Neutron Capture Gamma-Ray Spectra*

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Photographic plates soaked in D_2O may be used for gamma-ray studies. By measuring the ranges of the photo-protons resulting from the photo-disintegration of the deuterons in the D_2O, the γ-ray energies can be found. The method is limited to γ-rays of energy above 3 Mev. The neutron capture γ-ray spectra of Al, Cl, Fe, Br, Cd, La, W, Au, and Hg have been obtained by this method. The results suggest a possible dependence of the shape of the spectra on the odd-even character of the compound nucleus.

I. INTRODUCTION

ALTHOUGH a large amount of research has been done on the problem of radiative capture cross sections for thermal neutrons, very little attention has been given to the study of the instantaneous gamma-rays emitted. Most of the studies[1] of these capture gamma-rays have been made by studying their absorption in some material for the purpose of finding either an average gamma-ray energy or a maximum energy. Since such spectra are usually complex, the absorption technique cannot be expected to yield very accurate average energies. As for the maximum energy determinations, here one is faced with finding the end point of a spectrum and only in those researches where neutron beams from a chain reacting pile have been used has the intensity been high enough for reasonably accurate measurements.

In all of the above-mentioned work no direct study of the spectra was made. Recently, studies made with a γ-ray pair spectrometer[2] have been reported. A pair spectrometer should be capable of giving good results at the high energy end of the γ-ray spectra. In the work of Kinsey *et al.*, the energy values reported do not go below 4.5 Mev.

The researches described in this paper were carried out for the purpose of obtaining some detailed knowledge about the instantaneous γ-ray spectra of various elements resulting from neutron capture. It was hoped that by studying various elements some information concerning the dependence of the shape of the spectrum on atomic number or nuclear type could be found.

II. EXPERIMENTAL METHOD

By introducing heavy water into a photographic emulsion[3] and then exposing the plate to γ-rays it is possible to obtain the energies of the γ-rays by measuring the ranges of the photo-protons resulting from the photo-disintegration of the deuterons in the D_2O. The energy of the γ-ray, E_γ, is related to the energy of the proton, E_p, by the relation

$$E_\gamma = 2E_p + 2.2 \text{ Mev}. \quad (1)$$

This formula neglects the effect on the energy of the angular dependence of the reaction. This effect will be a cause of poor resolution for the method unless one has sufficient γ-ray intensity to be able to collimate the gamma-ray beam. Furthermore, since it is difficult to measure accurately the range in an emulsion of a proton of energy very much below 0.5 Mev (6 microns), this method will be limited to γ-rays above 3 Mev.

Amounts of D_2O of at least 60 percent by weight can be introduced into an emulsion by soaking in D_2O. The emulsion need not be[3] stripped but may be left on a glass backing to simplify the handling problem.

Figure 1 shows the experimental arrangement used. A beam of neutrons from the thermal column of the Argonne heavy water moderated pile impinges on a calcium fluoride crystal. The crystal is rotated until a Bragg maximum is found at an angle of 30° to the incident neutron beam. In this manner one avoids placing the plates in the direct pile beam which contains many gamma-rays, which would cause a very large background of tracks and make the search for photo-proton tracks impossible.

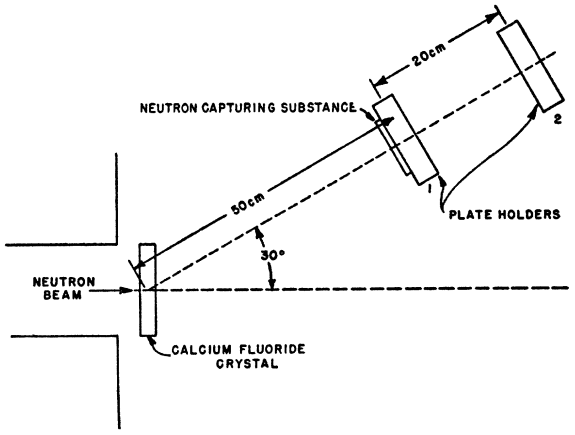

FIG. 1. Diagram of apparatus.

* A preliminary report of this work was presented at the Washington meeting of the American Physical Society, 1949; Phys. Rev. **76**, 182 (1949).

