Conversion of the Pt^{196} Gamma Rays*

M. T. THIEME† AND E. BLEULER
Department of Physics, Purdue University, Lafayette, Indiana
(Received October 31, 1955)

The conversion coefficients of the 331- and 354-kev transitions in Pt^{196}, following electron capture of Au^{196}, were measured with the aid of a lens spectrometer adapted for coincidence measurements. The lower, 354-kev, gamma ray is an $E2$ transition whereas the K-conversion coefficient of the 331-kev gamma ray was found to be $18\pm5\%$ larger than that of a pure $E2$ transition. From the good agreement of this excess with that expected for the 4.5% $M1$ admixture derived from angular correlation measurements ($18\pm2\%$), it is concluded that the fraction of conversion electrons due to an electric monopole transition is $0\pm5\%$ (standard error). The intensity ratio of the two transitions is 0.278 ± 0.010.

INTRODUCTION

THE decay scheme of Au^{196}, as established by Steffen *et al.*[1] and Staehelin,[2] is shown in Fig. 1. From the conversion coefficients, the two gamma rays in Pt^{196} were found to be electric quadrupole transitions.[1] Steffen and Roberts[3] confirmed this assignment by angular-correlation measurements which also yield spin 2 for both excited states. With improved angular-correlation equipment, Steffen[4] was able to show that the 331-kev transition is a mixture of $5\pm1\%$ $M1$ and 95% $E2$ radiation. More recent, unpublished measurements yield an $M1$ admixture of $4.5\pm0.8\%$ (error limit).[5] The independence of the angular correlation from the type of source used (metal, $AuCl_3$, aqueous solution of $AuCl_3$) is taken as proof that the angular correlation is not disturbed by extranuclear fields.[6]

The present investigation started as an attempt to check the mixed character of the 331-kev transition by measuring its conversion. According to the tables of Rose *et al.*[7] the K-conversion coefficient is 0.050 for pure $E2$ radiation, 0.059 for a 4.5% $M1$ admixture. The difference between these values appears large enough to allow a determination of the $M1$ admixture to within perhaps 25%. There exists, however, the possibility of a further contribution to the conversion line by electric monopole transitions.[8] From the formulas of Thomas,[9] with a matrix element $M=R^2=(1.2\times10^{-13}A^{1/3})^2$, one obtains $\lambda_e(E0)\sim10^{11}$ sec^{-1}. For 0−0 transitions, Drell and Rose[10] found the decay rate to be reduced by factors of 10 to 50, giving in the present case $\lambda_e(E0)\sim2\times10^9$ to 10^{10} sec^{-1}. This should be compared with the emission rate of $E2$ and $M1$ conversion electrons which, however, is not known at the present time since the lifetime of the 685-kev state has not been measured. The theoretical estimates for single-particle gamma-ray transitions,[11] $\lambda_\gamma(E2)\sim10^9$ sec^{-1} and $\lambda_\gamma(M1)\sim10^{12}$ sec^{-1}, are not compatible with observation. The $E2$ decay is probably enhanced to $\lambda_\gamma(E2)\sim10^{10}$ to 10^{11} sec^{-1}, the $M1$ transition weakened to 5×10^8 to 5×10^9 sec^{-1}. The rate of emission of $E2$ and $M1$ conversion electrons would be of the order of $\lambda_e(E2+M1)\sim5\times10^8$ to 5×10^9 sec^{-1}, and the monopole conversion rate (2×10^9 to 10^{10} sec^{-1}) might be quite important. An argument for the reduction of its intensity, however, may be found in the collective character of the 2^+-2^+ transition which is thought to be connected with the disappearance of a phonon, with a corresponding (vector) change of the nuclear angular momentum.

The interpretation of the measured conversion coefficient, then, will depend on whether one assumes the $E0$ transition to be negligible—which allows a check of the $M1-E2$ mixing ratio—or whether one adopts the mixing ratio obtained from the angular-correlation measurements and tries to determine the amount of monopole conversion.

