# Determination of the Energy of Photo-Neutrons Liberated from Deuteron by Radium C Gamma-Rays*

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### Abstract

The author studied in detail the photo-neutrons liberated from deuterium and berilium by Ra C $\gamma$-rays, and especially determined, for deuterium, the thickness of paraffin wax which was necessary to slow them down to the energy of the resonance absorption of iodine. From this it was ascertained that 2.198 MEV. $\gamma$-rays really act in the disintegration of deuterium, and the initial energy of liberated neutrons was estimated to be between 0.001 and 0.0076 MEV., and hence the binding energy of deuteron was $2.189 \pm 0.007$ MEV. which gave the mass of neutron 1.00895. The relative atomic cross-section for photo-disintegration of deuterium against berilium was $\sigma_D : \sigma_{Be} = 1 : 13$.


## Introduction

The phenomena of photo-nuclear disintegration were first observed by Chadwick and Goldhaber(1) in the case of deuterium irradiated by the $\gamma$-rays of thorium C$''$ and radium C. The same effect was also found by a later experiment by Szillard and Chalmers(2) for berilium exposed to the $\gamma$-rays of radium C.

The phenomena, besides being of high interest in themselves, give us sometimes a superior method of studying atomic nuclei. One of the most useful applications of this effect is to determine the binding energy of the nucleus, and especially that of deuteron. For the latter purpose, most of the previous workers(3) measured the energy of photo-protons liberated from deuteron generally by using the $\gamma$-rays of thorium C$''$ of 2.6 MEV. energy.

In spite of its critical property, no experiment was made for the determination of photo-protons (or photo-neutrons) from deuterons irradiated by Ra C $\gamma$-rays, because the direct observation of the range of their tracks in the Wilson apparatus and of their ionizing power in the ionization chamber was almost impossible. Some workers(4) held the

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* Under the same title the author read a paper at the annual meeting of the Physico-Mathematical Society of Japan held at Kyoto Imperial University, April, 1939.

opinion that the weakly observed photo-disruption of deuteron by Ra C $\gamma$-rays was not actually effected by the rays of 2.198 MEV. energy but, in reality, by those of higher energies probably contained.

The present writer studied the phenoména for several months to determine whether deuteron be disintegrated or not, by Ra C $\gamma$-rays of 2.198 MEV. energy. Having ensured a positive answer, he made an estimation of the value of the energy of the photo-neutron, and of the binding energy of deuteron. The main part of this paper is composed of the study of the distribution of the number of count of neutrons liberated from deuteron by Ra C $\gamma$-rays as a function of the thickness of paraffin layers surrounding the neutron emitter, once in the presence and then in the absence of an iodine absorber which naturally determines the thickness of the paraffin layer sufficient to slow down the neutrons to the energy of the resonance absorption of iodine.

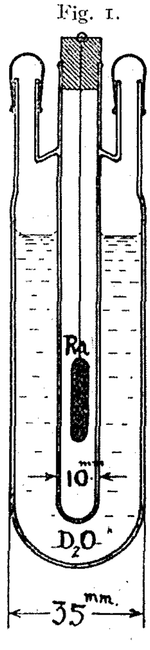
Some properities of photo-neutrons liberated from nucleus of berilum by the $\gamma$-rays of Ra C were investigated, and the ratio of the cross-section for disintegration of $Be^9$ to that of $D^2$ was estimated to be 13 : 1.

## Apparatus

*a) The source of photo-neutrons*

In order to obtain photo-neutrons from deuteron, 50 c.c. of heavy water of 99.5 per cent concentration was irradiated by gamma-rays from 50 mg of radium enclosed in a platinum container. The heavy water occupied the space between the double walls of a glass container (Fig. 1) constructed similarly to a cylindrical Dewer vessel. The radium was placed in the central cavity of the container, which was surrounded in turn by paraffin cylinders of several thicknesses from 4 mm. to 60 mm.

Fig. 1.

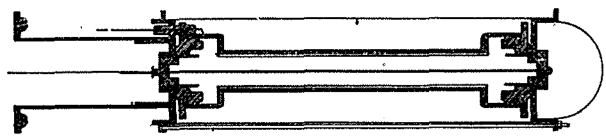



In the case of observing photo-neutrons from berilium, a thin walled (1 mm. thick) copper can was used for the berilium container which was filled with 20 grams of berilium powder and the $\gamma$-ray source of radium was placed in its central part.