[1] H. E. Kubitschek and S. M. Dancoff, Phys. Rev. **76**, 531 (1949). This paper is the most recent one using the absorption method. It contains a list of references to the earlier work on capture gamma-rays.

[2] Kinsey, Bartholomew, and Walker, Phys. Rev. **77**, 723 (1950); **78**, 77 (1950); **79**, 218 (1950); Bartholomew, Kinsey, and Walker, Phys. Rev. **79**, 218 (1950).

[3] G. Goldhaber, Phys. Rev. **74**, 1725 (1948).

The Bragg reflected neutron beam then strikes the capturing material. Figure 1 shows the location of the plate holders and the (n, γ) source. The first plate (experimental plate) is right next to the gamma-ray source, whereas the second plate (background plate) is relatively very far from the gamma-ray source. On the other hand, the two plates are comparatively close together with respect to any source of background radiation such as fast neutrons from the pile.

The details of the plate holders are shown in Fig. 2. The holders are of $\frac{1}{8}''$ lead and are coated with a thick layer of boron carbide which has been painted on with Zapon. The holders are then covered with a thin sheet of aluminum foil to protect the boron carbide coating. The lead prevents the very low energy capture γ-rays from striking the plate. These would serve no useful purpose but would fog the plates. The boron carbide absorbs any thermal neutrons that may have penetrated the (n, γ) source and does not yield any capture gamma-rays that could photo-disintegrate deuterium. The plate holders are filled with D_2O. Ilford C_2 emulsions, 100 microns thick, have been used for all the elements that have been studied. The plates are immersed in the heavy water throughout the irradiation.

The thickness of the (n, γ) source has been chosen to be approximately one thermal neutron capture mean free path, provided this length is less than the mean free path for Compton scattering of a gamma-ray of 3 Mev. For small capture cross sections, the last criterion required using only a small fraction of a mean free path, in which case the searching of the plates was much more difficult.

The irradiations were of 30 to 50 hours duration. When longer irradiations were used, the background became too heavy to allow for easy searching of the plates.

III. DATA AND CORRECTIONS

After irradiation, the plates were processed and then searched with microscopes with oil immersion objectives. The projected lengths of the tracks were measured with calibrated eyepiece scales and the dips of the tracks were measured with the calibrated fine depth adjustments of the microscopes. The plates were each searched until 300 tracks were found, with the exception of the plates exposed to aluminum and lanthanum capture gamma-rays. The background plate corresponding to a given experimental plate was then searched over the same total area as had been covered on the experimental plate. The number of tracks found on the background plate averaged below 20 percent of the number found on an experimental plate.

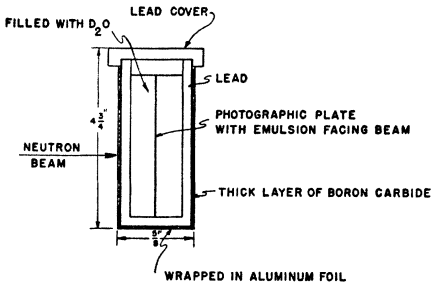

FIG. 2. Detail of plate holder.

The actual track lengths were calculated by correcting for emulsion shrinkage and for the dip of the track. In order to be able to use the Ilford range-energy[4] curves, a comparison of the range of protons in wet and dry emulsions was needed. Goldhaber[3] gives the range in dry emulsion as equal to 0.93 times the range in wet emulsion. This relation was used and then the energies of the photo-protons could be calculated. The γ-ray energy was then determined from Eq. (1). Figure 3 shows the range distribution of tracks from cadmium. The associated background tracks are shown on the same figure.

The range distribution must be corrected for the following: (1) the probability of tracks leaving the emulsion, (2) the variation with energy of the photo-disintegration cross section of the deuteron.

The first correction is purely a geometrical one. It may be calculated exactly in terms of emulsion thickness and track length. The second correction is more difficult to determine. The measurements[5] of the (γ, n) cross section of the deuteron have only been made at a few energies. The experiments are very difficult ones and the results are not very accurate. However, the theoretical calculations of the cross section should be reasonably good in the range of energies 3 to 9 Mev.

