PHYSICAL REVIEW VOLUME 75, NUMBER 5 MARCH 1, 1949

The Radiations from 2.7-Day Au^{198}

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(Received November 3, 1948)

The radiations from the 2.7-day Au^{198} activity were investigated with a counter and photographically recording beta-ray spectrometer. The shape of the continuous beta-ray spectrum is essentially that of an allowed spectrum with some additional intensity in the region below 0.6 Mev. The maximum energy of the spectrum is 0.966 ± 0.010 Mev. Very strong conversion lines are observed from a gamma-ray of 0.408 ± 0.004 Mev, and weak conversion lines attributed to gamma-rays of 0.157 ± 0.002 and 0.208 ± 0.002 Mev were detected. These weak lines and the deviation of the spectrum from the theoretically permissable spectra suggest that the beta-ray spectrum may be complex with a component of 0.601 ± 0.016 Mev maximum energy occurring in 15 percent, or less, of the disintegrations. This suggested mode of decay is consistent with the theoretical analysis of the spectrum and half-life. In terms of this theory the 0.966-Mev transition is described by the polar-vector or pseudo-scalar interaction corresponding to a nuclear spin change of ±1, with no parity change.

I. INTRODUCTION

ONE of the first radioactive substances obtained by neutron irradiation was the 2.7-day activity of Au^{198} produced by a $n-\gamma$ reaction on the stable Au^{197} isotope. The radiations from this activity have been studied many times, and yet there is still no positive assurance that they are unambiguously known. The earliest determinations of the beta- and gamma-ray energies naturally employed absorption methods.[1-6] The energies reported for the beta-particles are in the region of ≈0.5 to ≈1.0 Mev and for the gamma-rays from ≈0.065 to 0.7 Mev with one report[2] of a gamma-ray of 2.5 Mev. Some increase in the precision of the radiation measurements was obtained by Richardson,[7] who used a cloud chamber to observe the momentum distribution of the beta-rays, and electrons ejected by quanta from a radiator. Lawson and Cork,[8] using a beta-ray spectrometer, located the internal conversion lines of a Au activity of approximately 3-day half-life. Siegbahn[9] has reported that the 2.7-day radiations consist of a single beta-ray of 0.92 Mev, followed by a gamma-ray of 0.401 Mev. Recently, Saxon[10] and Wilkinson and Peacock[11] have confirmed the 0.970 ± 0.010 Mev value for the maximum energy of the beta-rays we had previously reported.[12] They did not find evidence for the low intensity component of ≈0.6 Mev maximum energy which our work indicates may be present.

In addition to the energy measurements, many investigators[13-18] have employed coincidence absorption methods in their work on this activity. Again, in this coincidence work, there is disagreement among the various experimenters. In particular, the existence of gamma-gamma coincidences is disputed.

Considering the disagreement among the various workers, the fact that many different decay schemes for Au^{198} have been proposed is

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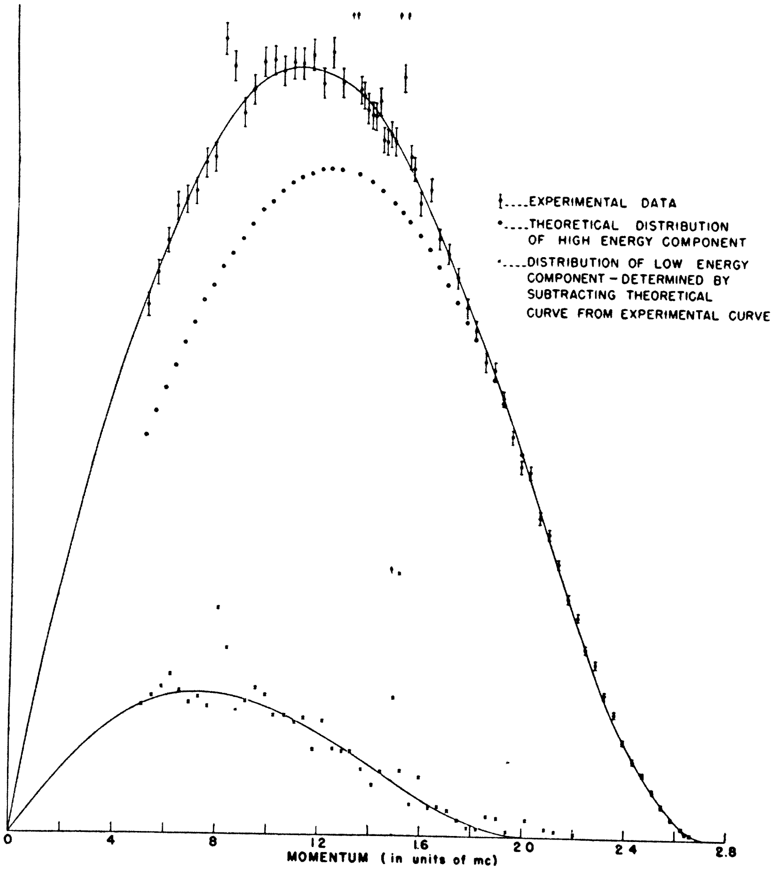

