Scintillation Spectrometer Determination of L-Capture to K-Capture Ratio in Cd^{109} and I^{125}†

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A useful technique of scintillation spectrometry developed in this laboratory—that of growing crystals containing radioactive nuclei—was employed to measure the L-capture to K-capture ratio in the orbital electron capture occurring in Cd^{109} and I^{125}. Marshak's formulas were used to calculate the energy of the transition. The L-capture to K-capture ratio and the energy of the transition are reported as 0.28 ± 0.03 and 73_{-6}^{+12} kev for Cd^{109}, and 0.23 ± 0.03 and 108_{-10}^{+20} kev for I^{125}, respectively.

INTRODUCTION

WHEN a nucleus decays by means of orbital electron capture, a knowledge of the L-capture to K-capture ratio is significant because this ratio is a function of the order of and the energy available for the transition.[1] If the total disintegration energy is known (as would be the case if positron emission competed with orbital electron capture) as well as the spin and parity change, the experimentally determined L-capture to K-capture ratios can be compared with calculated values as a check of β theory.[2] On the other hand, as belief in β theory strengthens, the L-capture to K-capture ratio becomes a useful and important tool in the study of the systematics of energy levels, for it offers a way to determine the energy differences between states connected by orbital electron capture alone.

Not many experimental data exist on L-capture to K-capture ratios, the reason being that the detection and absolute measurement of soft x-radiations has been experimentally difficult.[3] A technique developed in this laboratory,[4] whereby radioactivity is introduced into a NaI scintillating crystal, lends itself readily to the study of L-capture to K-capture ratios by providing one with a detector which has no window corrections for soft radiations and, at the same time, has an efficiency of approximately 100 percent.

EXPERIMENTAL DETAILS

The method consists of growing a thallium-activated sodium iodide single crystal from a mass of sodium iodide to which has been added a trace of the radioactive material under investigation. The single crystal of sodium iodide which is obtained in this manner contains evenly dispersed throughout its volume a source of radiations which is for practical purposes of zero thickness. If the ratio of the weight of radioactive matter to thallium activator is kept small (<10 percent) the crystal exhibits in most cases scintillation properties equal to those of an ordinary sodium iodide scintillator. Observations are made with the crystal mounted on a photomultiplier tube connected through an Atomic Instrument Company type 204B linear amplifier to either a Dumont type 301 oscilloscope (for photographic recording) or an Atomic Instrument Company single channel pulse-height analyzer.

The chief source of error in this method for determining this ratio is the escape of photons from the crystal. If large crystals (1 cm^3) are used, the escape of x-rays for atoms of low or intermediate Z will be small and approximate corrections can be made.

Cd^{109}

The radioactive isotope Cd^{109} decays by orbital electron capture to the first excited state of Ag^{109}, which in turn decays through the emission of a highly converted 87-kev gamma ray to the ground state.[5] The half-life of Cd^{109} is reported to be 330 days or greater. The first excited state of Ag^{109} is metastable and has a half-life of 39.2 sec. Orbital electron capture leading to the ground state of Ag^{109} has not been reported, and the probability for such a transition is small for any spin assignment made to the Cd^{109} in view of the spins assigned to the first excited state and to the ground state of Ag^{109} (7/2 and 1/2, respectively). Positron emission has been looked for and not seen.[6] Thus each orbital electron capture has associated with it an 87-kev gamma ray, which is internally converted over 90 percent of the time.

When a source of Cd^{109} is dispersed throughout a crystal by the above described method and the pulses resulting in the crystal are observed with a pulse-height analyzer, one sees a pulse peak which corresponds to the conversion electrons of the 87-kev gamma ray and another peak which corresponds to the K x-ray which accompanies the K-electron capture (Fig. 1). Although both these pulses originate in the detector and the

† Work carried out under the auspices of the U. S. Atomic Energy Commission.
[1] R. E. Marshak, Phys. Rev. **61**, 431 (1942).
[2] P. Radvanyi, Compt. rend. **235**, 428 (1952).
[3] See for example, Pontecorvo, Kirkwood, and Hanna, Phys. Rev. **75**, 982 (1949); G. Friedlander and W. L. Orr, Phys. Rev. **84**, 484 (1951).
[4] Scharff-Goldhaber, der Mateosian, Goldhaber, Johnson, and McKeown, Phys. Rev. **83**, 480 (1951).
[5] References are given in Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953); M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).
[6] Dreyfus, Major, and Radvanyi, Compt. rend. **232**, 617 (1951).

detector has a 4π solid angle geometry, the x-rays and the 87-kev gamma rays are not added together in the crystal since the two pulses are usually separated in time by an interval greater than the resolving time of the circuit. Because of the high degree of conversion of the Ag^{109m} internal transition and low loss of 87-kev gamma rays from the crystal, the area under the 87-kev gamma-ray peak may be taken as a measure of the total number of orbital electron captures in a given unit of time, regardless of whether capture takes place in the K, L, or a higher shell. The area under the K x-ray peak is a measure of the K-capture rate and by difference one may obtain the L-capture rate, assuming M and N capture to be negligible. The experimentally obtained L-capture to K-capture ratio is 0.28 ± 0.03.