The values of $\sigma(\gamma, n)$ for the deuteron were obtained from a calculation based on the[6] simple theory of the effect, neglecting tensor forces. The calculations were extended to 5 Mev. Beyond 5 Mev, the formula for the photoelectric cross section was used, since the photomagnetic cross section is negligible at these energies. The recent measurements[7] at 6.13 and 17.6 Mev agree very well with the results of the above calculations.

Both corrections raise the number of high energy tracks relative to the number of low energy tracks. The corrected spectrum of cadmium is shown in Fig. 4.

Similar sets of histograms for the other substances studied were obtained. The background data for all the elements studied were then averaged and the distribution of the average background was determined. The histograms were then redrawn using the average background.

Finally, the data was smoothed by taking the average of the number of tracks in two adjacent blocks of the previously obtained histograms. A point corresponding to this average number of tracks was then plotted

[4] Lattes, Fowler, and Cuer, Proc. Phys. Soc. London **59**, 883 (1947).
[5] A. Wattenberg, *Photoneutron Sources*, Preliminary Report No. **6**, Nuclear Science Series, Division of Math. and Phys. Sciences, Nat. Res. Council.
[6] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 122 (1936).
[7] Barnes, Stafford, and Wilkinson, Nature **165**, 69 (1950).

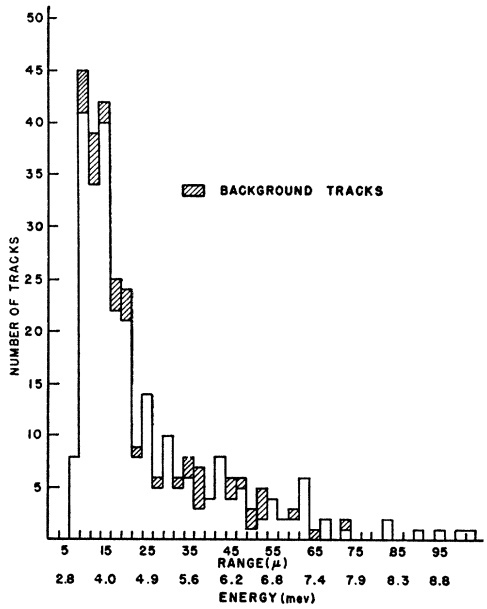

FIG. 3. Range distribution of tracks from cadmium.

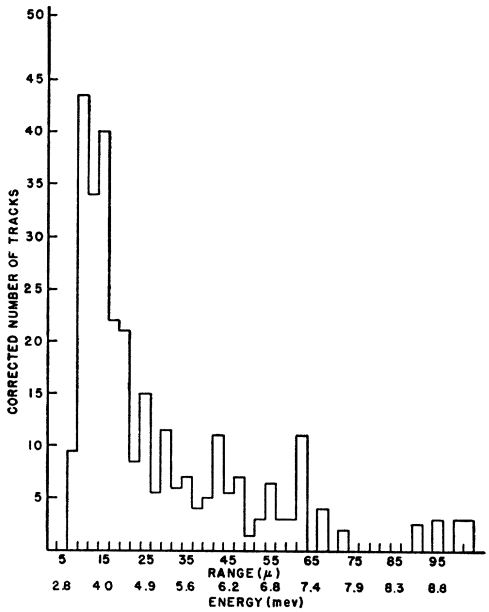

FIG. 4. Neutron capture γ-ray spectrum of cadmium.

against the average of the energy represented by the two corresponding blocks of the histograms. The points were then joined by straight lines. Figure 6 shows the smoothed spectrum of cadmium.

The energy resolution of the method is of the order of ± 0.25 Mev. Most of this spread arises from the dependence of the energy of the photo-proton on the angle between the incident γ-ray and the proton's direction of motion. The thermal flux available after Bragg reflection was too low to permit collimation of the γ-rays so as to try to minimize the angular effect on the resolution.

IV. RESULTS

Before the capture γ-ray spectra are examined, it should be recalled that there is a low energy cutoff for the detector at about 3 Mev. Furthermore, by the very nature of the photographic method it will be difficult to determine a high energy cut-off of a spectrum. In addition, the poor resolving power of the method will tend to mask evidence of line structure in the spectra.