FIG. 1. Momentum distribution of Au^{198} electrons. The theoretical curve is

$$CF(W^2-1)(W_0-W)^2,$$

where W is the total electron energy in mc^2 units, W_0 is the end-point energy, 2.89_5, F is the Fermi Coulomb effect function, and C is the single component correction factor shown in curve I of Fig. 5. Arrows indicate experimental points off the figure arising from the conversion lines of the 0.41 Mev gamma-ray.

not surprising. In this paper we shall present the evidence and arguments leading to still another decay scheme.

II. EQUIPMENT

The measurements to be described below were all made with two beta-ray spectrometers.[19] One employs G-M tube recording, has a radius of 10 cm, and is similar to that constructed by Lawson and Tyler.[20] It possesses the additional feature that more than one counter can be located in the electron trajectory. With this arrangement it is possible to record coincidences between these counters, as produced by a particle in the required trajectory, with a very substantial reduction of the background due to gamma-rays from the source. This method is particularly advantageous when observing photoelectrons ejected from a radiator to determine gamma-ray energies. For measurements of continuous spectra, however, the shielding was sufficient to reduce the background to such a low level that the coincidence feature was not used. The other spectrometer employs photographic recording and is similar to those used by Ellis[21] and Valley and McCreary.[22]

The magnetic field for both of these spectrometers is provided by an electromagnet of the usual design except for the field coils. Instead of coils which have a few turns and require large currents, these coils have several thousand turns, high resistance, and require currents from 3 to 150 milliamperes. With this arrangement electronic stabilization of the field current proved to be extremly successful. All magnetic field

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[19] This equipment is described in detail in Plutonium Project Report CP-3702, which is to be included in the nuclear physics section of the Plutonium Project Record.

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measurements were made with a search coil arrangement that was calibrated with a standard mutual inductance. With this equipment the total error in the energy measurements was less than one percent for particles greater than 100 kev and less than two percent for particles between 50 and 100 kev.

The sources used for the continuous spectrum and conversion electron studies consisted of gold foils mounted on Zapon films 0.05 mg/cm^2 thick. The thinnest foil used was 0.26 mg/cm^2. Thicker foils were occasionally used for conversion electron pictures, since, for energy measurements, they did not impare the precision and provided more activity. Also, thicker foils were used to determine how the foil thickness affected the shape of the continuous spectrum. Initially, it was found that usually the foils did not adhere sufficiently to the Zapon to permit handling. This situation was remedied by covering the Zapon film with extremely small droplets of water from an atomizer before placing the foils on the film. The action of the water droplets, due to surface tension, was to produce intimate contact between the foil and film. When the source was put in a vacuum, the water between the foil and film evaporated rapidly.

III. THE BETA-RAY SPECTRUM

The continuous spectrum was investigated with the 180° counter-recording spectrometer. The momentum distribution obtained with the thinnest source used, 0.26 mg/cm^2, is shown in Fig. 1. The Kurie plot, Fig. 2, was calculated from this data. It is apparent that this plot yields the essentially straight line theoretically attributed to allowed transitions and typical of single component beta-ray spectra. The final value for maximum energy obtained for the continuous spectrum is 0.966±0.010 Mev. From the energy of the spectrum and the 2.7-day half-life, one would expect this to be a second forbidden transition for which the Kurie plot would not be linear. It was found that the curve obtained could not be fitted by any of the second forbidden correction factors predicted by theory. In particular, there was greater intensity in the region of the spectrum below 0.6 Mev than could be accounted for by a one-component theoretical spectrum. The appearance of weak conversion lines indicated that there were additional gamma-rays present other than the well-known one of 0.41 Mev.[23,24] Some of these low energy conversion lines had been reported previously by Plesset[25] and a low