*b) Neutron detector*

For detecting neutrons, there was used a cylindrical ionization chamber (Fig. 2), the inner surface (126 cm.$^2$) of which was coated with a thin layer of

Fig. 2.

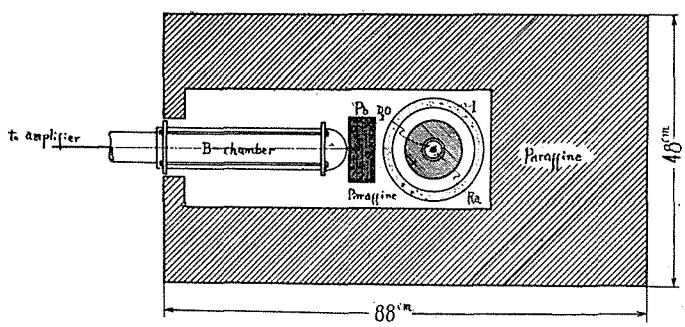


Boron chamber

borax, and the ion-collector was connected to the grid of the first valve of a system of linear amplifier and oscillograph or speaker.*,(5) In order to reduce the unsteady background due to the presence of $\gamma$-rays, a circular block of lead 7 cm. thick and ·12 cm. in diameter was interposed between the source and the ionization chamber.

To increase the efficiency of counting neutrons, the whole arrangement (Fig. 3) (consisting of radium source + heavy water or berilium, paraffin cylinder, iodine absorber, lead and ionization chamber) was enclosed in a thick paraffin box of the dimension indicated in Fig. 3.

Fig. 3.




With this arrangement it was found that, even though no paraffin cylinder was inter-placed, the number of counts due to photo-neutrons from deuteron + Ra C $\gamma$-rays was about 300 per hour, while otherwise the counts were too weak to be measured with sufficient accuracy by using such a weak $\gamma$-ray source as we had.

## The nature of photo-neutrons of deuterium and berilium

On counting, by means of the apparatus described in the preceding paragraph, the number of slowed down neutrons, it was found that the kicks appeared in two different groups by their amount and unless

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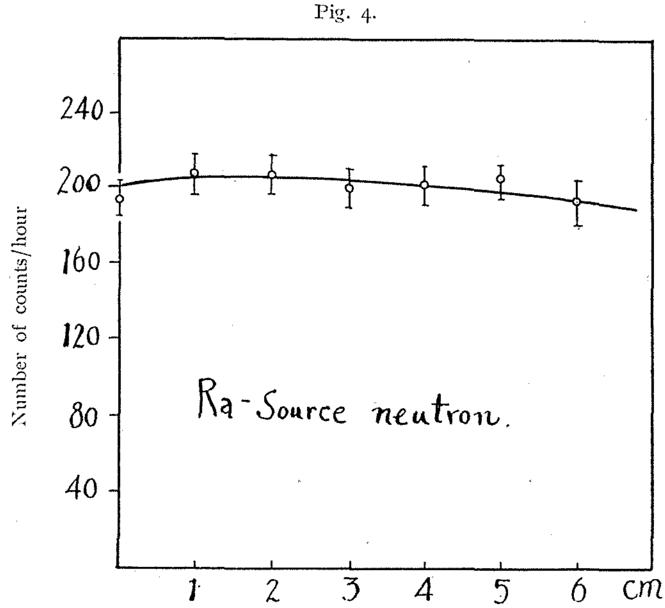
* The same apparatus had been previously used for the investigation of the neutrons liberated from lead exposed to cosmic rays.[5] The detail of the system of linear amplifier will be reported in this Memoir before long.

the counting system was carefully adjusted the group of smaller kicks was liable to be omitted from the count.

The natural background—the number of the count in the absence of radium—was about 10±2 per hour. With a source of radium of 50 mg alone or together with surrounding ordinary water, the number of counts was abonut 200±10 per hour. This indicated that a measurable number of neutrons was emitted from radium salt in a platinum container. When ordinary water was replaced by heavy water the counts increased to more than 300±15 per hour (without paraffin cylinder). We see, thus, the number of photo-neutrons liberated from $D_2O$ is almost the same order of magnitude as that of the "source neutrons" from radium in the container.