The elements reported herewith are Al, Cl, Fe, Br, Cd, La, W, Au, and Hg. The solid element was used in all cases except for Cl, Br, and Hg. For these C_2Cl_6, NaBr, and HgO were used. These were finely powdered and were placed in thin-walled aluminum containers. The thermal neutron capture mean free path of aluminum is 83 cm so that a thin-walled container gives a negligible contribution to the γ-ray spectrum that is being studied. The thickness of material used was one thermal neutron capture mean free path for all substances with the exception of Al, La, and Cd. In Al, only $\frac{1}{5}$ of a mean free path was used, since there is no advantage in using a thickness greater than a Compton mean free path. (For Al this is about 20 cm for 3 Mev gamma-rays.) In La, only enough material was available for $\frac{1}{2}$ a mean free path. For Cd, a thickness of 4 mean free paths was used.

In those cases where compounds were used (C_2Cl_6, NaBr, and HgO) the companion element in the compound yielded a negligible contribution to the capture γ-ray spectrum. In all cases, with the exception of Al and La, 300 tracks were obtained. In the two exceptions, 150 and 200 tracks respectively were obtained.

Aluminum (*Fig. 5*). There seem to be two possible peaks in the spectrum at 5.3 and 6.3 Mev. The group at 7.6 Mev reported by[2] Kinsey *et al.* does not stand out. The high energy cutoff is between 8.0 and 8.5 Mev.

Chlorine (*Fig. 5*). The spectrum is relatively flat over a wide range of energies beginning at the low energy cutoff of the detector. The intensity falls to a low value near 8.0 Mev and then a very intense group appears at 8.5 Mev. This is probably a single line. This γ-ray has been used to photo-disintegrate[8] various substances. The high energy cutoff is at about 9.2 Mev.

Iron (*Fig. 5*). The intensity is a maximum at 5.2 Mev. The intensity is definitely decreasing on each side of this energy. There is evidence of a line at 6.0 Mev. The high energy cutoff is between 7.5 and 8.0 Mev.

Bromine (*Fig. 6*). The intensity is probably still rising as one goes to energies below the low energy cutoff of the detector. The intensity decreases sharply towards higher energies. The peaks at 5.3, 6.3, 7.3, and 8.0 may be evidence of line structure. The high energy cutoff is between 8.5 and 9.0 Mev.

Cadmium (*Fig. 6*). The intensity is probably still

[8] B. Hamermesh and A. Wattenberg, Phys. Rev. **76**, 1420 (1949).

rising below the low energy detector cut-off. The intensity falls steadily to the high energy cut-off between 7.5 and 8.0 Mev. There is no evidence of structure.

Lanthanum (*Fig. 6*). There is an intensity maximum at 4.6 Mev. The high energy cutoff is between 7.5 and 8.0 Mev.

Wolfram (*Fig. 7*). There is an intensity maximum at 4.6 Mev. The peaks at 4.6, 5.3, 6.0, and 6.6 Mev indicate a possible line structure. The high energy cutoff lies between 8.5 and 9.0 Mev.

Gold (*Fig. 7*). The spectrum is relatively flat from 4.8 to 8.2 Mev. The intensity decreases on each side of this region. The intensity falls to a very low value at 8.8 Mev. A possible line at 9.2 Mev is in evidence.

Mercury (*Fig. 7*). The intensity is probably still rising below 3 Mev. The intensity decreases steadily to a high energy cutoff between 7.5 and 8.0 Mev.

V. DISCUSSION

From the meager theoretical knowledge at our disposal, one would think that for sufficiently heavy nuclei the predictions of a statistical theory of nuclear structure would apply. This theory predicts spectra of the form of the[9] cadmium spectrum. However, the types of spectra reported herewith may be placed in three general classes which can be described as follows:

(1) The intensity is probably still rising below 3 Mev. The intensity falls steadily to a high energy cut-off. There is no evidence of line structure. In this class we find Cd and Hg.

(2) The intensity is a maximum in the neighborhood of 5.0 Mev. The intensity decreases on each side of this maximum. There is evidence of line structure. In this class are Fe, Br, La, and W.

