Successive Neutron Capture in Gold*

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Transitions corresponding to 159 and 209 kev have been observed in neutron-activated gold. They have been observed to decay with a half-life of 3.3 days and have been ascribed to Au^{199}, which is formed by a process of successive neutron capture from Au^{197}. The thermal neutron capture cross section of Au^{198} has been estimated to be of the order of 10^4 barns.

THE 2.7-day beta-activity[1] of Au^{198}, produced by neutron capture from the single stable gold isotope Au^{197}, is well known[2] to have a continuous spectrum upper limit at 0.97 Mev and a conversion spectrum[3] from a 0.4112-Mev gamma-ray. Of somewhat disputed existence are two weak gamma-transitions of 0.157 and 0.208 Mev, observed by Levy and Grueling[4] and linked by them with a beta-spectrum having an apparent upper limit at 0.601 Mev. Other evidence in support of these lower energy transitions is reviewed in the paper by Levy and Grueling. However, more recent work by Siegbahn and Hedgran[5] and by Langer[6] indicated that the Au^{198} beta-spectrum was simple and no evidence of the lower energy transitions was found. Peacock and Wilkinson,[7] also, did not observe the transitions in deuteron-bombarded gold, and they believed that in earlier work the lines were due to an impurity of Hg^{197} which has gamma-transitions of 0.170 and 0.230 Mev.

Our experiments with neutron-activated gold were made originally with the aim of calibrating a magnetic beta-ray spectrograph against the 0.411 Mev conversion lines. In the course of these experiments we observed the low energy transition lines. They are difficult to detect, as Levy and Grueling pointed out, situated as they are in the most intense part of the strong continuous spectrum from Au^{198}. As far as photography is concerned, this leads to an optimum exposure of the film for visibility of the lines. Best contrast is also obtained when the source is thin, of the order of 0.2 mg/cm^2 or less. The sources were prepared by evaporation of gold on to thin aluminum backing before neutron bombardment in the pile.

A reproduction of the low energy part of the conversion spectrum, taken with a spectrograph using secondary electrostatic focusing,[8] is shown in Fig. 1. Although only the more intense lines are visible in the reproduction, there are at least fourteen lines visible on the best films. Of these lines, one of 63 kev was found to decay with a half-life of approximately 24 hours, and was attributed to W^{187} which undoubtedly arose as a contaminant from the tungsten heater from which the gold was evaporated in preparation. Two low energy lines of 33 and 35 kev were very weak and their existence was only observed on one exposure. They may arise from a 48 kev transition converted in the L_I, L_{III} sub-shells. The other lines can be ascribed to two transitions: one of 159 kev which is converted in the K, L_I, L_{III}, M and N shells of mercury; the other of 209 kev which shows only one line, presumably the K shell conversion.

Since it was previously claimed[4] that the low energy transitions decayed with a half-life of 2.7 day, we made an effort to determine a significant lifetime. By following the relative decays of the 411 and 159 lines over a period of many weeks, we have observed that the 159 lines decay more slowly than do the 411 lines. Decay plots for the 411-M and 159-L lines (which are of comparable intensity) are shown in Fig. 2. Their decays are consistent with half-lives of 2.7 day and 3.3 day, respectively. A 3.3-day decay is known to be associated with Au^{199} and recently Beach, Peacock and Wilkinson[9] have observed transitions of 24, 51, 70, 156, 207 and 230 kev from Au^{199} produced as a disintegration product from 31-minute Pt^{199}. We therefore assign our 159 and 209 kev transitions in neutron-activated gold to Au^{199}.

We have also observed that the low energy transitions are suppressed by enclosing the gold source in a cadmium box during neutron activation. In order to

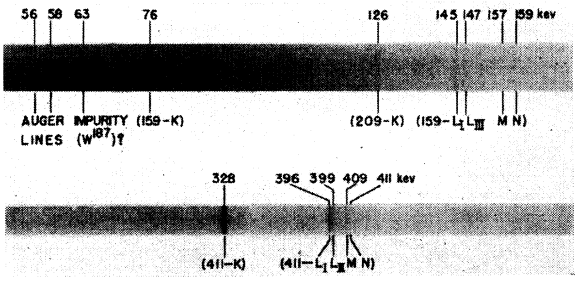

FIG. 1. Conversion electron spectra. Upper spectrum taken with $B=100$ gauss instrument and secondary electrostatic focusing. Lower spectrum taken with $B=200$ gauss instrument.

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make quantitative measurements of this effect, we obtained two identical sources and had them irradiated under the same conditions, except that one source was enclosed in $\frac{1}{16}$ inch cadmium. It was found that the 159 and 209 kev transition lines were absent, and certainly reduced in intensity by a factor of 10, in the cadmium shielded source; whereas the 411 kev lines were reduced in intensity only by a factor of approximately two. As will be shown later, these results cannot be accounted for by the presence of a platinum impurity in the gold, but can be attributed to the production of Au^{199} through a very large thermal neutron capture cross-section of Au^{198}. The remainder of the discussion will be concerned with obtaining an estimate of this cross section.

