

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

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

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 $\text{cm}^{-2}\text{-sec.}$, σ is evaluated to be about 3.5×10^4 barns.

It is well known that the $\text{Au}^{197}(n, \gamma)\text{Au}^{198}$ reaction has been used¹³ to produce Hg^{198} in centigram amounts for use in discharge tubes as a source of light. Bradt and Mohler¹⁴ found mass-spectrographically that one particular sample contained 0.16 percent of Hg^{199} ,

¹³ J. H. Wiens and L. W. Alvarez, Phys. Rev. **58**, 11, 1005 (1940); **65**, 58 (1944).

¹⁴ P. Bradt and F. L. Mohler, Phys. Rev. **73**, 925 (1948).

which they noted may well have come from a secondary nuclear reaction. Kessler¹⁵ 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 $\text{Hg}^{199}/\text{Hg}^{198}$ 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.

¹⁵ K. G. Kessler, Phys. Rev. **77**, 559 (1950).

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 $\sim 10^{-13}$ 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

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

¹ Amaldi *et al.*, Proc. Roy. Soc. (A) **149**, 522 (1935).

² J. Griffiths and L. Szilard, Nature **139**, 323 (1937).

³ B. Hamermesh, Phys. Rev. **76**, 182A (1949).

⁴ Kinsey, Bartholomew, and Walker, Phys. Rev. **77**, 723 (1950).

Therefore:

$$c/\bar{s} = [2\epsilon_1\epsilon_2(\bar{\nu}_\gamma - 1)]/(\epsilon_1 + \epsilon_2).$$

The ratio of the coincidence to the average single counting rate is proportional to $\bar{\nu}_\gamma - 1$. Such a measurement is independent of cross section and of neutron flux. Furthermore, since at least one γ -ray is always emitted in every cascade event, this minimum of one γ -ray is, in effect, deducted from the average number, $\bar{\nu}_\gamma$. The measurement is more sensitive to the value of $\bar{\nu}_\gamma$.

One can, of course, continue along these lines and employ three or more counters in higher order coincidences. With three counters, the ratio of threefold to twofold coincidences is proportional to $\bar{\nu}_\gamma - 2$. Such a measurement is most sensitive to the value of $\bar{\nu}_\gamma$ in the case of a material which emits a minimum of two γ -rays in *each* cascade process.

The method employed in this work is that of making simple twofold coincidence measurements; i.e., one step beyond the zero-order measurement which depends on nuclear constants.

It should be remarked in passing that this method can be used to measure neutron absorption cross sections.² $\bar{\nu}_\gamma(1)$ and $\bar{\nu}_\gamma(2)$ are first measured for substances (1) and (2) respectively. If substance (1) has a known cross section, $\sigma(1)$, the unknown cross section, $\sigma(2)$ of substance (2) can be determined via the single counting rates $\bar{s}(1)$ and $\bar{s}(2)$:

$$\sigma(2) = \sigma(1) [\bar{s}(2)/\bar{s}(1)] \cdot [\bar{\nu}_\gamma(1)/\bar{\nu}_\gamma(2)].$$

III. APPARATUS

A collimated beam of neutrons from the Argonne heavy water reactor passes between two γ -ray scintillation counters in coincidence and through a thin neutron absorbing foil as shown in Fig. 1. Coincidence, c , and single, s_1 and s_2 , counting rates from the capture γ -ray bursts are recorded. Anthracene crystals having about 6 percent detection efficiency are cemented to each of the 5819 photo-multiplier tube windows. The

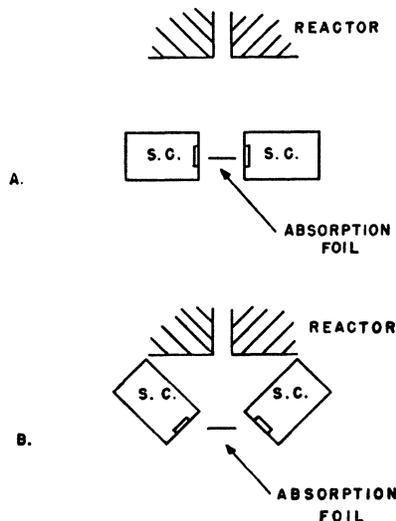


FIG. 1. Schematic diagram of counter and foil arrangement for measurement of capture γ -ray coincidences. (A) 180° apart. (B) 90° apart.

counters are partially shielded by bismuth from pile γ -rays, and operate at room temperature with a one-electron sensitivity. The mixer circuit is of a simple pulse addition type and has a resolving time of 0.15 μ sec.

