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Gamma-Ray Absorption Coefficients for NaI, Cu, Ta, and W†

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Gamma-ray absorption coefficients have been measured over a range of energies from 0.279 Mev through 1.113 Mev for NaI, Cu, Ta, and W, using an energy-selective scintillation detector. Measurements were made for the absorber position which gave the maximum coefficient, and the results are in good agreement with theory.

INTRODUCTION

SEVERAL recent papers[1–5] have reported experimental gamma-ray absorption coefficients in the region of 1 Mev which are consistently several percent below theoretical values for heavy elements above tin in the periodic table. Since the Klein-Nishina cross section is highly reliable and the pair cross section is small, the theoretical photoelectric cross section would have to be reduced from 10 to 15 percent in order to agree with experiment in this region.

Rudnick,[6] in reinvestigating the absorption coefficient of tantalum for the two Co^{60} gamma rays, found a definite dependence of absorption coefficient on absorber position. Agreement was found between the experimental and theoretical values when the measurement was made at the position which gave the maximum absorption coefficient. It is Rudnick's conclusion that the position of maximum absorption coefficient gives the most reliable experimental result because this position minimizes the various scattering errors.

In the present work several absorption coefficients near 1 Mev have been investigated with an improved experimental technique in an attempt to ascertain whether there are any significant differences between experimental and theoretical values that cannot be assigned to experimental error. It was felt that an improvement in experimental technique could be obtained by employing the scintillation detector method of Colgate[4] and by making experimental measurements in the position giving maximum absorption coefficient, within the limits of the geometry concerned. It should be noted that Colgate's experimental measurements at 0.6616 Mev, made by measuring absorption coefficients as a function of position and taking the maximum value, gave good agreement with theory. However, his measurements at 1.33 Mev, using Co^{60}, were made under a fixed geometry which was apparently not the position of maximum absorption coefficient. However, even when theoretical scattering corrections were applied, the experimental values for elements above tin in the periodic table ($Z > 50$) were lower than the theoretical values by amounts several time the experimental error. It is of course possible that insufficient allowance was made for the amount of 1.173-Mev gamma rays detected.[3]

EXPERIMENTAL EQUIPMENT

Detector and Collimator

Figure 1 is a schematic of the collimating apparatus. It is essentially the same as that described by Davisson and Evans[1] with the exception that the Geiger counter is replaced by a scintillation detector. A 1.5-in diameter NaI(Tl) crystal, together with a RCA 5819 photomultiplier is used as the detecting element. A resolution of 11.3 percent was obtained for the Cs^{137} gamma ray at 0.6616 Mev. The output of the photomultiplier was fed into a Radiation Instrument Corporation non-overloading amplifier and then into a Johnstone[7] single-channel differential pulse-height analyzer. The counting channel had a voltage width of ten percent of the voltage at the photopeak maximum. Initially, this channel was adjusted to have the photoelectric peak maximum at its center. The counting rate in the channel was recorded for various absorber thicknesses. An energy-selective detector of this type has a high detecting efficiency for the primary gamma-ray quanta and rejects all scattered radiation except that scattered through small angles. By setting the channel width base line on or above the photoelectric peak, a further reduction in the amount of scattered radiation detected is effected.

Careful regulation and monitoring of the line and photomultiplier voltages insured a reproducibility of counting rate as good as the statistics used in the experiment for periods up to 15 minutes. Long-time counting rate drifts were no greater than ±1 percent per hour under optimum conditions.

The equipment reached temperature equilibrium after a three-hour warmup and was always operated under equilibrium conditions.

As a result of good detector shielding, the background counting rates were less than one part in several thousand and no background corrections were required.

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† Supported by Bureau of Ships, U. S. Navy.

[1] C. M. Davisson and R. D. Evans, Phys. Rev. **81**, 404 (1951).

[2] Shimizu, Hanai, and Okamoto, Phys. Rev. **85**, 290 (1952).

[3] G. R. White, Natl. Bur. Standards (U. S.) Rept. No. 1003, May 13, 1952 (unpublished).

[4] S. Colgate, Phys. Rev. **87**, 592 (1952).

[5] S. J. Wyard, Proc. Phys. Soc. (London) **66**, 382 (1953).

[6] N. Rudnick, Mass. Inst. Technol. Laboratory for Nuclear Science and Engineering, Progress Report, May 31, 1952 (unpublished), p. 180.

[7] C. W. Johnstone, Nucleonics **11**, No. 1, 36 (1953).