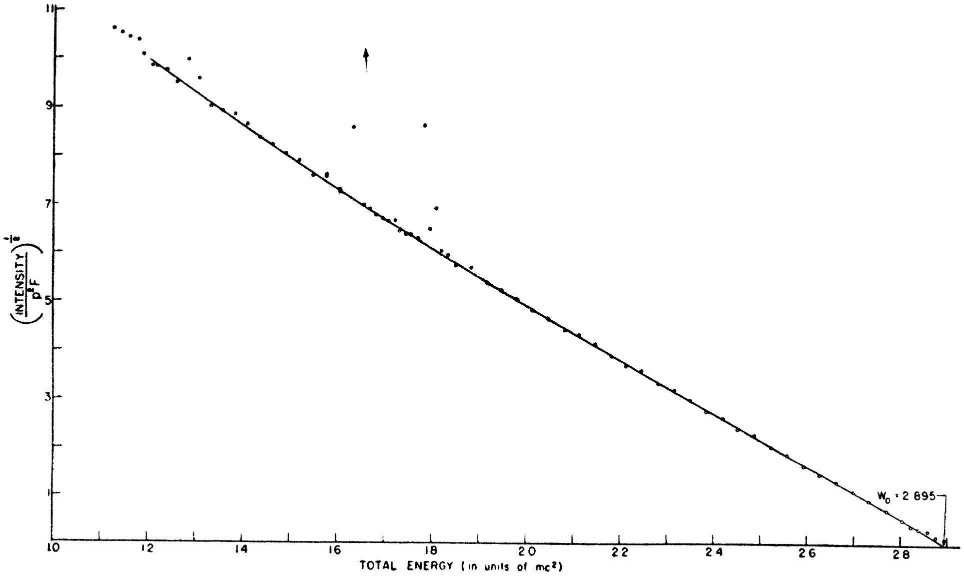

FIG. 2. Kurie plot. Arrows indicate points off the figure. The intensity is (counting rate)/$H\rho$.

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[24] DuMond, Lind, and Watson, Phys. Rev. **73**, 1392 (1948).

[25] E. H. Plesset, Phys. Rev. **62**, 181 (1942).

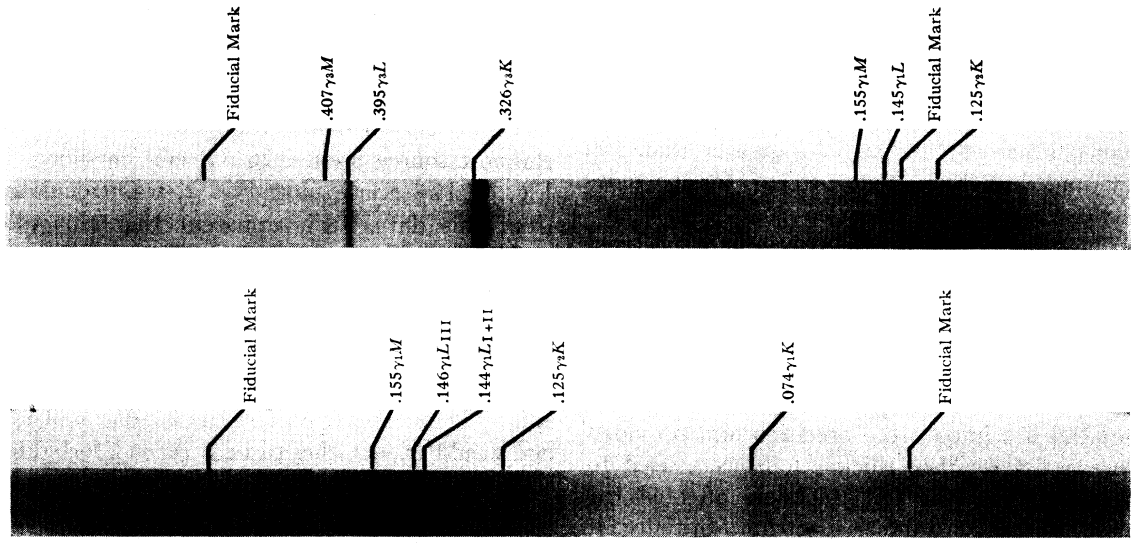

FIG. 3. Conversion electron pictures. Typical examples of the "pictures" obtained with the photographic spectrograph of the lines found in the Au spectrum. The weaker lines were lost in the process of making the reproductions.

energy gamma-ray by Richardson.[7] These weak gamma-rays could not be fitted in a decay scheme involving a single beta-ray component.