IV. COUNTING TECHNIQUE

Measurements were taken for two different counter positions as illustrated in A and B of Fig. 1. This was done to indicate possible γ -ray angular correlations.

Foils were of such a thickness that γ -ray source strengths of $\sim 10^4$ /sec. resulted. The natural backgrounds of the counters were negligible. Other sources of background which were significant were as follows:

- (a) pile γ -rays independent of pile power,
- (b) pile γ -rays proportional to pile power,
- (c) capture γ -rays from scattered neutrons,
- (d) fortuitous coincidences resulting from single γ -ray scattering,
- (e) chance coincidences.

Expressions for the single and coincidence background counting rates contained terms proportional to time, monitor counts, and the scattering cross section of the material being studied. In addition, the coincidence background contained terms proportional to the average single counting rates (scattering) and the product of the single counting rates (chance). Single background counting rates ranged from 5 to 50 percent of the total effect. Calibration was effected with active 5-yr. Co⁶⁰ ($\nu_\gamma = 2$) and active 12.8-hr. Na²⁴ ($\nu_\gamma = 2$). Foils of these materials having the same area as the neutron beam were placed between the counters with the pile inoperative. The net ratio, c/\bar{s} , of coincidence to average single counting rate was measured, and "k" in the following expression was determined:

$$c/\bar{s} = k(\bar{\nu}_\gamma - 1) = k \text{ for } \nu_\gamma = 2.$$

Though the average γ -ray energy, \bar{E}_γ , is different for Co⁶⁰ (1.2 Mev) and Na²⁴ (2.1 Mev), the same value of k (~ 0.002) was obtained for each. This was fortunate since it indicated that the counters had approximately constant sensitivity in the energy range from 1 Mev to 2 Mev. As will be shown later $\bar{\nu}_\gamma \sim 3$ to 4 for most substances, and this gives $\bar{E}_\gamma \sim 1.5$ to 2.5 Mev. That is, most capture γ -rays fall in or near the energy range over which the counters have nearly constant sensitivity.

In all cases measurements were made with either thermal plus resonance neutrons (i.e., open pile beam) or resonance neutrons only (cadmium and boron filtered pile beam). This was done to investigate a possible dependence of $\bar{\nu}_\gamma$ on neutron energy.

In several instances a short-lived activity was induced in the sample by the neutron bombardment. Measurements were made after equilibrium had been reached. To illustrate how this situation was analyzed consider the case of $V^{51} + n \rightarrow V^{52}$ (3.7 min. β^- with one γ -ray). The extra delayed γ -ray could not coincide with the capture γ -rays thereby making the ratio c/\bar{s}

too small. That is:

$$(c/\bar{s}) \cdot (\bar{\nu}_\gamma + 1) / \bar{\nu}_\gamma = k(\bar{\nu}_\gamma - 1)$$

from which one can solve for the proper value of $\bar{\nu}_\gamma$. Dividing \bar{s} by $\bar{\nu}_\gamma + 1$ yielded the single counting rate per γ -ray. This figure was checked by turning the pile off and allowing the 3.7-min. γ -ray activity to decay. The total single γ -ray counts were recorded and the single counting rate per γ -ray was again computed. The two figures agreed to within 1 percent.

V. RESULTS

Results on the measurements of $\bar{\nu}_\gamma$ for twenty-seven isotope sets are given in Table I. Several important features should be noted:

(a) $\bar{\nu}_\gamma$ increases on the average with increasing atomic weight. This is to be expected for approximately constant binding energy and increasing level density.

(b) In the light element region, $\bar{\nu}_\gamma$ is approximately the same (~ 2.5) for odd Z -even N and even Z -odd N nuclei. In this same region, $\bar{\nu}_\gamma$ is less (~ 1.7) for even Z -even N nuclei. This observation is in agreement with the fact that neutron resonance levels are much more widely separated in the compound nucleus resulting from even Z -even N neutron capture than in compound nuclei resulting from odd Z or odd N neutron capture.⁵

(c) $\bar{\nu}_\gamma$ is low for a closed shell nucleus⁶ (such as La^{139} with 82 neutrons). This observation is in agreement with the fact that the neutron level spacing of compound nuclei resulting from closed shell nuclear neutron capture is large.⁵

(d) The isotope with the largest value of $\bar{\nu}_\gamma$, (${}_{62}\text{Sm}^{149}$ for which $\bar{\nu}_\gamma = 5.6$) is of the even Z -odd N type. It has one and a half more γ -rays than the isotope (${}_{48}\text{Cd}^{113}$ for which also Z is even and N is odd) having the next largest number of γ -rays.