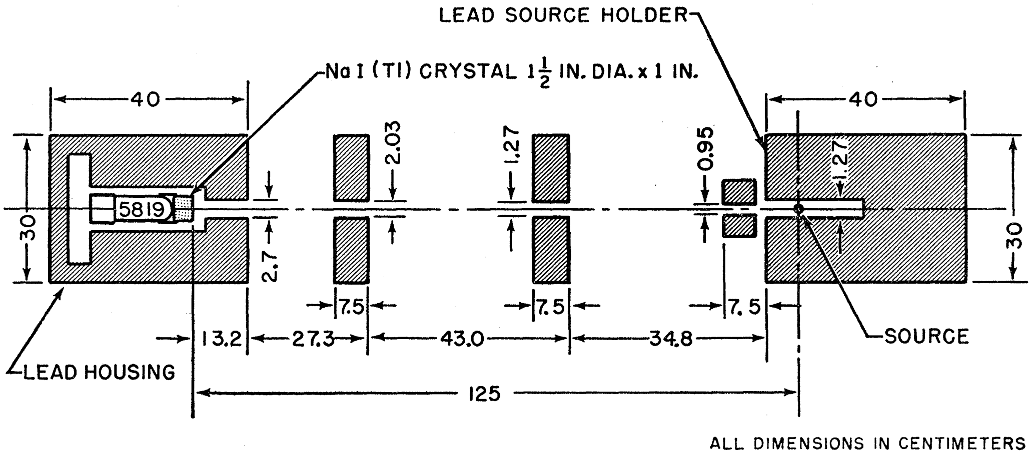

FIG. 1. Geometrical arrangement of collimators.

Counting rate linearity was checked by the method of adding sources and the maximum counting rates were kept in the linear range.

Sources

Radioactive sources obtained from Oak Ridge were placed in Pyrex glass vials of 7-mm outside diameter and 1-mm wall thickness with the exception of the Zn^{65} source whose glass vial had an outside diameter of 8 mm. The tips containing the radioactive material were rounded and of the same wall thickness as the cylindrical sides.

The Hg^{203} source was 50 mC of high specific activity mercury metal reduced from an acid solution with metallic zinc. It was approximately 1 cm in length.

The Au^{198} source was 50 mC of gold foil weighing 0.016 g. The foil was rolled and placed within the tip of its glass container.

The 50-mC source of Cs^{137} was in the form of an evaporated solid. The active portion was less than 3 mm in length.

The Zn^{65} source was 40 mC in strength and was also an evaporated solid extending to a length of about 0.7 cm in its glass vial.

Absorbers

Absorber densities were determined to an estimated accuracy of 0.04 percent. The densities were determined from the dimensions and weights of the various absorbers after machining to the desired sizes. The copper absorbers were made from electrolytic copper bar and were 1 in. in diameter. Spectrochemical analysis showed only trace impurities. The sodium iodide obtained from the Harshaw Chemical Company was in the form of small blocks 2×2×1 in. The density in this case was taken as 3.667 g/cc.[8] The blocks were sealed in light polyethylene bags during the measurements. The absorption coefficient correction due to the bags was found to be negligible. The tantalum and wolfram absorbers were obtained from the Fansteel Metallurgical Corporation and were ground to a diameter of ¾ in. They were certified pure to at least 99.9 percent.

EXPERIMENTAL RESULTS

Table I compares experimental and theoretical absorption coefficients.

Since good agreement between theoretical and experimental absorption coefficients for copper has been reported,[1–5] copper absorption coefficients were measured as a check on the reliability of the present experimental equipment. No significant dependence of absorption coefficient on absorber position was noted in the case of copper. This indicated that the degradation of the primary gamma rays by the collimators was negligible since the amount of collimation between source and absorber increased as the absorber was moved nearer the detector. Similar results were presented by Rudnick.[6] There was no measurable dependence of the copper absorption coefficient on the position of the pulse-height analyzer channel with respect to the photoelectric peak. The results of the copper measurements indicated satisfactory operation of the equipment. The measurements for thallium-activated sodium iodide were included because of the widespread use of sodium iodide as a gamma-ray detector. Good agreement with theory is obtained. Bell[8] measured slightly lower values under a somewhat different geometry.

A definite dependence of absorption coefficient on absorber position was found in the case of tantalum and wolfram at both 0.6616 Mev and at 1.113 Mev. The coefficients were measured at the position giving the maximum value. Some dependence of absorption coefficient on channel position was also noted and for this reason the channel base line was set at the highest energy portion of the photoelectric peak consistent with

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[8] Bell, Davies, Hughes, Jordan, and Randall, Oak Ridge Laboratory Rept. No. 1415, 1953 (unpublished).

