Excitation of $\gamma$-rays by Slow Neutrons.$^{(1)}$

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1. Introduction. In the course of experiments on the action of slow neutrons on various substances, we have noticed a marked increase in the number of kicks of the Geiger counter placed near the neutron source when it is surrounded by a block of paraffin. This is probably the similar effect as that observed by Lea$^{(2)}$ and Fleischmann$^{(3)}$. On substituting the Geiger counter with a counter of Wynn-Williams' type we could observe no increase in the number of kicks. Hence, the increase in number of kicks observed with the former counter may mainly be due to $\gamma$-rays. We can consider two possibilities as to the origin of these $\gamma$-rays. In the first place the protons in paraffin itself emit $\gamma$-rays as the result of combining with neutron to form deuteron, as Lea$^{(2)}$ and Fleischmann$^{(2)}$ considered on interpreting the results of their experiments. This effect is the inverse process of the famous photo-dissociation of the deuteron observed by Chadwick and Goldhaber. But this explanation is not straightforward as we now know about the intense action of slow neutrons in various nuclear transmutation processes. The second and more natural explanation is thus offered by the secondary action of the slow neutron produced in the paraffin block. The slow neutron escaping the block may be captured by some of the surrounding substances emitting $\gamma$-rays as is evidenced by the experiment of Fermi and his collaborators$^{(4)}$. Since our apparatus is constituted to a greater part from iron, and the counter was surrounded by sheets of Pb to protect it from X-rays, the action of slow neutron on these elements must especially be taken into account.

The results of our experiments indicate in favour of the second alternative and show, moreover, that the $\gamma$-ray emission does not necessarily occur in concomitance with the production of artificial radio-element,

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(3) R. Fleischmann: Naturw. 50, 839 (1934).
thus pointing out the possibility of the transmutation of a stable nucleus into another stable nucleus with mass number increased by unity.

2. Experimental method. Neutron was obtained by deuteron-deuteron collision in the apparatus described before\(^{(1)}\). The accelerating potential was about 300 k.v. and some ten microamperes was received by the target. As $\gamma$-ray detector served an Al-walled Geiger counter filled with argon. It was accidentally of a good choice, that we have used aluminium, as we could confirm afterwards that Al emits no detectable $\gamma$-rays in the presence of slow neutrons. As to the artificial $\beta$-activity of Al we infer from the result of a trial experiment that its intensity is so weak that it needs not to be taken into account in our present research. In order to eliminate the fluctuation of the intensity of the neutron source due to the variation of discharge conditions we have employed a second similarly constructed Geiger counter as standard, which was always used under the same condition to indicate the intensity of primary neutrons. The two counters were placed diametrically about the target and their distance was long enough that we could neglect the influence of the $\gamma$-rays emitted from the substance placed before the first counter upon the standard counter. The natural effect of the counters lies between 5 to 15 countings per min.

It was a pity that the ratio of the countings of the two counters under certain definite conditions was not absolutely constant so that the values obtained in different occasions were not directly comparable with each other. But within a day or two the ratio was confirmed to be constant, and our conclusions on the relative values of the effect for various conditions were fairly accurate.

The effect of X-rays excited in the discharge tube was tested by a controlling experiment using proton beams instead of deuterons. It was confirmed that a Pb sheet of 1 cm. in thickness was sufficient to protect X-rays completely from the counter while those of 1 cm. in thickness permit about one stray kicks per min. per micro ampere with the accelerating potential of 300 k.v.

The impulses of the counters, after passing through a two staged amplifier, excite thyratrons, which in their turn operate mechanical recorders. The maximum separation of kicks by our present recording system amounts to 0.04 sec., hence by greater intensity of the radiation it was often necessary to correct the recorded countings for the finite

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separation.

Now, when the discharge tube system is set in to operate, we get about hundred kicks per min., which effect we shall refer to as "back ground." The number of kicks is increased up to about 300 per min. if the target is surrounded by a paraffin block of cylindrical form 20 cm. high and 20 cm. in diameter.