The intensity of the 411-K line relative to the 159-K line, for a particular gold source bombarded 8 days in the pile, was determined as 31 to 1, at a time immediately after removal from the pile. This figure does not represent the relative populations of the 411 and 159 kev excited states, but must be corrected for the magnitudes of the conversions. Although the latter have not been directly determined, we may obtain some idea of them from the conversion line intensities within a particular transition group. The experimental observations of these intensities are given in Table I.

The value of $N_K/N_L=2.1$ for the 411 transition is in reasonable agreement with Siegbahn and Hedgran's[5] value of 3. The theoretical value of N_K/N_L for a 2^2-electric transition of 411 kev in mercury is 3.8 and the K-conversion coefficient is 0.033. Siegbahn and Hedgran[5] obtain $\alpha_K=0.031$, Peacock and Wilkinson[7] a value $\alpha_K=0.025$ and Plesset[10] a value $\alpha_K=0.04$.

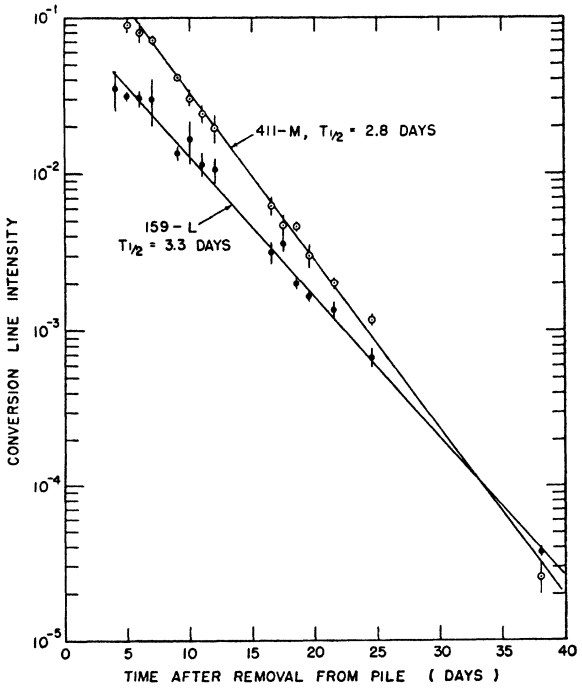

FIG. 2. Decay of conversion lines from neutron-activated gold.

TABLE I. Relative intensities of conversion lines.

Conversion line	411-K	411-L_I	411-L_{III}	411-M	411-N
Relative intensity	1.0	0.335	0.135	0.099	0.026
Conversion line	159-K	159-L_I	159-L_{III}	209-K	
Relative intensity	1.0	0.685	0.43	0.65	

The value of $N_K/N_L=0.87$ for the 159 transition agrees excellently with one of 0.8 obtainable from the Au^{199} spectrum published by Beach, Peacock, and Wilkinson.[9] The theoretical values of N_K/N_L lying closest to the experimental ones are: 0.5 for electric 2^3, 2 for electric 2^2 and 3 for magnetic 2^3 transitions. Thus, if electric 2^3 is the most probable assignment for the 159 transition, the theoretical K-conversion coefficient $\alpha_K=0.8$.

Thus a very tentative estimate of the 411 to 159 gamma-ray intensity ratio is

$$(N\gamma)_{411}/(N\gamma)_{159}=750.$$

Then, if we assume with Beach, Peacock, and Wilkinson[9] that the 159 kev transition occurs in 30 percent of the Au^{199} decays, and also that the 411 kev transition occurs in 100 percent of the Au^{198} decays, the population ratio of Au^{198} to Au^{199} is

$$(Au^{198})/(Au^{199})=95.$$

It is now clear that the Au^{199} cannot be produced from a Pt^{198} impurity in the gold sample. The isotopic thermal capture cross section of Pt^{198} for the formation of 31-minute Pt^{199} is known[11] to be 25 times less than that of Au^{197} for the formation of 2.7-day Au^{198}. Thus an isotopic impurity of 25/95, or approximately 26 percent, of Pt^{198} in Au^{197} would be required to produce the observed ratio of Au^{199} to Au^{198}. Since Pt^{198} is present only to the extent 7.2 percent in ordinary platinum that is clearly impossible; moreover chemical analysis of the gold showed no platinum present to within 10 parts in 10^6. This conclusion is also supported by the fact[12] that cadmium shielding reduces the 31-minute Pt^{199} activity to 75 percent, whereas our experiments indicated that the 159 and 209 kev transitions were certainly reduced to less than 10 percent, of their unshielded intensities.

