Radioactive Decays of Au^{194}, Au^{196}, and Au^{198}†

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The de-excitations of the 2+ second excited states of Pt^{194} and Pt^{196} have been studied to compare crossover transition intensities to the 0+ ground states relative to the known cascade transitions to the 2+ first excited states. An intermediate-image beta-ray spectrometer was used to measure the internal-conversion electron spectra occurring in the decays of Au^{194} and Au^{196}. A conversion line is found in the decay of Au^{194} corresponding to a 0.618-Mev crossover having a transition intensity of 25% relative to the 0.290-Mev cascade, provided the latter is assumed to be $E2$. In Pt^{196} an upper limit of 0.04% is placed on the intensity of a possible crossover transition compared with the 0.331-Mev cascade. Au^{194} exhibits the internal-conversion lines of 36 gamma-ray transitions which fit energetically into a proposed energy level scheme for Pt^{194}. A search in the electron spectrum of Au^{198} showed no gamma-ray transitions other than the three already known.

INTRODUCTION

THE systematics of the first two excited states in even-even nuclei up to $N=108$ have been studied by Scharff-Goldhaber and Weneser.[1] They have shown that, while the first excited state is 2+ in most instances, the character of the second excited state is 0+, 2+, 4+, or (odd, −), the largest number of cases being 2+. With respect to the ratio E_2/E_1 of the second to the first excited state energies, two groups occur, those of $90 \leq N \leq 108$ which can be described by the Bohr-Mottelson collective model and for which $E_2/E_1 \cong 3.33$, and those of $N \leq 88$ for which $E_2/E_1 \cong 2.2$. In the latter group the modes of motion are pictured as vibrational, the states being characterized by phonon quantum numbers of 0 and 1 for the ground and first excited states, respectively, and 2 for a triplet of states which should exist above the first excited state and which should have spins of 0+, 2+, and 4+.

In the region above $N=108$ but below the closed shell $N=126$ at Pb^{208} the above-mentioned spin sequence is followed by the levels in even-even nuclei but the ratio E_2/E_1 fluctuates somewhat more, i.e., for the isotopes $Pt^{192,194,196}$ $E_2/E_1 \cong 1.95$ while for $Hg^{198,200}$ $E_2/E_1 \cong 2.6$. It is of interest not only to establish the ratio E_2/E_1 but when the second excited state is 2+ one wishes to know the $M1/E2$ mixing ratio in the cascade transition and the ratio of reduced $E2$ matrix elements for the crossover relative to the cascade transition. If the motions are to be described as vibrational, then the two-phonon jump from the second excited to ground state should be inhibited.

The three even-even nuclides $Pt^{192,194,196}$ are thought to have 2+ second excited states. However, information on the relative crossover transition intensities from all of these states has been incomplete. For Pt^{192}, as studied in the decay of Ir^{192} by Baggerly *et al.*[2] and by Johns and Nablo,[3] the 0.612-Mev crossover is $\frac{1}{4}$ as intense as the 0.296-Mev cascade and hence the ratio of crossover to cascade reduced transition probabilities as obtained by correcting for the E^5 energy dependence is 0.007.

Pt^{194} levels have been investigated in the decay of Ir^{194} by Johns and Nablo,[3] Mandeville *et al.*,[4] and Butement and Poë.[5] A 0.620-Mev crossover gamma ray was found by Johns and Nablo, its intensity being $\frac{1}{3}$ as great as the cascade as determined from an external conversion-electron spectrum. In both Ir^{194} and in the decay of Au^{194} to Pt^{194} as reported by Thieme and Bleuler[6] the gamma-ray spectra are exceedingly complex. Owing to insufficient resolution Thieme and Bleuler were not able to study the photopeak of the 0.62-Mev crossover in a NaI scintillation spectrometer or the corresponding conversion lines in their lens-type beta-ray spectrometer at 2.7% resolution.