3. Controlling experiment (a). Inactivity of lead. Since there was a strong suspicion that sheets of lead, the employment of which in our experimental arrangement was inevitable for the purpose of protecting the counter from X-rays, may emit $\gamma$-rays in the presence of the slow neutron, this element has been repeatedly investigated. For this purpose we have employed a large shielding box of lead and compared the number of kicks of the counter placed inside the box in following two cases; (1) the lead box was covered with layers of boric acid (at least 2 cm. in thickness), so that no slow neutron can reach to the lead walls, (2) the layers of boric acid were removed and the lead box was exposed to slow neutrons. In this second case layers of boric acid of same thickness as in the first case was placed inside the shielding box to eliminate the effect of absorption of the back ground by boric acid or the emission of $\gamma$-rays emitted by proton contained in boric acid, if such an effect is really existing. We could observe no difference in the number of kicks in two cases and have been led to the conclusion that the layers of lead sheets used for X-ray shielding have no share in the observed $\gamma$-rays of unknown origin.

4. Controlling experiment. (b) Absorption of $\gamma$-rays of unknown origin. The absorption by lead of the $\gamma$-rays of unknown origin was studied by inserting sheets of lead between the counter and the target. We can not draw any definite conclusion from the absorption measurement of this kind. For the source of the radiation was not concentrated in the neighbourhood of the target alone but distributed widely over different parts of the apparatus. This last circumstance was confirmed by comparing the number of kicks of the counter at various positions relative to the apparatus.

It may, however, be of some significance to report the results, since we know until now very little about these $\gamma$-rays. Fig. 1 (a) gives the absorption curve when the target was covered with paraffin, and (b) the same for the back ground. These curves are plotted in arbitrary scale and direct comparison of the height of the two curves has no meaning, since the counters were placed asymmetrically to the apparatus, and
increases in number of kicks of two counters took place by different amount, when the target was covered with paraffin blocks.

As will be seen from Fig. 1 the curves are quite similar in two cases showing that there is no essential difference between the nature of the rays. It is therefore probable that, owing to the presence of waters in cooling systems and of wood works for supporting purpose, slow neutrons were already existing before we covered the target with blocks of paraffin, and that the back ground is due to $\gamma$-rays excited by these slow neutrons just in the same way as in the case with blocks of paraffin around the target.

We do not know whether the flattening of the head of the curves is real one or not. The absorption coefficient derived from the middle variable portion of the curve (not corrected for the divergency of the radiation) is 0.6 cm$^{-1}$.

6. Experimental arrangements to detect $\gamma$-rays. After preceding preliminary researches we have adopted the following arrangement to detect $\gamma$-rays emitted from various substances under the bombardment of slow neutrons (Fig. 2). The counter, shielded by lead from X-rays, is put in a large box with walls of boric acid (at least 2 cm. thick) leaving the space A for a block of paraffin to produce slow neutron and the space B for the radiator.

The walls of boric acid, not only shut out the external slow neutrons, but also prevent internal slow neutron produced in A from escaping the box and exciting $\gamma$-rays in substances outside the shielding case.

If the substance under examination emit $\gamma$-rays, the number of kicks must be larger in the case, where the substance is in B and a block of paraffin is in A, compared with the case where the substance
is removed from B, while the block of paraffin remains in A. Whether
the \(\gamma\)-ray emission is due to the slow neutron or not can be settled by
repeating the measurement without the block of paraffin in A. If the
increase in number of kicks takes place only in the former case, then
the effect is due to the presence of slow neutron.

The presence of the back ground radiation makes the effect to be
observed indefinite. For, as the substance, when inserted in B, absorbs
the back ground, the observed increase in number of kicks becomes an
algebraic sum of the increase due to \(\gamma\)-rays and the decrease due to ab-
sorption mentioned above.

In order to reduce the intensity of back ground as much as possible
we covered all the faces of the box by lead sheets of few centimeter in
thickness. In our present apparatus, however, the layers of lead was
not thick enough so that the results obtained need further refinements.