I^{125}

The decay of 60-day I^{125} is very similar to that of Cd^{109}, proceeding by orbital electron capture[5] to the

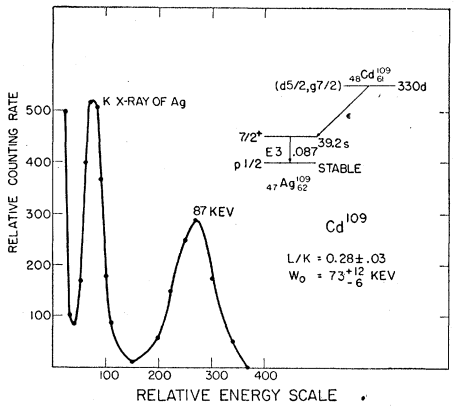

FIG. 1. Soft photons of Cd^{109} observed with a scintillation spectrometer. The output of a single channel pulse-height analyzer is plotted as a function of energy. Two peaks are observed, one corresponding to the K x-rays of Ag resulting from the orbital electron capture in Cd^{109} and the other corresponding to the 87-kev transition in Ag^{109} following the orbital electron capture.

first excited state of Te^{125}. Unlike Ag^{109m} however, the first excited state of Te^{125} is not metastable so that the internal transition to the ground state (35 kev) and the x-radiations accompanying orbital electron capture are not resolved and one sees, in a pulse-height analysis of the pulses resulting from a crystal containing I^{125}, peaks corresponding to the addition of conversion electrons and x-rays. This is a fortunate state of affairs, because both the L x-ray peaks and K x-ray peaks are shifted by 35 kev to higher positions on the energy axis, rendering both peaks resolvable above instrument noise (see Fig. 2). The ratio of the areas under the L x-ray peaks and K x-ray peaks is a direct measure of the L-capture to K-capture ratio, which in this case is 0.23 ± 0.03.

TRANSITION ENERGIES

The calculation of transition energies from L-capture to K-capture ratios was based upon formulas developed

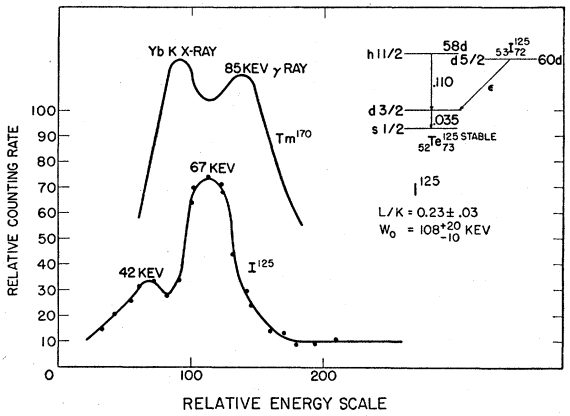

FIG. 2. Soft photons of I^{125} observed with a scintillation spectrometer. The output of a single channel pulse-height analyzer is plotted as a function of energy. Two peaks are observed, one corresponding to the addition of 35-kev pulses and L x-rays of Te and the other corresponding to the addition of 35-kev pulses and K x-rays of Te. The upper curve shows the radiations of Tm^{170} which were used to calibrate the instrument.

by Marshak[1] for the lifetime for orbital electron capture. In an allowed transition the relationship between the L-capture to K-capture ratio and the transition energy may be written as

$$R_{L/K}=\left[\frac{\Psi_{LI}}{\Psi_K}\right]^2\left[\frac{W_0+W_L}{W_0+W_K}\right]^2,$$

where $R_{L/K}$ is the L-capture/K-capture ratio, Ψ_{LI}/Ψ_K is the ratio of L- and K-shell electron densities at the nuclear radius and W_0, W_L, and W_K are the transition energy, the L-shell energy, and the K-shell energy, respectively.[7] Values for the quantity $[\Psi_{LI}/\Psi_K]^2$ as a function of Z have been published by Rose and Jackson.[8] Table I gives a summary of our data and results. It is of interest to note that in the region where $(W_0+W_L)/(W_0+W_K)$ is appreciably different from 1, W_0 is a slowly varying function of $R_{L/K}$, hence sizeable experimental errors can be tolerated in the L-capture to K-capture ratio.

TABLE I. Measured L-capture to K-capture ratios and calculated transition energies for orbital electron capture in Cd^{109} and I^{125}.

Isotope	L-capture to K-capture ratio	Transition energy (kev)[a]	Log(ft)
Cd^{109}	0.28 ± 0.03	73_{-6}^{+12}	5.1
I^{125}	0.23 ± 0.03	108_{-10}^{+20}	4.8[b]

[a] Values for W_K and W_L in these calculations were computed from the expressions given by Marshak (reference 1): $W_K=1-\frac{1}{2}\alpha^2Z_{\text{eff}}^2$ and $W_L=1-\frac{1}{8}\alpha^2Z_{\text{eff}}^2$ where $Z_{\text{eff}}=Z-0.3$.