The decay scheme of Au^{196} was established by Steffen *et al.*[7] and by Staehelin.[8] Electron capture takes place to levels at 0.354 and 0.686 Mev in Pt^{196} and a beta branch occurs to a level in Hg^{196} at 0.426 Mev. The gamma-gamma angular correlation of the 0.332–0.354-Mev cascade in Pt^{196} has been investigated by Steffen[9] who has shown that the second excited state must be 2+ and that the $M1$ admixture in the cascade transition is (5 ± 1) %. Thieme and Bleuler[10] examined the internal-conversion spectrum of Au^{196} to investigate the possibility of an electric monopole component in the cascade transition. According to Church and Weneser[11] electric monopoles can occur between states of the same spin and parity. From the relative conversion-line intensities of the 0.332- and 0.354-Mev transitions combined with the $M1/E2$ mixing ratio in the 0.332-Mev transition as established previously by angular correlation, Thieme and Bleuler concluded that no

† Under contract with the U. S. Atomic Energy Commission.
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[11] E. L. Church and J. Weneser, Phys. Rev. **100**, 943 (1955).

monopole component was present within their limits of error. The possible existence of conversion lines corresponding to the 0.686-Mev crossover in Pt^{196} was not mentioned. Angular correlations between conversion electrons and gamma rays in the decay of Au^{196} were measured by Kane[12] whose results were consistent with the $M1$ admixture reported by Steffen and which also showed no evidence for electric-monopole conversion electrons. Inasmuch as the crossover transitions to the ground states in both Pt^{192} and Pt^{194} are of nearly the same intensity relative to the cascades, it was felt desirable to obtain information on a possible crossover in Pt^{196}. This constituted the main object of the present work.

The decay of Au^{198} to Hg^{198} is known[13] to give rise to a strong 0.411 gamma ray and to two weak lines of 0.676 and 1.088 Mev. Bergström *et al.*[14] found the same two lower energy lines in the decay of Tl^{198} to Hg^{198} but reported additional lines corresponding to gamma-ray transition energies of 0.195, 0.284, and 0.402 Mev. According to Hill[15] their K-conversion line intensities are about the same as that of the 0.676-Mev transition. In the present work a search has been made for possible weak lines of these energies in the decay of Au^{198} which might have been missed previously.

EXPERIMENTAL METHODS AND RESULTS

Sources of mixed $Au^{194,196,198}$ were made by bombarding 0.01-in.-thick platinum foil with 22-Mev deuterons in the Brookhaven 60-in. cyclotron. A chemical separation procedure similar to that described by Thieme and Bleuler[10] was used to extract the gold activity which was then deposited on thin copper or mica backings for spectrometer sources. An analysis of the Pt target material by M. Slavin showed that it contained ~0.01% gold and hence the specific activity was limited by the extraction of 0.4 mg of inactive gold. Samples 2 mm in diameter and about 2 mg/cm² thick, made from 10–20% of the activity resulting from bombardments of 100–200 μa hours, were of sufficient strength for measurements at moderately good resolution.

All of the measurements were made with an iron-free intermediate-image beta-ray spectrometer[16] using a 3.5-mg/cm² thick mica end-window Geiger counter for detection. Runs on the electron spectrum were taken within a few days after a bombardment, one example being shown in Fig. 1. A spectrometer resolution setting of 0.6% was used but the source diameter gave an effective line width of 0.74% and the source thickness made the lower energy lines increasingly wider. Except

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[15] R. D. Hill (private communication to G. Scharff-Goldhaber).
[16] D. E. Alburger, Rev. Sci. Instr. **27**, 991 (1956).

TABLE I. Gamma-ray energies and K-conversion line intensities in the decay of Au^{194}.