(3) The intensity is relatively constant over a wide range of energies. There is evidence of line structure. In this class are Al, Cl, and Au.

The strange thing about the results is that from the data on cadmium one might expect that all heavier elements would have a similar type of spectrum. Of those elements heavier than cadmium that are included in this report, only mercury has a cadmium-like spectrum. Furthermore, the three elements W, Au, and Hg, which are so close together in the periodic table and are so much heavier than cadmium, have very different spectra. In fact, in the above rough classification, they are in different classes.

If one considers the compound nuclei from which the γ-rays are emitted, a possible reason for the differences between W, Au, and Hg will appear. In the case of gold the capturing nucleus is Au^{197}. However, for the other elements there are several isotopes that need be considered. Fortunately, in each case there is one isotope which outweighs all others by virtue of its relatively very large capture cross section. These isotopes are W^{186} and Hg^{199}. The three compound nuclei which give rise to the capture gamma-spectra are of the types:

$$\text{Even } Z, \text{ odd } N - W_{74}{}^{187},$$
$$\text{Odd } Z, \text{ odd } N - Au_{79}{}^{198},$$
$$\text{Even } Z, \text{ even } N - Hg_{80}{}^{200}.$$

The differences in shapes of the capture γ-ray spectra could be due to the differences in the nuclear types of the compound nuclei of W, Au, and Hg. This is not an unreasonable assumption, since level spacings should vary considerably from one nuclear type to another. Recent work[10] on capture γ-ray multiplicities indicates such a dependence on nuclear type. Additional elements are now being studied to check the above suggestion.

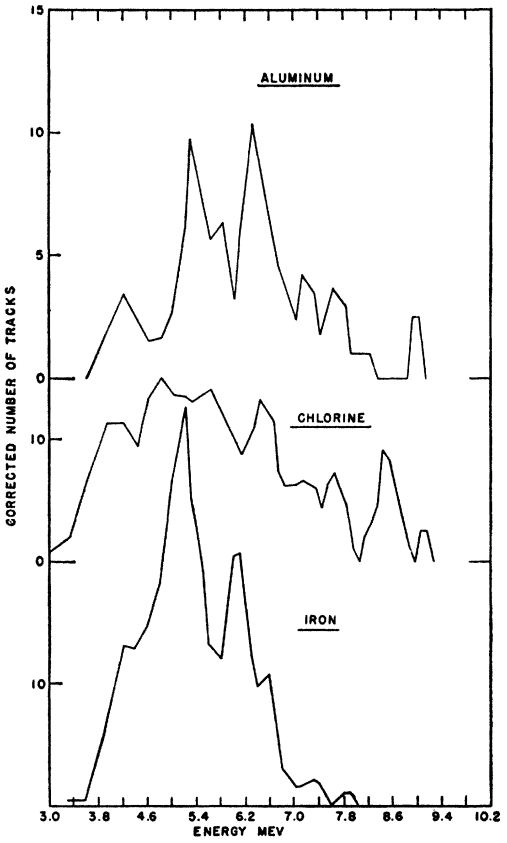

FIG. 5. Neutron capture gamma-ray spectra of aluminum, chlorine, and iron.

VI. CONCLUSION

The D_2O soaked plate technique of studying neutron capture gamma-ray spectra yields results which give a general picture of the shapes of such spectra. The poor resolving power of the method does not allow one to determine with any good accuracy line structure in the spectra. The results on the nine elements that are reported are rather surprising and indicate that a theory of the spectra based on a statistical nuclear model is not valid. The results suggest a dependence of

[9] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **9**, 231 (1937).

[10] C. O. Muehlhause, Phys. Rev. **79**, 219 (1950).