TABLE I. Comparison of experimental and theoretical gamma-ray absorption coefficients.

Energy Mev	Element	Position[a] cm	Meas cm²/g	Experimental Correction	Final cm²/g	Theoretical White cm²/g	Theoretical Davisson and Evans cm²/g
0.279 (Hg^{203})	NaI	83.5	0.182 ±0.001	0.0009	0.183 ±0.001	0.183	0.195
0.4118 (Au^{198})	W	58.5	0.1736±0.0005	0.0029	0.1765±0.0005	0.1768	0.1821
0.6616	Cu	83.5	0.0724±0.0002	0.0003	0.0727±0.0002	0.0724	0.0725
(Cs^{137})	NaI(Tl)	83.5	0.0759±0.0003	0.0006	0.0765±0.0003	0.0763	0.0763
	Ta	58.5	0.0925±0.0003	0.0010	0.0935±0.0003	0.0936	0.0959
	W	62.0	0.0937±0.0002	0.0011	0.0948±0.0002	0.0955	0.0970
1.113	Cu	83.5	0.0555±0.0003	0.0002	0.0557±0.0003	0.0554	0.0556
(Zn^{65})	NaI(Tl)	83.5	0.0541±0.0003	0.0003	0.0544±0.0003	0.0541	0.0551
	Ta	45.5	0.0598±0.0002	0.0006	0.0604±0.0002	0.0603	0.0606
	W	45.5	0.0606±0.0002	0.0007	0.0613±0.0002	0.0607	0.0611

[a] Distance from detector face to absorber face nearest detector.

good statistics. This apparently reduced the amount of scattered radiation detected and tended to give slightly higher coefficients.

THEORETICAL VALUES OF THE ABSORPTION COEFFICIENTS

Theoretical values for the absorption coefficients were obtained by interpolation, using the results of Davisson and Evans[9] and White.[10] Both these papers give calculations of absorption coefficients for gamma rays following the theoretical formulas of several authors. The absorption processes considered are photoelectric effect, Compton scattering as given by the Klein-Nishina formula, pair production in the field of the nucleus, and Rayleigh or coherent scattering. All other processes by which photons interact with matter have been neglected as being too small to effect the results. These include radiative corrections to the Klein-Nishina formula and others which have been discussed by various authors.[1–5]

Interpolations for energies and elements used in the experiment are made for the photoelectric and pair production partial absorption coefficients from accurate plots of the values quoted by Davisson and Evans.[9] The Compton scattering partial absorption coefficients were calculated using their formula 19. To these three partial absorption coefficients we have added a calculated coherent scattering coefficient[11] to give total absorption coefficients for the elements and energies used. The values for NaI(Tl) were obtained from the coefficients of Na, I, and Tl using formula 64 of reference 9 to calculate the values for the crystal. A 0.2 percent Tl content by weight was assumed.

The values according to White[10] are interpolations from plots of $(\mu/\rho)A$ *versus* Z at energies from 0.2 Mev to 1.5 Mev as taken from White's tables. From these plots the values of $(\mu/\rho)A$ are taken at the Z's of interest and plotted against energy. From these curves the values for the total absorption coefficients are obtained at the energies and for the elements used. Values for NaI are taken from a plot of White's values of μ/ρ for NaI.

CORRECTIONS

The theoretical absorption coefficients with which the experimental results are compared are calculated on the basis that any absorption or scattering process that acts on a gamma-ray photon in the absorber removes it from detection. However, the radiation detected actually includes a small amount of scattered radiation from the absorber. Although energy discrimination is used, it is possible to detect photons which have been scattered through as much as 15° under these energy discrimination conditions. Collimation reduces this scattering angle in the absorber to about 2° for scattered radiation which is detected, and it is therefore necessary to correct the intensity detected for that amount which undergoes Compton and coherent scattering in the absorber.

If I_0 is the intensity reaching the detector with no absorber present and I_1 and I_2 are the intensities reaching the detector with absorbers 1 or 2, respectively, present, then $I_1=I_0\exp(-\mu x_1)+I_{s1}$, where $I_{s1}=$ intensity scattered into detector by Compton and Rayleigh scattering and $\mu=$ total absorption coefficient. Also

$$I_2=I_0\exp(-\mu x_2)+I_{s2},$$

so that

$$\ln(I_1/I_2)=-\mu(x_1-x_2)+\ln(1+s_1)/(1+s_2),$$

where

$$s_1=I_{s1}/I_0\exp(-\mu x_1),$$

and

$$s_2=I_{s2}/I_0\exp(-\mu x_2),$$

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

[10] Gladys R. White, Natl. Bur. Standards Rept. No. 1003, 1952 (unpublished).