$E\gamma$ (Mev)	Relative K-line intensity	$E\gamma$ (Mev)	Relative K-line intensity
0.095	...	1.149	0.43
0.154	10	1.176	0.30
0.185	7	1.216	0.32
0.201	14	1.266	0.12
0.290	27	1.301	~0.1
0.327	100	1.341	0.54
0.525	2.6	1.423	0.09
0.543	0.38	1.479	3.7
0.590	1.5	1.593	0.79
0.618	1.2	1.713	0.11
0.642	1.3	1.831	0.07
0.664	0.24	1.885	0.52
0.699	0.61	1.923	0.30
0.732	0.22	2.043	0.50
0.895	0.23	2.084	0.23
0.944	1.8	2.162	0.73
1.046	0.50	2.309	0.012
1.102	0.29	2.360	0.014

for the strong Au^{196} 0.331- and 0.353-Mev conversion lines and the beta-ray continuum (endpoint energy 0.963 Mev) and 0.411-Mev conversion lines of Au^{198}, the remaining very complex structure of lines was found to decay with the 40 hour half-life of Au^{194}.

Assignments have been made for many of the 40-hour lines as indicated in Fig. 1. For the most part the structure is resolved but in some instances, as shown by question marks, the resolution was insufficient or else it was not possible to make an unambiguous assignment because of masking of mating lines. Gamma-ray energies, accurate to about 0.5%, and relative K-conversion line intensities corrected at lower energies for line width and counter-window thickness are listed in Table I.

In making a search for a possible 0.686-Mev crossover gamma ray in the decay of 5.6 day Au^{196}, it was first necessary to wait for decay of the 40-hour Au^{194} component. The remaining background consisted of the Au^{198} beta-ray spectrum which decays with a half-life of 2.7 days. It is interesting to note that when the ratio of half-lives is 2, as it is in this case, the possibility of seeing a conversion line of the longer-lived activity against a continuum of the shorter-lived activity is independent of the time at which the observations are made, at least as regards the ratio of the peak height of the conversion line to the statistical error of the count recorded during a given time interval. Thus, suppose that the counting rate of the Au^{198} beta-ray spectrum accepted by the spectrometer is 10 000/min while that of a superimposed Au^{196} conversion line is 100/min. The ratio of the peak to the statistical error for a 1-min count is then 1. At a time 5.6 days later the beta-ray background has dropped to 2500/min and the net conversion line to 50/min so the ratio of conversion line to statistical error is still 1. This relationship will be maintained until the natural counter background