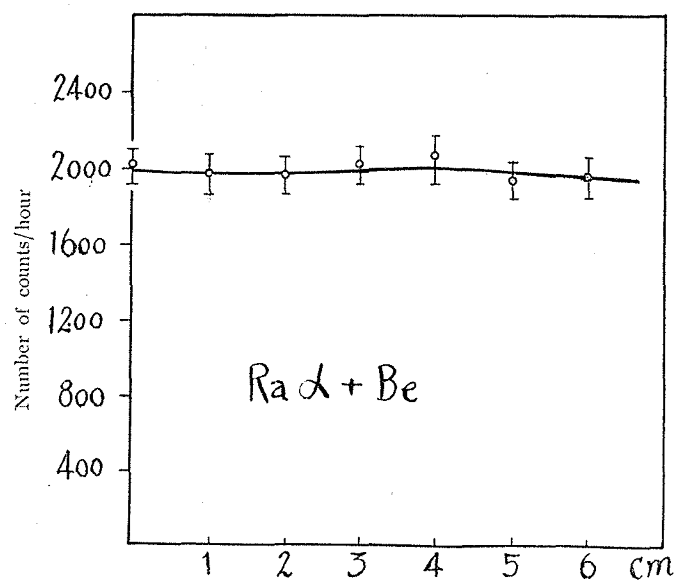
In order to know the nature of the "source neutrons," it was surrounded by a system of paraffin cylinders of various thicknesses respectively and the variation of the count of the neutrons was observed (Table I). The curve was found to be fairly flat at first up to 6 cm. of paraffin layer and then to decrease very slowly with increasing paraffin thicknesses (Fig. 4).

Fig. 4.




During the continued observation for several months a neutron source of 10 mg Ra $\alpha$+Be was used for control experiments, testing the efficiency of total counting system. By the way, the number of counts for various thickness of paraffin cylinder was taken and the result is tabulated in Table I and indicated in Fig. 5.

Fig. 5.




After the careful preliminary adjustment and observations the variation of counts of $D^2$-photo-neutrons was observed for various thicknesses (4~60 mm.) of paraffin cylinder surrounding the source

Table I.

| Paraffin mm. | Number of Counts per hour Raα+Be⁹ | Ra source |
|---|---|---|
| 0 | 2020 | 194 |
| 10 | 1980 | 204 |
| 20 | 1958 | 198 |
| 30 | 2016 | 203 |
| 40 | 2080 | 198 |
| 50 | 1936 | 204 |
| 60 | 1958 | 195 |

Table I. The number of counts of $Be^9+\alpha$ neutrons and "source neutrons" respectively. The statistical fluctuations are at most 5 per cent in each case.

placed in the paraffin box. The results are shown in Table II and Fig. 6, curve I. Since the counted number amounted to several thousands for each thickness of paraffin cylinder, the statistical fluctuation of each value is taken to be sufficiently small.

Table II.

| Paraffin mm. | Clear Number of Counts of $D^2$-photo-neutrons per hour Without Iodine | With Iodine 2.2 g/cm² | Difference |
|---|---|---|---|
| 0 | 109 | 110 | − 1 |
| 4 | 144 | 143 | + 1 |
| 7 | 170 | 168 | + 2 |
| 10 | 196 | 130 | + 66 |
| 13 | 200 | 98 | +102 |
| 17 | 172 | 90 | + 82 |
| 20 | 140 | 100 | + 40 |
| 30 | 106 | 96 | + 10 |
| 40 | 90 | 90 | 0 |
| 50 | 84 | 80 | + 4 |
| 60 | 74 | 80 | − 6 |

Table II. Clear number of counts of $D^2$-photo-neutrons per hour. The number both in the second and the third column respectively are the net amount subtracted by 200 per hour, which is the mean value of the counts of "source neutrons" vs. thickness of paraffin cylinder. The statistical fluctuations are at most 5 per cent in each case.