7. **Results of the experiment.** We examined following elements:
Pb, Hg, Au, Cd, Cu, Ni, Fe and Al. Among these elements the experi-
ment of Fermi and his collaborators\(^{(1)}\) already showed Cd, Hg, Au and Cu
to emit \(\gamma\)-rays, which is also confirmed by our experiment. As to the
other elements, Ni and Fe emitted appreciable amount of \(\gamma\)-rays when
bombarded by slow neutrons, while on the other hand Al and Pb did
hardly show the same property (at least not to such an extent as
the others did). The results are tabulated in Table I. In the 3rd and
4th columns the percentage increase in number of kicks, when the

<table>
<thead>
<tr>
<th>Element</th>
<th>Form and size of radiators</th>
<th>% increase with paraff.</th>
<th>% increase without paraff.</th>
<th>relative cross-sec.</th>
<th>Abs-scattering cross-sec. (Dunning)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>10\times15\times1.5 cm.</td>
<td>-39</td>
<td>-</td>
<td>-</td>
<td>61</td>
</tr>
<tr>
<td>Hg</td>
<td>1000 gr. in two bottles</td>
<td>+10</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Au</td>
<td>a disc of 8 cm. in dia. 300 gr.</td>
<td>+56</td>
<td>+38</td>
<td>32</td>
<td>-</td>
</tr>
<tr>
<td>Cd</td>
<td>10\times15\times0.06 cm.</td>
<td>+143</td>
<td>-3</td>
<td>&gt;300</td>
<td>very large</td>
</tr>
<tr>
<td>Cu</td>
<td>10\times15\times2.0 cm.</td>
<td>+44</td>
<td>+2</td>
<td>14</td>
<td>59</td>
</tr>
<tr>
<td>Ni</td>
<td>10\times15\times2.1 cm.</td>
<td>+59</td>
<td>+1</td>
<td>16</td>
<td>-</td>
</tr>
<tr>
<td>Fe</td>
<td>10\times15\times2.4 cm.</td>
<td>+33</td>
<td>+2</td>
<td>10</td>
<td>78</td>
</tr>
<tr>
<td>Al</td>
<td>10\times15\times4.0 cm.</td>
<td>0</td>
<td>-2</td>
<td>&lt;0.12</td>
<td>19</td>
</tr>
</tbody>
</table>

\(^{(1)}\) loc. cit.
substance was introduced in space B, once with a block of paraffin $10 \times 15 \times 3.5 \text{ cm}^3$ in size in space A and once without it, are given. We see that in cases of Cd, Cu, Ni, and Fe the increase in number was observed only when there was a block of paraffin in A, while in the case of Au appreciable increase was observed even without a block of paraffin. It is therefore certain that Au emits $\gamma$-rays under the bombardment of fast neutron too, while it is difficult to say anything about Cd and the other elements since, as already mentioned, the absorption of background radiation may compensate the effect of $\alpha$-rays emitted. In fact, we see from the table that in the case of Pb the number of kicks decreases by appreciable amount when a sheet of Pb $1.5 \text{ cm}$ in thickness was placed in B, probably being due to the absorption of background radiation. Hence, if Cd, Cu, Ni, Fe and Al emit no $\gamma$-rays under the bombardment of fast neutron, the figures given in the fourth column of table I must be negative for these elements, owing to the absorption of back ground. As the absorption coefficients of background radiation in these elements are not yet investigated, we leave these problems to our further investigation with improved arrangement.

8. Yield of $\gamma$-rays. To calculate the relative cross-section for the emission of $\gamma$-rays given in the 5th column of table I from the data obtained, we neglected the absorption of emitted $\gamma$-rays in the radiator itself and also the absorption of background radiations, and assumed that the percentage increase given in the 3rd. column is entirely due to slow neutrons. We notice that, except Pb, there is a parallelism between the absorption-scattering cross-section of slow neutron given by Dunning and others\(^{(1)}\) and that for exciting $\gamma$-rays obtained by us. To estimate the absolute value of the cross-section, we must know the number of slow neutrons created in a block of paraffin placed in A per unit time. Now, from the number of kicks of counter of Wynnwilliams' type number of neutrons emitted from the target per sec. is estimated to be of the order of $10^6$. The propriety of this estimation is supported by the intensity of radio-activity induced in Ag placed near the target. On the other hand, we know from the results of our previous work\(^{(2)}\), about one to tenth of the neutrons are converted into slow neutron on passing through a layer of paraffin of $3.5 \text{ cm}$ in thickness. The efficiency of the counter for $\gamma$-rays was estimated from

\(^{(2)}\) loc. cit.
the number of kicks observed when known amount of Ra and its products were placed at a certain distance from the counter. We obtained the results that about one in 500 quanta of γ-rays passing through the counter is registered.