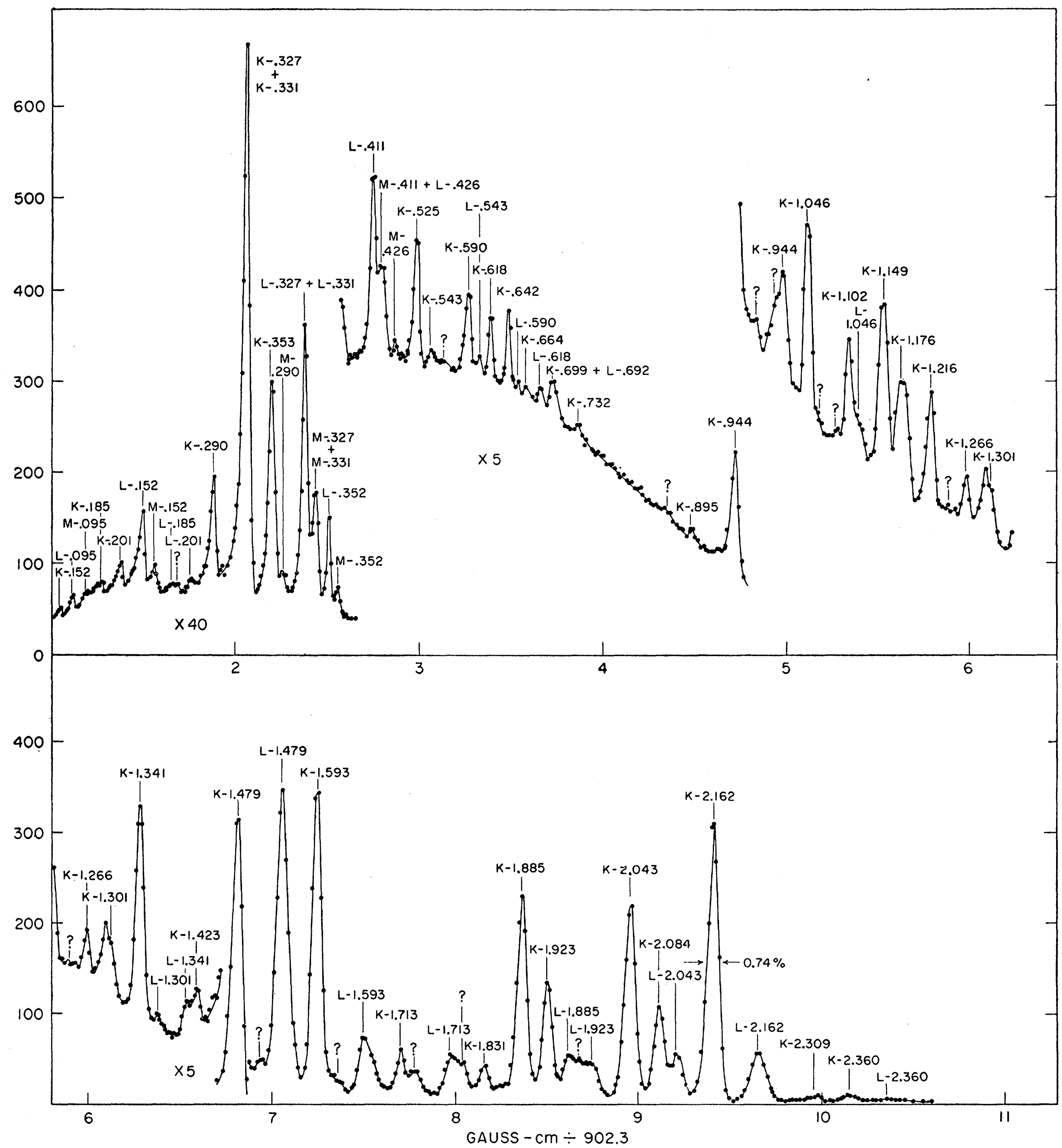

FIG. 1. Electron spectrum of $Au^{194,196,198}$ activity.

begins to become an appreciable fraction of the total count.

However, when one takes nonstatistical fluctuations into account, such as counter voltage drifts, etc., it is desirable to wait as long as possible in order to increase the ratio of the intensity of a possible conversion line to the absolute counting rate of the underlying continuum. Another consideration in this case is the presence of conversion lines of the 0.675-Mev transition in the decay of Au^{198} which are close to the 0.686-Mev Au^{196} lines in question. From the measurements of Elliot *et al.*[13] the number of 0.675 K-conversion electrons per beta ray is 1.88×10^{-4}. To determine the expected height of this peak relative to the underlying beta-ray spectrum counting rate, an allowed spectrum was constructed having the 0.963-Mev end point of Au^{198}. The fraction of the total spectrum accepted by the spectrometer at the $K-0.675$-Mev momentum position was determined by area measurements to be 2.85% when the resolution is 3.5%. Hence the ratio of the $K-0.675$-Mev conversion line to the beta-ray count at that point is $1.88\times10^{-4}/2.85\times10^{-2}=0.66\%$ for this resolution.

The above considerations suggest that if there is no way to change the initial ratio of Au^{196}/Au^{198}, such as by the use of enriched isotopes or by optimum selection of bombarding energy, then the sensitivity of detecting a possible $K-0.686$-Mev conversion line will depend on the initial source strength and on the transmission-resolution characteristics of the spectrometer.