Measurements were undertaken to determine whether the additional intensity below 0.6 Mev could be attributed to source thickness effects. First the continuous spectrum was determined with sources ≈ 25.0 and 1.46 mg/cm^2 thick. It was found that the spectrum obtained from the 25.0 mg/cm^2 thick source contained considerably more intensity in the low energy region than had been obtained from the thinnest source. However, there was only a slight difference in the spectra obtained from the two thinner sources. If these were the only pertinent data, one could not exclude the possibility that the observed extra intensity in the low energy half of the spectrum is an effect of source thickness.

A second possibility is that the low intensity lines, as well as the extra intensity, could come from contamination in the source. Accordingly, the decay of this activity was followed with the spectrometer, and it was found that all parts of the spectrum, and the low intensity lines, decayed at the same rate over a period of two half-lives. The half-life observed was less than one percent lower than the reported value of 2.70 days.[26] A spectrochemical analysis of the gold foil used was made in an attempt to detect impurities. None were found which were present in quantities greater than traces except silver. Since the $K-L$ separations of the observed lines (Fig. 3) is approximately that expected for gold, these lines could not be attributed to silver. The separations could not be determined with sufficient accuracy to decide in what heavy element the conversion of the gamma-ray took place. Also, considering the large *activation* cross section of gold as compared to that of silver and the other impurities present, one is led to the conclusion that these lines are not due to contamination or impurities. Since the cross section for neutron capture is usually considerably higher than for other reactions, e.g., $n-p$ reactions, one would not expect to find activities other than the Au^{198} one.

On the basis of this evidence it is feasible to consider the possibility that there is in reality a second low intensity component in the continuous spectrum. Proceeding on this assumption, one can determine its shape by subtracting the theoretically predicted curve from the data as is indicated in Fig. 1. The maximum energy of the spectrum so obtained is approximately 0.6 Mev. This value is in good agreement with what is to be expected if this low intensity component is

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[26] N. Sugarman and co-workers; unpublished Plutonium Project Work. See also reference 20.

followed by gamma-rays of 0.157 and 0.208 Mev, as the conversion electron studies described below indicate are present. However, assuming that the suggested decay scheme, Fig. 4, is correct, a more accurate procedure to determine the maximum energy of this component is to subtract the low intensity gamma-ray energies from the maximum energy, 0.966, of the intense spectrum. The value so obtained is 0.601±0.16 Mev.

From the curves shown in Fig. 1 one can ascertain that the low intensity component is present in only 15 percent, or less, of the disintegrations. It is difficult to assign an error to the value of 15 percent, since, in order to determine the branching, we had to extrapolate the spectrum below the lowest momentum measured.

IV. GAMMA-RAY MEASUREMENTS

In order to obtain detailed information on the conversion lines that were found superimposed on the continuous spectra, they were studied with the photographic spectrograph. The lines from the low energy gammas were considerably weaker than the lines from the 0.41 Mev gamma. In fact, only by carefully controlling the exposure could both the K and L lines of the low intensity gammas be detected. The conversion line data and an analysis of it is contained in Table I. Figure 3 contains typical examples of the pictures obtained with the spectrograph. Considering the various factors that produce variations in the energy determinations of the lines, the best values for the gamma-ray energies are 0.408 ±0.004, 0.157±0.002 and 0.208±0.002 Mev. It is to be noted that if a gamma-ray of 0.065 Mev were present with an intensity greater than a few percent, as suggested by Feather and Dainty,[15] it should have produced intense conversion lines. No such sharp lines were detected. However, there was some evidence of blackening of the negative where the Auger electrons produced by Hg and Au x-rays were focused. We have not been able to explain, at the moment, why Jnanananda[27] should have found these Auger electrons and not the lines from the 0.157 and 0.208 Mev gammas.

Most of the lines detected have been observed by Plesset, who suggests that they are due to an isomeric state of Hg^{198}, to which the 0.970 Mev beta-ray decays. If this were the case these gammas should be as intense as the 0.41 Mev gamma and would be readily detectible by absorption methods. Recently DuMond, Lind, and Watson[24] measured the energy of the 0.41 Mev gamma very accurately with a crystal spectrometer. The value they obtained, 0.4112±0.001, agrees with ours, to within the experimental error. They were not able to detect the diffracted beam from the weaker gammas because of their low intensity, but by using their apparatus to obtain a finely collimated gamma-ray beam, an absorption experiment was performed that

TABLE I. Gamma-rays and conversion lines in Au^{198}.