Other important observations made in this work are as follows:

(e) No angular γ -ray correlations were observed. That is $\bar{\nu}_\gamma$ (position A) = $\bar{\nu}_\gamma$ (position B). This result is not surprising since a typical value of $\bar{\nu}_\gamma$ is ~ 3 . This implies about 10 important levels (i.e., $\bar{\nu}_\gamma^2$) leading to many different modes of cascade. Such a complex decay scheme is likely to average all individual correlations to about zero.

(f) $\bar{\nu}_\gamma$ measured with and without cadmium plus boron filtering (see conclusion of Section IV) was observed to be equal for the three cases studied. These

⁵ Harris, Muehlhause, and Thomas, Phys. Rev. **79**, 11 (1950).

⁶ M. Mayer, Phys. Rev. **75**, 1969 (1949).

TABLE I. Values of $\bar{\nu}_\gamma$.

(odd, even) →(odd, odd)	Nuclear types		Comment
	(even, odd) →(even, even)	(even, even) →(even, odd)	
${}_{11}\text{Na}^{23}$ <2	—	—	—
${}_{13}\text{Al}^{27}$ ~ 2	—	—	—
${}_{17}\text{Cl}^{35}$ 3.1	—	—	for KCl
${}_{23}\text{V}^{51}$ 2.5	—	—	—
—	${}_{24}\text{Cr}^{53}$ 2.3	${}_{24}\text{Cr}$ <2	—
${}_{25}\text{Mn}^{55}$ 2.6	—	${}_{26}\text{Fe}$ 1.7	therm. and res.
—	—	—	—
${}_{29}\text{Cu}$ 2.6	—	—	—
—	${}_{32}\text{Ge}^{73}$ ~ 4	${}_{32}\text{Ge}$ 2.0	73 impure? therm. and res. large res. abs.
${}_{33}\text{As}^{75}$ 2.7	—	—	—
${}_{35}\text{Br}$ 3.4	—	—	—
${}_{41}\text{Nb}^{93}$ 2.6	—	—	—
${}_{47}\text{Ag}$ 2.9	—	—	—
—	${}_{48}\text{Cd}^{113}$ 4.1	—	—
${}_{49}\text{In}^{115}$ 3.3	—	—	—
${}_{57}\text{La}^{139}$ 2.7	—	—	82 neutrons maximum
—	${}_{62}\text{Sm}^{149}$ 5.6	—	—
${}_{63}\text{Eu}$ 3.8	—	—	—
—	${}_{64}\text{Gd}$ 3.9	—	—
—	—	${}_{66}\text{Dy}^{164}$ 3.7	—
—	—	${}_{72}\text{Hf}$ 3.8	even- N abs.
${}_{75}\text{Re}$ 3.2	—	—	—
${}_{77}\text{Ir}$ 3.6	—	—	—
${}_{79}\text{Au}$ 3.5	—	—	therm. and res.
—	${}_{80}\text{Hg}$ 3.3	—	—

were ${}_{25}\text{Mn}^{55}$, ${}_{33}\text{As}^{75}$, and ${}_{79}\text{Au}^{197}$. With some uncertainty in the case of manganese, the thermal absorption cross section can be calculated from the resonance parameters of the principal resonance level⁷ at 345 ev. However, one might suspect that the thermal absorption cross section is compounded of a different proportion of spin states ($J=2$ or 3) than is the resonance absorption cross section (principally $J=3$). This, in turn, might lead to different $\bar{\nu}_\gamma$ values for thermal and resonance absorption. In the case of gold no such difference in $\bar{\nu}_\gamma$ is to be expected, since the principal resonance level⁸ at 4.8 ev does yield the proper thermal absorption cross section. That is, thermal energies lie in the wing of the 4.8 ev state. In none of the three cases was $\bar{\nu}_\gamma$ found to depend on neutron energy. Doubtless neutron absorption at energies of around 1 Mev would result in a different value of $\bar{\nu}_\gamma$ from that at thermal or at low energy resonance absorption, since an extra 1 Mev is carried into the nucleus.

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⁷ High speed Argonne resonance region time-of-flight data by W. Selove.

⁸ Havens, Wu, Rainwater, and Meaker, Phys. Rev. **71**, 165 (1947).