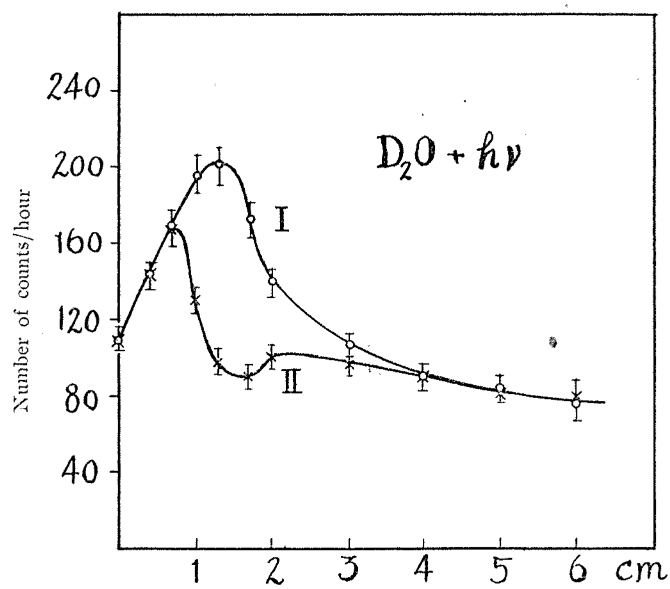
Table III.

| Paraffin mm. | Number of Counts per thirty minutes Be+$h\nu$ (Ra) |
|---|---|
| 0 | 176 |
| 4 | 204 |
| 7 | 220 |
| 10 | 248 |
| 13 | 257 |
| 17 | 250 |
| 20 | 268 |
| 30 | 275 |
| 40 | 260 |
| 50 | 186 |
| 60 | 146 |

Table III. Number of counts of $Be^9$-photo-neutron by Ra C $\gamma$-rays. The statistical fluctuations are 10∼5 per cent.

In the same way berilium photo-neutron was also observed and the results were summarised in Table III and plotted on the curve in Fig. 7. From the curves in Figs. 5, 6, and 7 respectively, we see that for $D^2+h\nu$ neutrons a relatively sharp maximum is found at 13 mm.

Fig. 6.

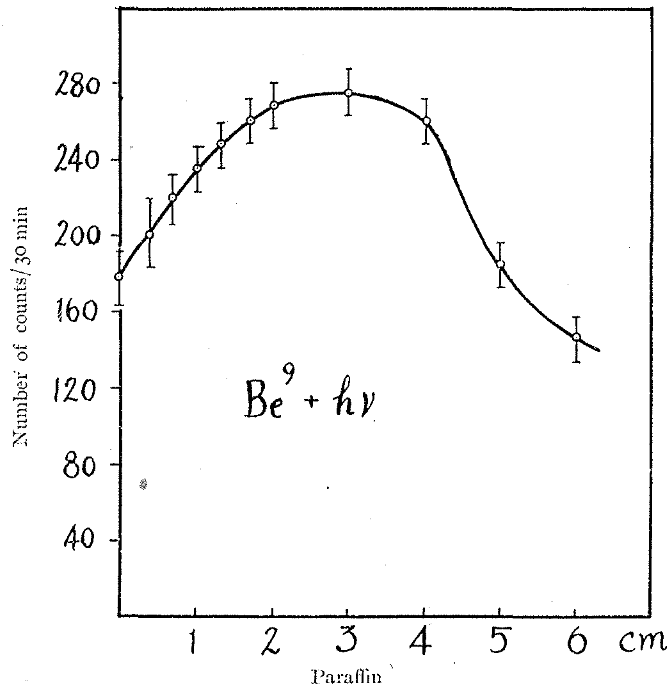



Paraffin

of surrounding paraffin layer, while the curve for $Be^9+\alpha$ is flat and no maximum can be found up to 60 mm. These differences of aspect are reasonable if we consider the effect of the degree of mixture of the neutrons of different energies as well as the difference of the initial values of their kinetic energies according as their disintegration mechanisms. We may conclude in this way that the neutrons from deuteron are "simple" or "monochromatic" while those of $Ra\alpha+Be^9$ neutrons and the "source neutrons" are "complex" or of "mixed colour." The neutrons from $Be^9+Ra\,C\gamma$ reaction may be taken as being composed of a few kinds of monochromatic neutrons which agrees with the expectation from the energies of acting $\gamma$-rays (2.198 and 1.76 MEV.) and the binding energy of the neutron in $Be^9$ (1.6 MEV.).