Gamma-ray	Conversion lines observed	Estimated intensity of conversion lines	Energy of conversion lines (Mev)	Energy of gamma-rays (Mev)
1	K	Medium	0.074	0.157
	$L_{\mathrm{I}}+L_{\mathrm{II}}$	Weak	0.144	0.158
	L_{III}	Very weak	0.146	0.158
	M	Faint	0.155	0.157
2	K	Weak	0.125	0.208
	L	Very faint	0.193	0.207
3	K	Very strong	0.326	0.409
	L	Strong	0.395	0.407
	M	Strong	0.407	0.409

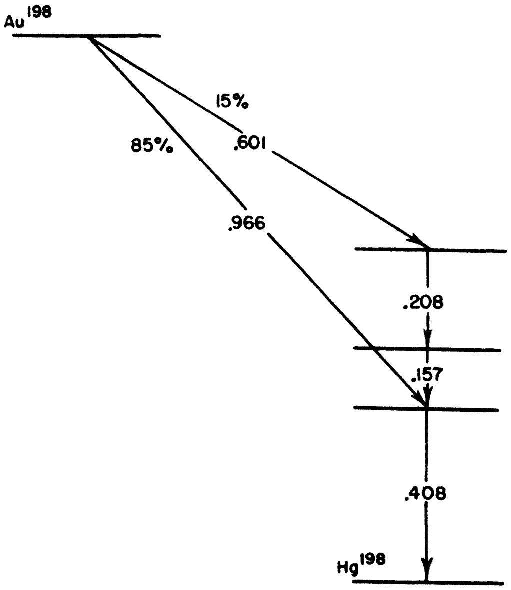

FIG. 4. Suggested decay scheme for Au^{198}. The cascade order of the two weak gamma-rays is not known.

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[27] Swami Jnanananda, Phys. Rev. 70, 812 (1946).

indicates these gammas are present to about the intensity we observed.

A determination of the average gamma-ray energy has been made by Barker,[28] who gives a value of 0.366 Mev of gamma-ray energy per electron (beta-particles plus conversion electrons) emitted. This corresponds to 0.383 Mev of gamma-ray energy per disintegration, using the 4.7 percent internal conversion coefficient for the 0.41-Mev gamma-ray reported by Wiedenbeck and Chu.[16] Barker's value is consistent with a decay scheme containing only the 0.41-Mev gamma.

Using calorimetric methods, Cannon and Jenks[29] find that the total energy per disintegration for this activity is 0.788±0.028 Mev. From our intensity distribution curve we find that the average energy of the beta-rays is 0.310±0.01 Mev. Adding the gamma-ray and conversion electron energies to this one obtains 0.773 Mev for the average energy per disintegration. Thus our results are in agreement with those obtained by Cannon and Jenks, but not with the result of Barker.

V. COMPARISON WITH THEORY

Considering the abundance of conflicting data and proposed dècay schemes published in the literature, it is obvious that the decay scheme proposed by the authors, Fig. 4, must be considered as tentative. It is worth while, however, to point out that no contradiction with the theoretically predicted spectra, half-life, or branching ratio was found.

The most obvious characteristic of the beta-spectrum measured is its close similarity with the so-called allowed type spectrum. The fact that the half-life of Au^{198} is large enough to require that it be classified as either a first or second forbidden transition prompted one of us to