The most likely process for the production of Au^{199} in neutron-irradiated gold is by neutron capture in radioactive Au^{198}. If we consider the following successive reactions:

$$Au^{197}+n=Au^{198}\xrightarrow{\beta^-}Hg^{198}+\gamma(411\text{ kev})$$

$$Au^{198}+n=Au^{199}\xrightarrow{\beta^-}Hg^{199}+\gamma(159,\ 209\text{ kev}),$$

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the relative populations of Au^{198} and Au^{199}, immediately after activation for a time T, are:

$$\frac{(Au^{199})}{(Au^{198})}=\frac{F\sigma}{\lambda_2-\lambda_1}\left\{1-\frac{\lambda_1(1-\exp-\lambda_2 T)}{\lambda_2(1-\exp-\lambda_1 T)}\right\},$$

where F is the activating neutron flux, σ is the total capture cross section of Au^{198}, and λ_1 and λ_2 are the decay constants of Au^{198} and Au^{199}, respectively. For a thermal flux of 10^{12} neutrons cm^2-sec., σ is evaluated to be about 3.5×10^4 barns.

It is well known that the $Au^{197}(n,\gamma)Au^{198}$ reaction has been used[13] to produce Hg^{198} in centigram amounts for use in discharge tubes as a source of light. Bradt and Mohler[14] found mass-spectrographically that one particular sample contained 0.16 percent of Hg^{199}, which they noted may well have come from a secondary nuclear reaction. Kessler[15] has also found the presence of forbidden Hg^{199} lines in the spectrum of mercury obtained from the neutron bombardment of gold. The amounts of Hg^{199} contaminant in the two samples prepared in this way were stated to be 3.6 and 0.3 percent respectively. Since the anticipated Hg^{199}/Hg^{198} ratio in a successive reaction process is $F\sigma/\lambda_1$, our values of σ and F would lead to a 1.2 percent Hg^{199} contaminant. It seems probable that the variation of Hg^{199} yield is due to different values of F, and thus the purest Hg^{198} can be obtained by using the lowest admissible neutron flux.

The gold sources were prepared at the Argonne National Laboratory, Chicago, Illinois. We wish to acknowledge our indebtedness to Dr. C. O. Muehlhouse for arranging for the source preparation, and to Dr. M. Goldhaber for helpful criticism of this work.

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Neutron Capture Gamma-Ray Multiplicity

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The average number of γ-rays, $\bar{\nu}_\gamma$, per neutron capture has been measured for about thirty isotopes. Correlations of $\bar{\nu}_\gamma$ with atomic weight and nuclear type are presented.

I. INTRODUCTION

WHEN a nucleus of atomic weight A absorbs a neutron, a compound nucleus of atomic weight $A+1$ is formed in a highly excited state (6 to 9 Mev). This excess energy may be released in the form of cascade γ-rays[1,2] involving many[3,4] levels of the compound nucleus. In any particular cascade process only a few of the total number of available levels are excited. Unless one or more of these levels is metastable, the total decay time is $\sim10^{-13}$ sec. The many levels immediately below the capture state (i.e., for ~1 Mev) are most likely not involved in the cascade process, since the probability for γ-ray emission is proportional to $E^{2(l-1)}$ (E is the transition energy, and l is the transition angular momentum change). This leaves a total number of levels, L, between which γ-ray transitions have a significant cascade probability. Also the number of γ-rays, ν_γ for any given capture process is such that $1\leq\nu_\gamma\leq L$. For example, let us take an oversimplified view of this process. If one allows all transitions from any given energy state to all lower energy states to have the same probability, then $\bar{\nu}_\gamma{}^2\simeq L$. Here $\bar{\nu}_\gamma$ is the average number of γ-rays per capture.

It is the purpose of this paper to present measurements of $\bar{\nu}_\gamma$ for various nuclei and to attempt to reveal the dependence of $\bar{\nu}_\gamma$ on atomic weight and nuclear type.

II. METHOD

Measurements of $\bar{\nu}_\gamma$ can be made in a number of ways. For example, with one γ-ray counter, a collimated neutron beam of flux, f, and n atoms/cm^2 of absorption cross section, σ_a, in the neutron beam, a γ-ray counting rate, s, results which is given by:

$$s=\epsilon f n\sigma_a a\bar{\nu}_\gamma.$$

Here ϵ is the over-all counting efficiency and a is the area of the neutron beam. Determination of $\bar{\nu}_\gamma$ by this method, however, depends on a knowledge of σ_a and f.

It is preferable to employ a method which is independent of uncertain nuclear data and difficult flux measurements. If two γ-ray counters in coincidence are used and both single, s, and coincidence, c, counting rates are recorded, then:

$$s_1=\epsilon_1 f n\sigma_a a\bar{\nu}_\gamma,\quad s_2=\epsilon_2 f n\sigma_a a\bar{\nu}_\gamma,$$

and

$$c=\epsilon_1\epsilon_2 f n\sigma_a a\bar{\nu}_\gamma(\bar{\nu}_\gamma-1).$$

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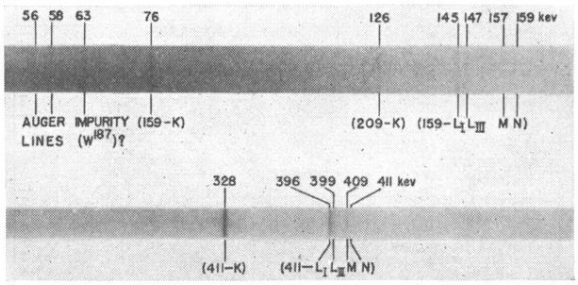

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