It is to be noticed that the positions of maximum intensity for both deuterium and berilium appear in the present experiment at a smaller thickness of the paraffin layer than do the observed values of Mitchell and others.[4] But, remembering that, in the present case,

Fig. 7.




the neutrons which emerged from the paraffin cylinder are again slowed down by the wall of the paraffin box, causes of the differences are recognised at once. It is actually observed that, in the neighbourhood of the maximum count of neutrons from deuterium + Ra C $\gamma$-rays, the thermal neutrons are not strongly decreased by placing Cd absorber (1 mm. thick) enclosing the paraffin cylinder.

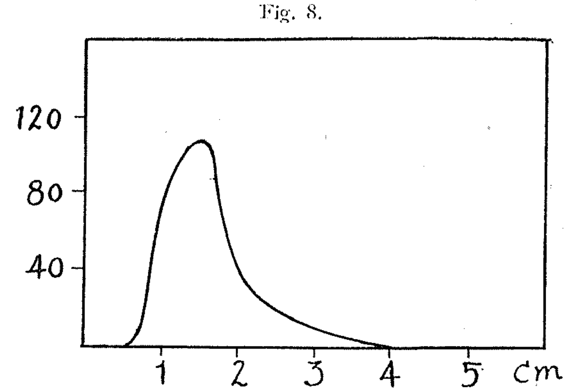
It may be possible to do a rough estimation of relative probability for photo-disintegration of $D^2$ and $Be^9$ by the gamma-rays of radium C. For this purpose the areas enclosed by the distribution curve, the parts of abscissa and ordinate were compared for the two different cases (Fig. 6, I and Fig. 7). Thus the area for $Be^9$ : the area for $D^2$ $= 37 : 6.5$, and the relative amount of $Be^9$ and $D^2$ used is 20 gr : 10 gr, hence the ratio of disintegration probability for each atom is 13 : 1. Though from the ratio of both maxima we have 7 : 1, the former method seems more reasonable than the latter. These relations agree fairly well with that of Chadwick and Goldhaber.(1)

## Absorption of Photo-Neutrons from $D^2$ by Iodine

In the course of the measurement of absorption of photo-neutrons from deuterium by various elements, the absorption by iodine was closely observed. In this case, the neutron source with the system of paraffin cylinders of varying thickness was surrounded by an iodine absorber (2.2 g/cm$^2$.), which was contained in a thin double-walled copper cylinder (15 cm. high, 17.5 cm. and 18.5 cm. in inner and outer diameter respectively), and all the other arrangements were exactly the same as previously (Fig. 3).

The distribution of the counts for various thickness of paraffin is as shown by the Table II and the curve II in Fig. 6. Comparing this with the curve I in Fig. 6, we find marked difference in distribution, namely, when the iodine absorber was inserted the number of counts increases at first with increasing thickness of paraffin cylinder in almost the same way as before, but then it soon ceases to increase and commences to decrease rather rapidly forming a slight groove in the neighbourhood of 17 mm.* If we take the differences of corresponding values on curve I and curve II respectively, we have a curve showing the variation of the absorption of iodine for the neutrons slowed down in different grade by passing layers of paraffin of different thicknesses after they were photo-electrically liberated (Fig. 8). We see that the absorption curve is fairly steep and has a sharply defined maximum at about 15 mm. and that a layer of paraffin of this thickness is necessary to slow down the initial photo-neutrons to the kinetic energy corresponding, in average, to the resonance energy of iodine.

Fig. 8.



Paraffin

## Energy of Neutrons Liberated from Deuteron

We are now to estimate the initial energy of neutrons liberated from deuteron by the knowledge obtained from the preceding experiment.

As the effective thickness of heavy water is 1.25 cm. and photoneutrons are liberated everywhere in it, we must take the slowing down

* In the case of the source neutrons no effect was observed by placing iodine absorber surrounding the paraffin cylinder.

effect in heavy water itself into account. By Dunning and others[6] the slowing down process of neutrons in $D_2O$ is $\frac{1}{5}$ of that of $H_2O$, while Kikuchi and others[7] observed no effect for slowing down in the former.

To measure the slowing down process in heavy water, it is more desirable to use iodine or bromine for the detector than to use Ag or Cd, because the probability of neutrons having thermal energy in heavy water will obviousely be small compared with that in ordinary water.