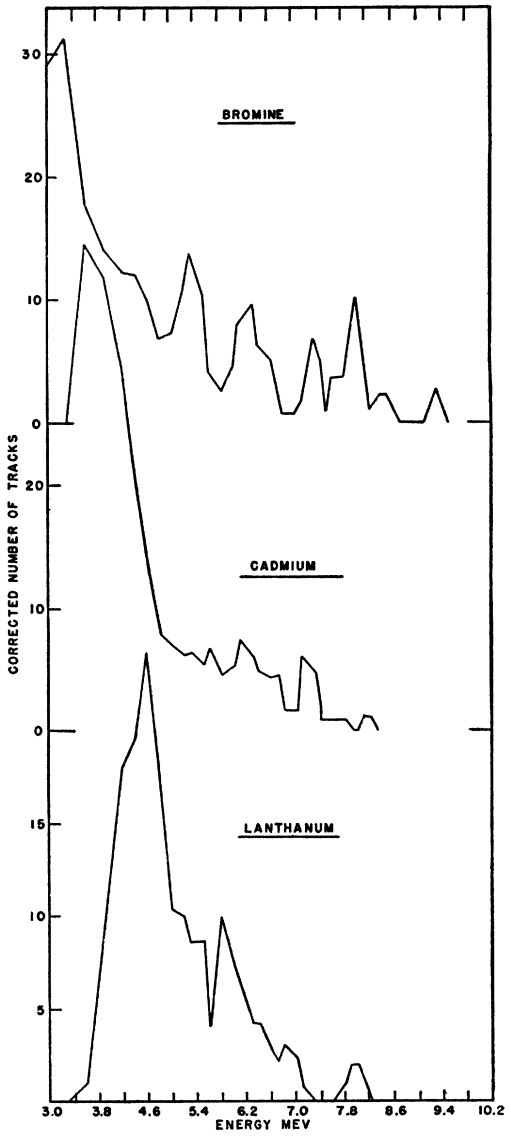

FIG. 6. Neutron capture gamma-ray spectra of bromine, cadmium, and lanthanum.

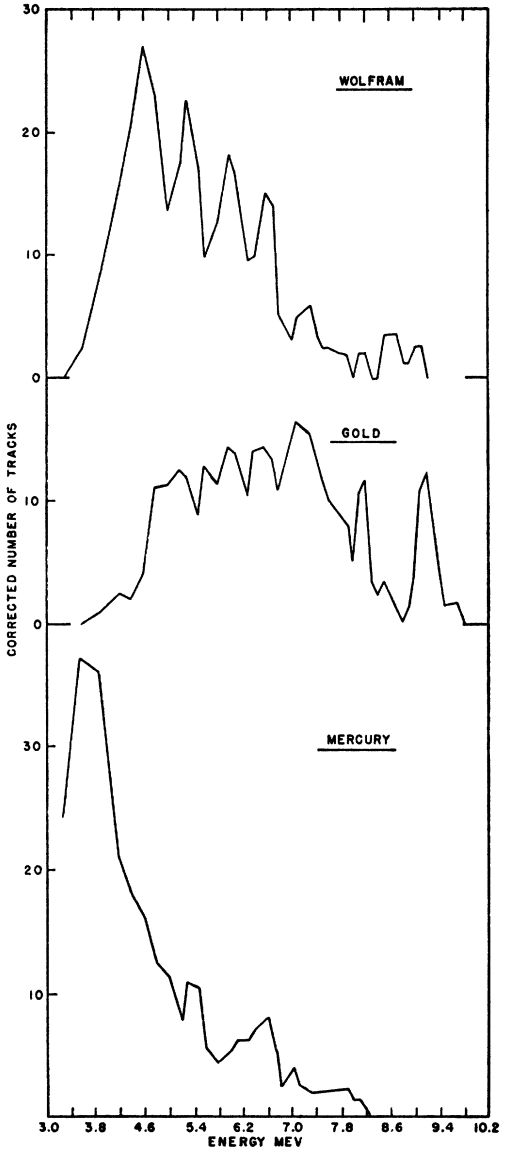

FIG. 7. Neutron capture gamma-ray spectra of wolfram, gold, and mercury.

the spectral shapes on the odd-even character of the compound nuclei which emit the spectra.

It is a pleasure to acknowledge the advice and assistance of the many people who have aided in the above research. Most of the very tedious plate searching was done by Louise Kollman. Without her help the work would have progressed at a much slower pace. John Dalman constructed the plate holders and assisted in setting up the apparatus. Drs. Albert Wattenberg and Morton Hamermesh were of incalculable help in offering advice and encouragement in the many discussions that were held as the research progressed.