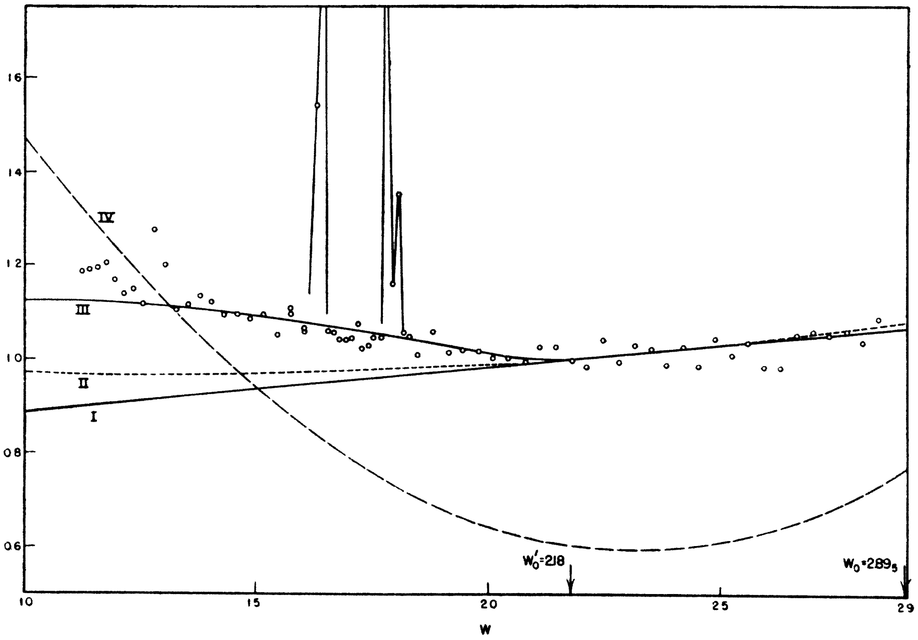

FIG. 5. Dependence of the Au^{198} correction factor on the total energy of the emitted electrons. The points are proportional to the thin source (0.26 mg/cm^2) (counting rate)/($H\rho$) data divided by $F(W_0-W)^2(W^2-1)$. Curve I is the theoretical correction factor for a single component forbidden transition with $W_0=2.89_5$, i.e., C_{1P} with $\Delta J=\pm1$, "no" or C_{2V} with $\Delta J=\pm1$ "no" for a $0\leftrightarrow1$ transition or $|\boldsymbol{\alpha}|\gg\rho$ (not $0\leftrightarrow1$). Curve II is C_{2V} with $\Delta J=\pm1$ (not $0\leftrightarrow1$) for $|\boldsymbol{\alpha}|=\{W_0\rho\}$. Curve III is C_{2V} or C_{1P} obtained by adding a low energy component with end-point $W_0'=2.18$ mc^2 for $\approx$15 percent of the transitions. Curve IV shows the energy dependence of the term in C_{2V} containing $|R_{ij}|^2$. The ordinates are arbitrary. Curves I, II, III and the data normalized to 1 at $W=2.2$ mc^2.

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[28] Edgar C. Barker, Phys. Rev. **72**, 167 (1947).

[29] C. V. Cannon and G. H. Jenks, private communication.

compute the correction factors to the allowed spectral shape one would expect according to the present theory of forbidden beta-decay transitions.[30]

All of the first forbidden correction factors are nearly independent of the electron energy if the spin and parity change for the transition is $\Delta J = \pm 1$ or 0 "yes." In the expressions for these correction factors the quantity M_0 of Konopinski and Uhlenbeck's paper[30] is essentially energy independent for high Z. This quantity M_0 containing $(\alpha Z/\rho)^2$, which is the square of the potential energy of an electron at the surface of the nucleus, predominates. $(\alpha Z/\rho)$ is much larger than the electron and neutrino energies appearing in the other terms of the correction factor. Only C_{1T} and C_{1A} for $\Delta J = \pm 2$, "yes," show such a strong variation with energy that they can be ruled out as possibilities for the Au^{198} transition upon comparison with the observed spectrum.

Of the second forbidden correction factors, the only ones that are weakly energy dependent are C_{2V}, $\Delta J = \pm 1$, "no" and C_{2T}, $\Delta J = 0 \rightarrow 0$, "no." The second forbidden tensor interaction with $\Delta J = 0 \rightarrow 0$ can be ruled out on two counts. First, the 0.41 Mev gamma-ray transition from the excited state of Hg to its ground state, which has spin zero, would be $0 \rightarrow 0$ and therefore is completely forbidden. Second, if the spin of Au^{198} is zero, one would expect to find the $0 \rightarrow 0$ beta-transition to the ground state of Hg out competing the $0 \rightarrow 0$ transition to the Hg excited state.