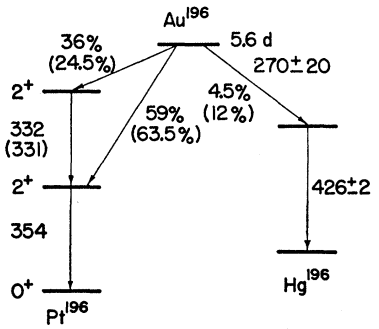

FIG. 1. Decay scheme of Au^{196}. The energies (in kev) are taken from Staehelin,[2] the branching ratios from Steffen *et al.*[1] The values given in parentheses are obtained in the present work. It is assumed that electron capture to the ground state of Pt^{196} is negligible.[1]

* Work supported by the U. S. Atomic Energy Commission. Part of a doctoral thesis submitted by M. T. Thieme to the faculty of Purdue University.
† Now at Los Alamos Scientific Laboratory, Los Alamos, New Mexico.
[1] Steffen, Huber, and Humbel, Helv. Phys. Acta **22**, 167 (1949).
[2] P. Staehelin, Phys. Rev. **87**, 374 (1952).
[3] R. M. Steffen and D. M. Roberts, Phys. Rev. **83**, 222 (1951).
[4] R. M. Steffen, Phys. Rev. **89**, 665 (1953).
[5] R. M. Steffen (private communication).
[6] R. M. Steffen, Advances in Phys. **4**, 294 (1955).
[7] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).
[8] *Note added in proof.*—See E. L. Church and J. Weneser, Phys. Rev. **100**, 943 (1955).
[9] R. Thomas, Phys. Rev. **58**, 714 (1940).
[10] S. D. Drell and M. E. Rose, Progr. Theoret. Phys. (Japan) **7**, 125 (1952).
[11] V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).

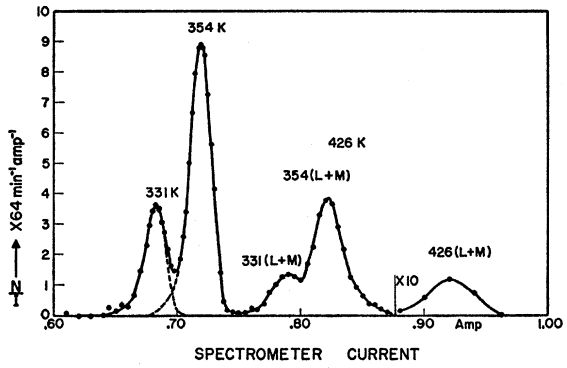

FIG. 2. Conversion electrons of Au^{196}, measured three weeks after bombardment. Spectrometer resolution: 2.7%.

SOURCE PREPARATION AND APPARATUS

Au^{196} was produced by bombarding Pt with the internal 9.5-Mev deuteron beam of the cyclotron. The simultaneous production of Au^{194}, Au^{198}, and Au^{199} complicated the measurements, but no other production method [e.g., $Ir^{193}(\alpha,n)Pt^{196}$] gave sources strong enough for spectrometer and coincidence studies. The Pt was dissolved in aqua regia, the nitric acid boiled away, adding 6N HCl, and the gold extracted carrier free with ether. After evaporating to dryness in a Teflon beaker, the gold chloride was taken up in a few drops of water and deposited on a $\sim$1-mg/cm^2 Mylar foil. Charging effects were prevented by covering the source with a 10-μg/cm^2 nylon film upon which a thin Al coat was evaporated.

The measurements were performed with a double-coil lens spectrometer adapted for coincidence measurements,[12] using scintillation detectors for both the electrons focused in the spectrometer and the radiations coincident with them.

MEASUREMENTS

The quantity that can be measured with the highest accuracy is the ratio of the K-conversion coefficients of the 331-kev and 354-kev transitions. Its determination requires the measurement of the intensity ratio of the two conversion lines and an accurate redetermination of the intensity ratio of the two gamma rays which had been given by Steffen *et al.*[1] as 0.38$\pm$0.03.