[b] A slightly higher value for log(ft) was reported for I^{129} by G. Friedlander and W. L. Orr, who extrapolated S. A. Moszkowski's nomograph [Phys. Rev. **82**, 35 (1951)] to make their calculation.

[7] Marshak defines W_K as $1-\frac{1}{2}\alpha^2Z_{\text{eff}}^2$ and W_L as $1-\frac{1}{8}\alpha^2Z_{\text{eff}}^2$, where $Z_{\text{eff}}=Z-0.3$ and $\alpha=1/137$.

[8] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

DISCUSSION OF RESULTS

Friedlander and Orr[3] determined the L-capture to K-capture ratio in I^{125} by using a windowless proportional counter to compare the ratio of 35-kev gamma rays to K x-rays in an I^{125} source with the same ratio in a Te^{125} source. They reported a transition energy of 80_{-18}^{+160} kev, which is confirmed by our measurement. As an additional point of reassurance, these authors calculated $\log(ft)$ values for the K capture and showed that the energy was consistent with an allowed transition. The $\log(ft)$ for Cd^{109} calculated by means of a formula appearing in Feenberg and Trigg's[9] article, appears in Table I. It also is compatible with an allowed transition. In both cases the classification of the transition as allowed agrees with spin assignments for the initial and final states of the nuclei involved.[10]

The author wishes to express his appreciation to Dr. M. Goldhaber for his interest in this problem and for many helpful suggestions generously offered.

[9] E. Feenberg and G. Trigg, Revs. Modern Phys. **22**, 399 (1950).
[10] M. Goldhaber and R. D. Hill, Revs. Modern Phys. **24**, 179 (1952).

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Excitation Function for the Photodisintegration of Beryllium*

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Using filtered betatron bremsstrahlung, the $Be^9(\gamma,n)$ excitation function has been determined from threshold to 24 Mev. The results show two peaks, one due to excitation of the odd neutron and the second to the excitation of the Be^8 core.

THE photoneutron disintegration cross section of beryllium in the vicinity of the threshold (1.6 Mev) has been measured by several investigators,[1] and the results are in substantial agreement with the calculations of Guth and Mullin[2] for $P\rightarrow S$ and $P\rightarrow D$ transitions of a neutron moving in the field of a Be^8 core. The photoproton excitation function,[3] as determined by successive subtractions of yields produced by betatron bremsstrahlung, shows the characteristic dipole resonance found in other elements. Determination of the (γ,n) excitation function from threshold to 24 Mev by this method is of particular interest but is difficult because of the low threshold and the preponderance of low-energy photons in the bremsstrahlung, spectrum when the betatron is run at high energies.

We have measured the neutron yield as a function of betatron energy by the method of direct neutron detection[4] for betatron bremsstrahlung filtered by 30 cm of carbon absorber to flatten the bremsstrahlung distribution. Figure 1 shows the calculated bremsstrahlung curves at 10 and 20 Mev, normalized to 100r, with and without the carbon filter. As a test of the method, the bismuth (γ,n) yield curve[5] was repeated using filtered radiation, and the excitation functions computed therefrom are shown in Fig. 2. The data of reference 5 are drawn for comparison. Aside from a 20 percent discrepency in the absolute values of the cross sections, the data taken with the hardened beam are in good agreement with the previous work.

Figure 3 shows the excitation function for beryllium computed from yield data using the filtered beam. Also shown are the $Be^9(\gamma,p)$ data of reference 3, and

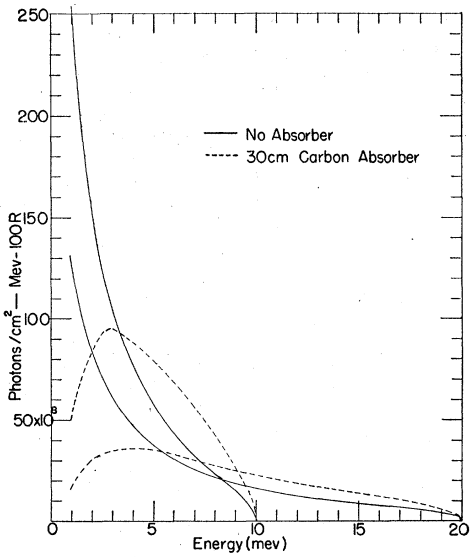

FIG. 1. Theoretical bremsstrahlung distributions for betatron energies of 10 and 20 Mev with and without 30-cm carbon absorber.

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[1] Russell, Sachs, Wattenberg, and Fields, Phys. Rev. **73**, 545 (1948), B. Hamermesh and C. Kimball, Phys. Rev. **90**, 1063 (1953).
[2] E. Guth and C. J. Mullin, Phys. Rev. **76**, 234 (1949).
[3] R. N. H. Haslam *et al.*, Can. J. Phys. **31**, 210 (1953).
[4] Halpern, Mann, and Nathans, Rev. Sci. Instr. **23**, 678 (1952).
[5] Halpern, Nathans, and Mann, Phys. Rev. **88**, 679 (1952).