In the most sensitive of the various searches for the 0.686-Mev K-conversion line a Pt target was given a 725 μa-hour bombardment with 22-Mev deuterons. Chemical separation was carried out 3 weeks later and almost all of the activity was deposited on the source holder. Measurements on the spectrum were begun at the end of 7 half-lives of Au^{196} (39 days) and were continued for one week. The spectrometer resolution setting was 3.5%, the corresponding transmission being 7.5% of 4π. Typical counting rates (at 43 days after bombardment) were 3000 per min for the $L-0.426$ Mev line, 60 000 per min for $K-0.331$ (calculated from the $K-0.331/L-0.426$ ratio measured with a weak source) and 180 per min at the $K-0.686$ Mev position (including the Au^{198} continuum plus 20 per min natural background). In runs giving statistics of 1.4% per point at the $K-0.686$ Mev position it was possible to show that no line was present which was greater than 5 counts/min, and thus

$$K_{0.686}/K_{0.331}<1/12\,000.$$

It may be noted that under these conditions the $K-0.675$ Mev line of Au^{198} would be only half as large as the probable errors of the points. In spite of this there was slight evidence of a line at the $K-0.675$ position which might be between 1 and 2 probable errors above the background. If it is real then its position is at too low a momentum value to be $K-0.686$. This small bump seemed to occur in every run but it did not increase with time relative to the continuum as one would expect if it were the sought-for $K-0.686$ line of Au^{196}. A possible explanation of the bump, if real, is that it is a composite of the $K-0.675$ internal-conversion electrons of Au^{198} accounting for a rate of 0.7% of the continuum, plus photoelectrons resulting from the relatively large thickness of the source which appeared to be more than 10 mg/cm^2 in thickness.

The search for possible lines of 0.195, 0.284, and 0.402 Mev in the decay of Au^{198} was made at 0.56% spectrometer resolution in order to enhance any such peaks relative to the beta-ray continuum. Sources consisted of 2-mm diameter gold leaf (0.22 mg/cm^2) attached to Scotch tape and irradiated in the Brookhaven reactor for 10 hours. Owing to source diameter the actual half-width observed for the $K-0.411$ Mev line was 0.69% and its net intensity was 4 times as great as the underlying beta-ray continuum. Counting rates were sufficient to obtain a statistical accuracy of 0.7% for each point on the beta-ray continuum or 0.18% relative to the $K-0.411$ line. Assuming that a line could be seen if it were more than two probable errors above the background then its upper limit would be 0.36% of the $K-0.411$ line. From the work of Elliot *et al.*, the $K-0.675$ Mev line is 0.6% as strong as $K-0.411$ and hence the upper limits of the 0.195, 0.284, and 0.402-Mev K-line intensities are $\frac{1}{2}$ as great as the $K-0.675$ Mev line.

DISCUSSION

From Table I the ratio $K-0.618/K-0.290$ in the decay of Au^{194} is 0.05. If it is assumed that the 0.290-Mev cascade transition is predominantly $E2$, as in the second excited-state cascade transition of both Pt^{192} and Pt^{196} then the ratio of transition intensities may be derived by using the internal-conversion coefficients calculated by Rose *et al.*[17] The resulting intensity ratio of the 0.618- to 0.290-Mev transitions is 0.25 and agrees well with the ratio 0.33 as found by Johns and Nablo[3] in the decay of Ir^{194}.

In Pt^{196} on the other hand the above experiments show that the crossover must be extremely weak. When the limit 1/12 000 is corrected for the ratio of $E2$ K-conversion coefficients, the 0.686-Mev crossover transition intensity must be less than 1/2500 as strong as the cascade transition; hence the matrix element of the crossover is less than 10^{-5} as great as that of the cascade transition. The ratios for Pt^{192} and Pt^{194} are both 7×10^{-3}. While Pt^{192} and Pt^{194} show an average behavior as regards the inhibition of the crossover from the 2+ second excited state the additional inhibition factor of more than 700 in the case of Pt^{196} presents a substantial and interesting variation.