Conversion Electrons

From the spectrum of the conversion electrons (Fig. 2), the gamma-ray energies were determined to be 331.0$\pm$0.4 kev and 354.0$\pm$0.4 kev (all our errors quoted are standard deviations), in good agreement with the values found by Staehelin.[2] The ratio of the areas of the K-conversion lines is

$$N_e(331)/N_e(354)=0.381\pm0.005.$$

The unresolved $L+M$ line of the 426-kev transition in Hg^{196} allows an approximate determination of the beta-decay branching of Au^{196}. Using the value $\alpha_{L+M}(426)=0.009\pm0.003$ determined by Staehelin,[2] the measured value $\alpha_K(354)=0.042\pm0.003$ (see below), and the relative intensities of the respective conversion lines, one obtains a beta branching of 12$\pm$4%, if no electron capture to the ground state of Pt^{196} is assumed.

Photoelectrons

The ratio of the gamma-ray intensities was determined by measuring the photoelectrons from a 5-mg/cm^2 gold radiator. The ratio of the K-shell photoelectron cross sections for the two gamma rays was taken from Davisson and Evans[13] to be $\sigma(331)/\sigma(354)=1.196\pm0.005$. The resulting intensity ratio of the two gamma rays is

$$N_\gamma(331)/N_\gamma(354)=0.284\pm0.011.$$

Electron-Gamma Coincidences

Since the gamma-ray intensity ratio was found to be appreciably different from the earlier result,[1] a second, independent determination was made by measuring the probability that the 354-kev transition is preceded by a 331-kev gamma ray. The K-conversion electrons of the 354-kev transition were focused in the spectrometer (counting rate S) and coincidences (C) with the 331-kev gamma ray, detected by a NaI crystal behind the source, were recorded. The ratio of coincidences to spectrometer counts is given by

$$C/S=\epsilon(1-\kappa_{331})w\omega_\gamma.$$

Here, ϵ is the intensity ratio of the two transitions, κ_{331} the fraction of converted 331-kev transitions, and w a correction factor (1.068) for the angular correlation between the 354-kev conversion electrons and the 331-kev gamma rays. It was obtained by integrating the angular-correlation function $W(\vartheta)=1+0.129P_2(\cos\vartheta)$, calculated from the tables of Biedenharn and Rose,[14] over the solid angles subtended by the spectrometer and the NaI crystal according to the procedure given by Rose.[15] An admixture of 4.5% $M1$ radiation is assumed; the final result does not depend much on this assumption. The last factor, ω_γ, is the counting efficiency of the NaI crystal (for isotropically emitted gamma radiation). It is energy dependent and was measured, using the same technique, for the 354-kev line in Pt^{196}, and by β,γ coincidences for the 411-kev and the 279-kev gamma rays which follow the decays of Au^{198} and Hg^{203}, respectively. In these cases, the branching ratios, ϵ, are equal or close to unity. An angular-correlation correction (0.922) was applied only in the case of Pt^{196}; for Au^{198} the β,γ correlation is known

[12] J. W. Blue and E. Bleuler, Phys. Rev. **100**, 1324 (1955).

[13] C. M. Davisson and R. D. Evans, Revs. Modern Phys. **24**, 79 (1952).

[14] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1955).

[15] M. E. Rose, Phys. Rev. **91**, 610 (1953).

to be negligible,[16] and in the case of Hg^{203} it is presumably small since the beta spectrum has been found to have the allowed shape.[17,18]

With the gamma-ray sensitivity $\omega_\gamma(331)$ obtained by interpolation between these calibration points, the product $\epsilon(1-\kappa_{331})$ was found to be 0.245 ± 0.009. The ratio of the gamma-ray intensities is $N_\gamma(331)/N_\gamma(354)=\epsilon(1-\kappa_{331})/(1-\kappa_{354})$. The total conversion fraction of the 354-kev transition could be calculated from the theoretical K-conversion coefficient and the experimental ratio α_K/α_{L+M}. It was also determined directly with the aid of electron-electron coincidences.