So if we want to determine the maximum thickness of paraffin layer to slow down the photo-neutrons to the energy of the iodine resonance level on the average, a few millimeters, at least, have to be added to the observed value, hence 15 mm + 2 mm = 17 mm.

According to Halban and Preiswerk[8] and Goldsmith and Rassetti[9] the value of the resonance of iodine is 80 ev., while Bethe[10] takes it to be 140 ev. by correcting the observed value with reasonably taken scattering coefficient. From the paper of Downing and Ellis[11] it appears as if iodine has two resonance levels, one in the neighbourhood of 40 volts and the other of several hundreds of volt. Hornbostel and Valente[12] took them as 30 ev. and 140 ev. respectively. In the present state of knowledge, it is, therefore, to be taken as 140 ev. for the upper limit of resonance energy. Thus we know that the initial photo-neutrons can be slowed down on the average to 140 ev. by passing through a layer of paraffin 17 mm. thick. So, if we know the mean free path of neutrons of "I" group, we can estimate the value or at least the conceivably highest limiting value of the initial energy of neutrons liberated from deuterium.

Experiments were undertaken by several workers[13] to ascertain the value of the mean free path of the neutrons in paraffin, and different values in wide region (11 mm.$\sim$5.6 mm.) were taken for the neutrons of the energies from 10,000 to 1 ev. By applying Fermi's general formula[14] Horvay[15] gave a table which permits us to estimate approximately the initial value of energy when the final value of energy and the mean square distance of travel in water were known.

Considering these findings, it seems, in the present experiment, that the photo-neutrons collide, in the course of the 17 mm. path in paraffin, with hydrogen atoms twice or at most four times on the average before they achieve 140 ev. So the highest, estimated value of the initial energy of photo-neutrons is about 0.0076 MEV., while the lowest is about 0.001 MEV.

## Conclusion

This experimental result shows that the photo-neutrons from deuterium by Ra C $\gamma$-rays are almost monochromatic and mainly affected by the $\gamma$-rays of $h\nu=2.198$ MEV.*

Then, since the initial value of the energy of photo-neutrons is determined to lie between 0.0076 MEV. and 0.001 MEV., the binding energy of deuteron falls between 2.183 and 2.196 MEV., i. e. 2.189 $\pm$0.007 MEV.

It is certainly difficult, in general, to make a precise determination of initial energy of neutrons by such a means as here taken, but, since the neutrons are found to be homogeneous and the estimated value is fairly small in energy, the derived value of the binding energy of deuteron in the present paper is considered to be one of the most accurate values.

Then the mass of free neutron may be calculated by the usual method, and by taking

$$D^2=2.0147$$
$$H^1=1.0081$$
$$Q_D=2.189 \text{ MEV.}=0.00235 \text{ in mass unit,}$$

we have ${}_0n^1=1.00895$.

For the convenience of comparing these values with various others previously determined, they are tabulated in Table IV.

Table IV.

| Binding energy of deuteron in MEV. | Mass of Neutron | Observer |
|---|---|---|
| 2.14±0.1 | 1.009 | Chadwick & Goldhaber (1935)(1) |
| 2.25±0.05 | 1.009 | Chadwick, Feather & Bretcher (1937)(3) |
| 2.17±0.04 | 1.00893 | Bethe (1938)(16) |
| 2.189±0.022 | 1.00895 | Stetter & Jenschke (1938)(3) |
| 2.18±0.07 | | Richardson & Emo (1938)(3) |
| 2.174 | | Roger Jr. & Marguerite Roger (1939)(3) |
| 2.189±0.007 | 1.00895 | Kimura (1939) |

In conclusion the author wishes to express his cordial thanks to Prof. B. Arakatsu for his many helpful suggestions and valuable advice

* No effect of harder $\gamma$-rays, as pointed out by Mitchell and others,(4) could be observed in the present experiment.

so generously given throughout the course of this work, and is also indebted to Mr. Y. Uemura for his help in constructing the apparatus. Lastly his thanks are due to Messrs. Ensuiko-Sugar Company for the loan of radium which rendered this work possible, and to the Nippon Gakujutsu Shinkôkwai for financial support through Prof. B. Arakatsu.

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