From these values and the geometrical configuration of the apparatus we estimated the atomic cross-section of slow neutron for exciting γ-rays in Fe to be $5 \times 10^{-24}$ cm$^2$. This value may involve the error by a factor 2 or 3. The cross-section in other elements can be calculated easily by multiplying this value by the relative cross-section given in the 5th column of table I.

9. Nuclear reactions. It is interesting to note that Fe and Ni, which emit γ-rays, are generally accepted to show no Fermi proton effect, in particular Ni produces no radio-active nucleus under the action of neutron bombardment. If the observed γ-ray emission accompanies the radiative capture process of slow neutron by these elements, product nuclei must be either of very short life or of very long mean life (stable or metastable isotopes). The interpretation in terms of this second possibility is supported by the recent discovery of Aston$^{(1)}$ of the isotopes of Fe and Ni, i.e. Ni$^{63}$, Ni$^{62}$ and Fe$^{57}$, besides the already known Ni$^{58}$, Ni$^{60}$, Fe$^{54}$, Fe$^{56}$. We may be justified in assuming following reactions to take place:

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\text{Fe}^{56} + n^1 \rightarrow \text{Fe}^{57} + h\nu,
\]
\[
\text{Ni}^{63} + n^1 \rightarrow \text{Ni}^{64} + h\nu.
\]

In regard to Cu, we know that this element shows Fermi proton effect. Although this may lead us to consider that γ-rays observed by us might be emitted by the radiative capture process in concomitance with the production of radio-copper already known, this consideration seems to contradict with the fact that no detectable γ-rays were emitted from Al, as we observed in our previous experiment that the saturated intensity of activity induced in Al and Cu were nearly equal. Now, Cu has isotopes 63 and 65 and there is a general rule that the element of odd atomic number has never stable isotopes more than two. It is therefore possible that γ-rays emitted from Cu are not due to ordinary radiative capture process.

10. Combination of slow neutron with Proton. We have further investigated to see if slow neutron can combine with proton to form

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deuteron under the emission of $\gamma$-rays. By filling the space B with boric acid we protected the counter from slow neutrons which were produced inside the shielding box by introducing a block of paraffin in A. We observed no detectable increase in number of kicks when a block of paraffin of $10 \times 15 \times 3.5 \text{cm}^3$ in size was introduced in A. From this result we estimated the cross-section for such a process to be smaller than $0.5 \times 10^{-24} \text{cm}^2$.

II. Summary. Emission of $\gamma$-rays under the bombardment of slow neutrons was investigated for following elements: Pb, Hg, Au, Cd, Cu, Ni, Fe and Al. It was confirmed, except Pb and Al, all these elements emit $\gamma$-rays. The cross-section of slow neutron for exciting $\gamma$-rays in Fe, Ni and Cu was estimated to be of the order of $5 \times 10^{-24} \text{cm}^2$, while that in Cd is much larger than this value. From the fact that we could detect no $\gamma$-rays emitted from a block of paraffin $10 \times 15 \times 3.5 \text{cm}^3$ under the bombardment of neutron, the cross-section for the combination of slow, as well as fast, neutron was estimated to be smaller than $0.5 \times 10^{-24} \text{cm}^2$.

In conclusion we wish to express our thanks to Imperial Academy of Japan for the grant which enabled us to cover the cost of heavy water and also to Hattori Hôkô Kwai for the grant which was used in the expenditure involved in setting up the high tension apparatus.

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Note added in the proof. Recently the results of experiments of Lea on a related subject were published in the Proc. Roy. Soc. 150, 637 (1935). The method used by him introduces many ambiguous points in the interpretation of the results obtained. Especially, the effects of slow neutron are not separated from those of fast neutron. Following two points are noticed in connection with the results obtained by us.

1) $\gamma$-radiation from paraffin. As the walls of ionization chamber used by Lea were made of iron, which substance, after our confirmation, emits an appreciable amount of $\gamma$-rays when bombarded by slow neutron, at least a part of $\gamma$-rays observed by him, when a block of paraffin was used
as radiator, must be attributed to the interaction of slow neutron with the walls of the ionization chamber.

2) γ-rays from lead. In this respect our result contradicts with that of Lea. We could observe no γ-rays from lead bombarded by slow neutron as well as by fast neutron, while Lea has attributed the increase of ionization, when lead was used as scatterer, to γ-rays from lead excited by the bombardment of fast neutron.