancies as

$$e_i=\sum_j \alpha_{ij}N_j=\sum_j e_{ij}, \quad (i=K, L;\ j=0.051, 0.158, 0.209), \quad (1)$$

where e_i is the vacancy per disintegration in the ith-electron shell, N_j is the intensity of the jth γ ray per disintegration, and α_{ij} is the ith-shell internal conversion coefficient for the jth γ ray; the vacancy per disintegration of gamma j in shell i is e_{ij}. If relative values of the e_{ij} are available, the relation

$$\alpha_{ik}N_k=\alpha_{ij}N_je_{ik}/e_{ij} \quad (2)$$

may be used to write Eq. (1) as

$$e_i=\alpha_{ij}N_j(1+\sum_{k\neq j} e_{ik}/e_{ij}), \quad (3)$$

where the sum of the relative electron vacancies, or conversion electron yields, referred to the jth γ ray, is taken over the other k gammas which are observed. Thus, by letting j in turn represent each of the γ rays of Au^{199}, one may determine the α_{ij}.

Use of the absolute γ-ray and electron shell intensities of Table I, and the relative conversion electron yields of NBH for the K- and L-electron shells, in Eq. (3) has led to the experimental values of the α_{ij} presented in Table II; no knowledge of theoretical internal conversion coefficients was necessary to obtain these results. For comparison, calculated values of the α_{ij}, obtained by interpolation from Rose's compilation,[7] are also given in Table II. The experimental quantities, within the stated uncertainties, are in good agreement with the $E2$ values for the 0.158-MeV γ ray, and with the $M1$ value for the 0.051-MeV γ ray. These results corroborate the findings of NBH, who concluded that the 0.158-MeV transition is a pure $E2$ transition, and that the fraction of $E2$ mixing in the 0.051-MeV transition is 3.3×10^{-4}. For the 0.209-MeV transition, the degree of $E2$ and $M1$ mixing may be calculated from the experimental values of the α_{ij} by solving the simultaneous equations

$$\alpha_{i209}=a_iE+b_iM, \quad (i=K, L), \quad (4)$$

where a and b are the theoretical $E2$ and $M1$ conversion coefficients of Rose, and E and M are the fractional values of the $E2$ and $M1$ transitions, respectively. The values of E and M, which should sum to unity, are respectively 0.18 ± 0.11 and 0.88 ± 0.04. These values are comparable to these found by NBH: $E=0.113\pm0.01$ and $M=0.887\pm0.01$.

Using the previously determined $\alpha_{ij}N_j$, one may proceed to calculate the total transition probabilities associated with each γ ray, since

$$T_j=(1+\alpha_{Tj})N_j,$$

where α_{Tj} is the sum over the i-electron shells of the α_{ij}. Internal conversion coefficients from NBH for the M, N, and O shells were also used in the evaluation of the α_{Tj}. The resulting total transition intensities per disintegration for the 0.158-, 0.209-, and 0.051-MeV γ rays are 0.716 ± 0.025, 0.153 ± 0.003, and 0.036 ± 0.004, respectively. Referring to the decay scheme of Fig. 1, one sees that the intensities per disingegration of the 0.251-, 0.302-, and 0.460-MeV β particles are, respectively, 0.189 ± 0.005, 0.680 ± 0.025, and 0.131 ± 0.025. The corresponding transition intensities found by Haynes and Achor[13] by directly observing the three β groups are 0.243, 0.693, and 0.064, respectively. It should be also noted that the ratio of the intensities of the 0.251- and 0.302-MeV betas, 0.278 ± 0.013, from the present work is in agreement with the values determined by NBH, 0.280 ± 0.015, and by Bauer *et al.*,[5] 0.30 ± 0.03, while the Haynes and Achor ratio is 0.35.

TABLE II. Internal conversion coefficients, α_{ij}.[a]

Electron vacancies	Relative conversion electron yields[b]	Experimental α_{ij}	Calculated α_{ij}[c] $E2$	$M1$
0.158 K	1.961 ± 0.04	0.317 ± 0.013	0.310	
0.209 K	1.000	0.768 ± 0.032	0.154	0.843
0.158 L	3.06 ± 0.07	0.464 ± 0.039	0.475	
0.209 L	0.193 ± 0.006	0.139 ± 0.012	0.141	0.129
0.051 L	0.471 ± 0.01	8.5 ± 4.1		7.60

[a] α_{ij} is the internal conversion coefficient of γ ray $-j$ in electron shell $-i$.
[b] Values are from Nalls, Baird, and Haynes; see reference 4.
[c] Values are from Rose; see reference 7. As indicated, the values are given for $E2$ and $M1$ transitions.