[11] W. Franz, Z. Physik **98**, 314 (1935).

with the result that

$$\mu=1/(x_2-x_1)\{\ln(I_1/I_2)-\ln(1+s_1)/(1+s_2)\}.$$

The values for s_1 and s_2 are calculated using the method described in reference 1 and 4. These scattered intensities include the coherent and Compton-scattered radiation and depend on the maximum angle of scatter in the absorber of a photon which can reach the detector. Values of $\ln(1+s_1)/(1+s_2)$ are given in Table I. These corrections were minimized experimentally by placing the absorber in the position which gave the largest effective absorption coefficient.

DISCUSSION

The results of this experiment using differential energy selection indicate very good agreement with theory when the absorption coefficient is maximized by appropriate positioning of the absorber.

As a result it seems likely that the present theoretical expression[12] for the photoelectric cross section is reliable near 1 Mev for heavy elements through $Z=74$ (wolfram).

The authors desire to thank Mr. R. A. Taylor for invaluable help with regard to electronic problems.

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[12] Hulme, McDougall, Buckingham, and Fowler, Proc. Roy. Soc. (London) **A149**, 131 (1935).

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New Isotopes of Niobium: Nb^{89} and Nb^{89m}†

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A new 1.9±0.3-hour activity was found in niobium fractions isolated from proton irradiated niobium and zirconium metal foils. By identification of the 78-hour daughter activity, the mass assignment has been made to Nb^{89}. Evidence indicates that the decay is a simple positron emission of 2.9±0.4 Mev maximum energy. Evidence was also obtained for the existence of the $\frac{1}{2}-$ isomeric state in Nb^{89} which decays with a 0.8±0.3-hour half-life by positron emission to the similar 4.4-minute state in Zr^{89}.

OBSERVATION of the niobium fraction obtained from ~60-Mev proton bombardment of niobium metal in the internal beam of the Harvard synchrocyclotron indicated the presence of a new activity of approximately two-hour half-life. In order to identify and characterize this activity, niobium metal foils were irradiated for periods of 45–60 minutes with protons of maximum energy varying from ~50 Mev (roughly the threshold for the reaction) to ~75 Mev, and zirconium metal foils were bombarded for similar periods with protons of 40–45 Mev maximum energy.

After irradiation, the niobium target foils were dissolved in a mixture of nitric and hydrofluoric acids containing molybdenum, zirconium, and yttrium carriers. Zirconium was then separated from the solution as barium fluozirconate by the addition of barium ion, and this precipitation was repeated once or twice more. Upon dilution with water, yttrium fluoride also precipitated from the supernatant. After centrifugation, the niobium was precipitated with ammonia, washed, and dissolved in concentrated hydrochloric acid. This solution was made approximately $3M$ in HCl by first saturating with HCl gas and then diluting with the appropriate amount of water. Two extractions with tributyl phosphate removed the molybdenum, and the aqueous phase was again saturated with HCl gas. Niobium was then separated from residual zirconium and yttrium by extraction from the HCl solution with diisopropyl ketone. The zirconium target foils were treated in a somewhat similar manner except that they were dissolved in hydrofluoric acid alone, the barium fluozirconate precipitation was done at least three times, and there was no necessity for the molybdenum extraction with tributyl phosphate.

Samples of the niobium fractions so obtained were followed on an end-window (3.5 mg/cm^2) Geiger counter, and resolution of the decay curves indicated a two-hour activity plus the known 14.7-hour Nb^{90} and longer activities. An aluminum absorption curve taken immediately after the chemical separation showed that the major particulate radiation seemed to consist of positrons of 2.9±0.4-Mev maximum energy. Counting samples of the niobium fraction through 620 or 571 mg/cm^2 of aluminum cut down the fraction of interfering activities (mainly Nb^{90}) and showed that these high energy positrons had a 1.9±0.3-hour decay. There was also a suggestion of a small amount of a shorter-lived activity. The decay of samples followed with a scintillation counter employing a well-type NaI(Tl) crystal gave a similar value for the half-life and indicated more strongly the possible presence of an additional gamma activity of just under one-hour half-life. An example of the decay curves obtained is shown in Fig. 1. Crude gamma integral pulse-height curves taken with the scintillation counter indicated a greater percentage of annihilation radiation associated

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