That C_{2V} has essentially the same energy dependence as C_{1X} for $\Delta J = \pm 1$ is not apparent except for the $0 \leftrightarrow 1$ transition, in which case only the factor containing the nuclear matrix element, $\int \boldsymbol{\alpha} \times \mathbf{r}$, enters. R_{ij} and A_{ij} appearing in C_{2V} vanish for $0 \leftrightarrow 1$ transitions. For $\Delta J = \pm 1$ (not $0 \leftrightarrow 1$)C_{2V} contains all three of the above nuclear matrix elements and the variation of the coefficients of $|R_{ij}|^2$ and $|A_{ij}|^2$ with energy does not resemble the experimental correction factor shown in Fig. 5. One may estimate the magnitudes of these nuclear matrix elements[31] in terms of the nuclear radius ρ (in units of the Compton wave-length) and the average value of $|\boldsymbol{\alpha}| \approx$ nucleon velocity/velocity of light. Such an estimate yields, for $|\boldsymbol{\alpha}| \approx 0.1$, the following magnitudes for the three matrix elements appearing in C_{2V}:

$$\left|\int \boldsymbol{\alpha} \times \mathbf{r}\right|^2 \approx \tfrac{2}{3} |\boldsymbol{\alpha}|^2 \rho^2 = 36 \times 10^{-7},$$

$$\sum_{ij} |A_{ij}/2!|^2 \approx (5/9) |\boldsymbol{\alpha}|^2 \rho^2 = 30 \times 10^{-7},$$

$$\sum_{ij} |R_{ij}|^2 \approx \tfrac{2}{3} \rho^4 = 2.0 \times 10^{-7}.$$

The term in C_{2V} containing $\sum |A_{ij}|^2$ adds less than one percent to C_{2V} for all energies as compared to that part contributed by the $|\int \boldsymbol{\alpha} \times \mathbf{r}|^2$ term, because the energy dependent factor multiplying $|A_{ij}|^2$ does not contain the Coulomb energy of the electron. Curve IV of Fig. 5 shows the term containing $|R_{ij}|^2$. Curve II is C_{2V} obtained by using a minimum estimate of the magnitude of $|\boldsymbol{\alpha}|$, i.e., $|\boldsymbol{\alpha}| = W_0 \rho$, where $W_0 = 2.9$ is the end-point energy in mc^2 units. Curve I is proportional to the coefficient of $|\int \boldsymbol{\alpha} \times \mathbf{r}|^2$ and is thus appropriate for C_{2V} if $|\boldsymbol{\alpha}| \gg \rho$.

The only first forbidden correction factor which has the same order of magnitude as C_{2V} with $\Delta J = \pm 1$ and at the same time varies with energy just as shown in Curve I of Fig. 5 is C_{1P}. Of the correction factors which vary only slightly with energy, only C_{1P} and C_{2V} with $\Delta J = \pm 1$, "no" and C_{2T} with $\Delta J = 0 \rightarrow 0$, "no" have the order of magnitude 10^{-3}, which is the ratio of allowed to experimental ft value for Au^{198}. The other first forbidden correction factors with $\Delta J = \pm 1$ or 0, (not $0 \rightarrow 0$), "yes" are too large by a factor of 100 because of the Coulomb energy term. C_{1T} and C_{1A} have the order of magnitude 10^{-3} for $\Delta J = \pm 2$, "yes," but cannot possibly be reconciled with the almost allowed type spectrum. All the other second forbidden correction factors are too small by a factor of 1/100 and/or do not show any similarity with the energy dependence of the experimental correction factor.

The deviation of the low energy data from the theoretical correction factors (Curves I or II of Fig. 5) can be attributed to four possible alternatives. (1) The beta-decay of Au^{198} is complex,

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[30] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **60**, 308 (1931). The notation of this paper is used throughout.

[31] E. Greuling, Phys. Rev. **61**, 568 (1942).

(2) back scattering from the source and its support has not been entirely eliminated, (3) the present theory applied to low energy electrons is not correct,[32] or (4) an undetected contaminant activity may be present.

To resolve the observed spectrum into two components would be an indefensible procedure if there was no supporting evidence other than the slight deviation of the spectrum from one possibly predicted by theory. However, the existence of the two weak gamma-rays of 0.157 and 0.208 Mev and the possibility that the $\gamma-\gamma$ coincidences reported are real suggests the level scheme shown in Fig. 4. It may not be accidental that the inner end point, $W_0'=2.18$ mc^2, obtained by subtraction, coincides with the energy below which the observed spectrum is larger than predicted by the theory for a single component decay.