Electron-Electron Coincidences

The K-conversion electrons of the 331-kev transition were focused in the spectrometer (S) and coincidences (C) with the conversion electrons of the lower transition were measured with the aid of an anthracene crystal placed about $\frac{1}{2}$ in. behind the source. The ratio C/S is given by

$$C/S=\kappa_{354}\omega_e w.$$

The detection efficiency of the anthracene crystal for electrons, ω_e, is essentially the solid-angle fraction subtended by the crystal at the source. It was determined by measuring coincidences between the conversion electrons of Au^{198} (where κ is known[19]) and the preceding beta particles. The angular-correlation correction, w, was calculated to be 0.923 for the coincidences between the two K-conversion electrons. The correlation functions for the outer-shell electrons are not available and the same correction factor was assumed.

With the total conversion fraction $\kappa_{354}=0.060\pm0.004$ obtained in this manner, the ratio of the gamma-ray intensities becomes $N_\gamma(331)/N_\gamma(354)=0.261\pm0.010$, in fair agreement with the value obtained from the photoelectron measurements.

By reversing the roles of the two transitions in the electron-electron coincidence measurements, one obtains an independent determination of $\epsilon\kappa_{331}$ (0.022 ± 0.002). The K-conversion coefficients are then calculated from the ratio of the K-conversion lines to the sum of all conversion lines which is proportional to $\kappa_{354}+\epsilon\kappa_{331}$. In this manner, uncertainties in the separation of the two broad $L+M$ lines are avoided.

[16] S. L. Ridgway, Phys. Rev. **78**, 821 (1950).
[17] S. C. Wilson, Phil. Mag. **42**, 762 (1951).
[18] N. Marty, Compt. rend. **240**, 291 (1955).
[19] P. E. Cavanagh, Phys. Rev. **82**, 791 (1951).

TABLE I. Conversion coefficients of the Pt^{196} gamma rays.

$E\gamma$	331 kev	354 kev
κ	0.079 ± 0.008	0.060 ± 0.004
α_K/α_{L+M}	$2.0\ \pm0.2$	$1.9\ \pm0.2$
$\alpha_{K,\mathrm{exp}}$	0.059 ± 0.004	0.042 ± 0.003
$\alpha_{K,\mathrm{th}}(E2)$	0.050	0.042
$\alpha_{K,\mathrm{th}}(M1)$	0.253	0.210

SUMMARY AND DISCUSSION

The results of the measurements are summarized in Table I.

The ratio of the gamma-ray intensities is taken as $N_\gamma(331)/N_\gamma(354)=0.272\pm0.010$, the intensity ratio of the transitions as $\epsilon=0.278\pm0.010$.

The comparison of the experimental and the theoretical K-conversion coefficients shows, as expected, that the lower gamma ray is an $E2$ transition, whereas the conversion of the 331-kev transition is too high for pure $E2$ radiation, indicating the admixture of $M1$ and/or $E0$ conversion.

The ratio of the two K-conversion coefficients is 1.40 ± 0.06 (standard error). This is in very good agreement with the value, 1.41 ± 0.04 (error limit; it is assumed here that errors in the theoretical conversion coefficients are negligible compared to the experimental uncertainties) expected for the admixture of $4.5\pm0.8\%$ $M1$ radiation found from the angular correlation of the two gamma rays. If one assumes this admixture to be correct, the fraction of conversion electrons of the 331-kev transition due to monopole radiation is $0\pm5\%$ (standard error). Accepting a generous upper limit of 15% for the fraction of monopole conversion electrons and a lifetime of 10^{-11} sec for the second excited state (an enhancement of the $E2$ transition by a factor 100), one obtains $\lambda_e(E0)<10^9$ sec^{-1} and a matrix element $M<0.1R^2$. It would seem desirable to measure the lifetime—which is probably longer—in order to obtain a more significant upper limit of M.

ACKNOWLEDGMENTS

The authors wish to express their appreciation to Dr. R. M. Steffen for suggesting the problem, to Dr. D. J. Tendam and Mr. F. Hobaugh for the cyclotron bombardments, and to Mr. F. Block for helpful discussions concerning the preparation of the sources. One of us (M. T. T.) is indebted to the Purdue Research Foundation for the grant of an XR fellowship.