In attempting to construct a level scheme for Pt^{194}, a number of sum relations within the errors of measurement may be found from Table I, involving sets of 3 gamma rays. The level scheme must contain the already well established first two excited states at 0.327 and 0.618 Mev and all the levels must fall within the total decay energy of 2.57 Mev based on the end point of the positron group to the ground state.[6] These two limitations simplify the analysis considerably. Figure 2 shows a level diagram into which all of the resolved gamma transitions fit reasonably well and in fact several of the gamma rays, i.e., those of 0.642, 0.944, and 1.046 Mev, fit just as well in two places. Comparing with the levels reported previously[3,6] in both Ir^{194} and Au^{194} decay, those at 0.327, 0.618, 1.266, 1.479, 1.666, 1.794, 2.043, 2.212, and 2.309 Mev agree with previous results while the levels at 1.213, 1.923, 2.084, 2.162, and 2.364 Mev are unique to the present work. It may be noted that the 2.084-Mev state involves only one gamma-ray transition, the only possible alternative being to assign a level at 2.411 Mev.

Verification of the Pt^{194} level scheme proposed here, which should be considered as tentative, could be

[17] M. E. Rose, *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (North Holland Publishing Company, Amsterdam, 1955).

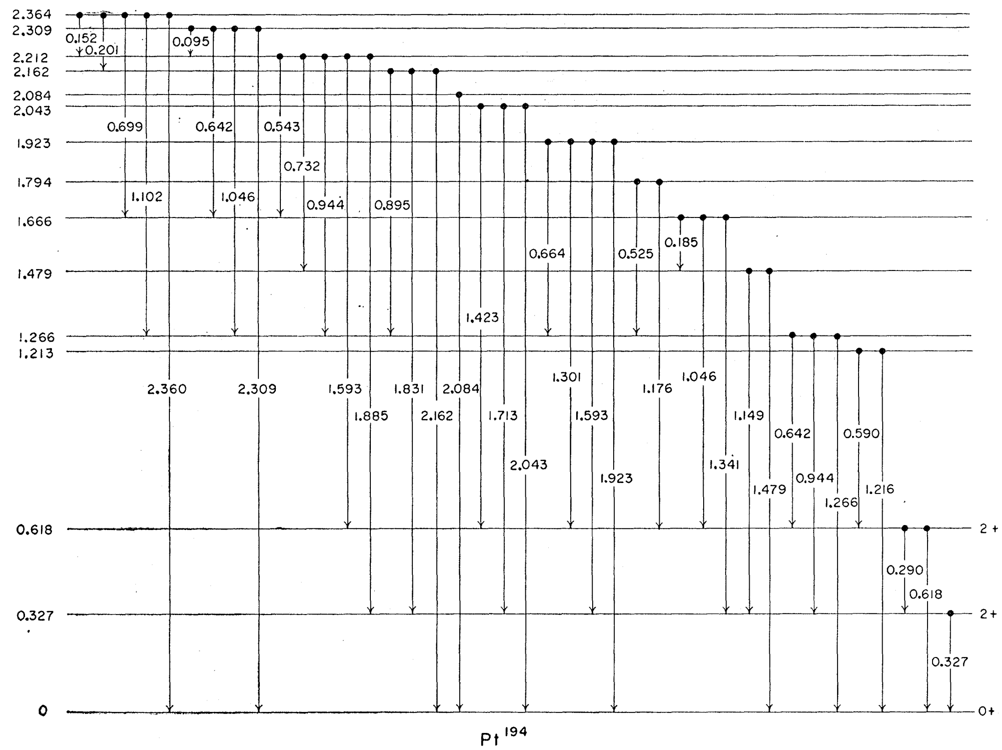

FIG. 2. Suggested energy level scheme for Pt^{194}.

carried out by measurements of the conversion-electron spectrum at higher resolution and possibly by detecting coincidences between pairs of conversion lines. The present results on Au^{194} should be considered mainly as a guide for higher resolution studies.

The results found for Au^{198} decay indicate that any levels in Hg^{198} which could account for the 0.195-, 0.284-, and 0.402-Mev transitions reported[14,15] in Tl^{198} decay are not excited in the decay of Au^{198} to the same extent. A reinvestigation of Tl^{198} to confirm the existence of these weak lines might be worth while.

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