Curve III in Fig. 5 was obtained by assuming a branching decay for which the theoretical correction factor has the form

$$C=C_{2V\text{ or }1P}[1+A(2.18-W)^2/(2.89_5-W)^2]$$

below 2.18 mc^2. $C_{2V\text{ or }1P}$ is Curve I shown in Fig. 5. The parameter A is the ratio of inner end point to outer end-point correction factors which must have the order of magnitude unity. To fit the data as shown, $A=0.7$ was used. This corresponds to a maximum branching ratio of 15 percent low energy to 85 percent high energy transitions. A smaller branching ratio would be obtained if part of the rise below 2.2 mc^2 is due to source thickness effects, or if C_{2V} for the main component is between Curves I and II shown in Fig. 5.

VI. CONCLUSIONS

It is shown that the 2.7-day Au^{198} beta-ray spectrum has a maximum energy of 0.966 ± 0.010 Mev. Measurements with the photographic spectrometer indicate that in addition to the strong 0.408-Mev gamma-ray two weak gammas of 0.157 and 0.208 Mev produce internal conversion electrons. The presence of the low energy lines suggests the possibility that the gold spectrum is complex with 15 percent, or less, of the transitions being to an excited level of Hg followed by the two weak gamma-rays in cascade to the level 0.408 Mev above the ground state. The maximum energy, 0.601 Mev, of the low intensity beta-rays is consistant with the theoretical analysis of the complete spectrum above 200 kev. In this region we believe no distortion of the spectrum, caused by counter window absorption, scattering in the source, or its backing, is present.

The theoretical analysis of the spectrum suggests that the 0.966-Mev beta-transition corresponds to a nuclear spin change of ±1 with no parity charge according to the polar vector or pseudoscalar form of interaction.

VII. ACKNOWLEDGMENT

The authors wish to acknowledge many helpful discussions with members of this laboratory too numerous to mention. Particularly appreciated is the counsel of Doctors S. DeBenedetti and E. O. Wollan. This work was done at the Oak Ridge National Laboratory under Contract W-7405 eng 26 for the Atomic Energy Commission.

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[32] The failure of the theory at low energies is suggested by the anomalous allowed spectra observed for Cu^{64} and Cu^{61} by C. S. Cook and L. M. Langer, Phys. Rev. **73**, 601 (1948) and Phys. Rev. **74**, 227 (1948).

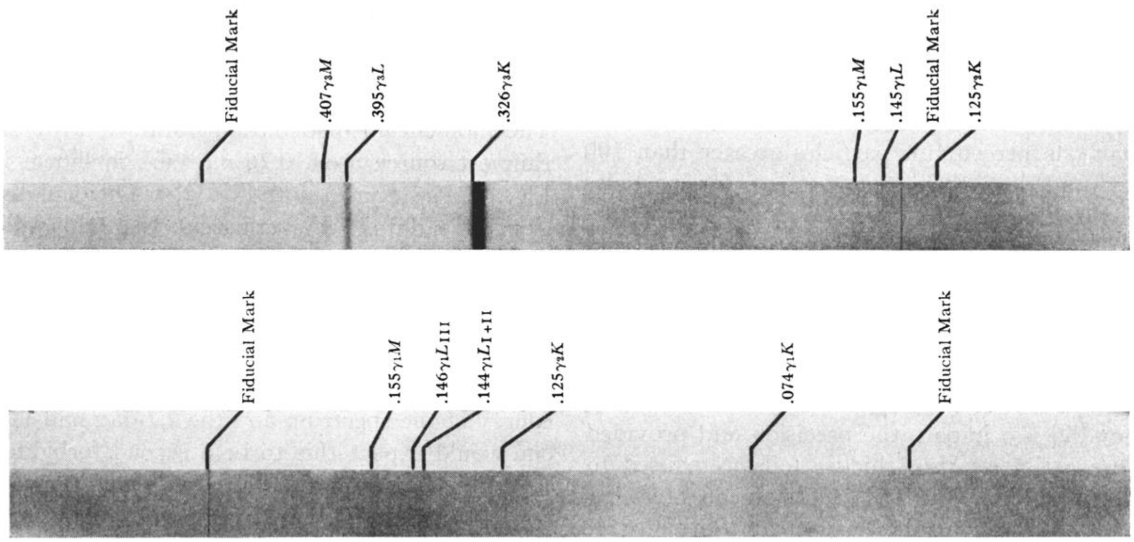

FIG. 3. Conversion electron pictures. Typical examples of the "pictures" obtained with the photographic spectrograph of the lines found in the Au spectrum. The weaker lines were lost in the process of making the reproductions.