TITLE:

The (γ,n) Reaction of Molybdenum produced by the Li(p,γ) γ-Rays

AUTHOR(S):

Shimizu, Sakae; Yasumi, Shinjiro; Saji, Yoshio; Muto, Jiro

The (γ, n) Reaction of Molybdenum
produced by the Li (p, γ) γ-Rays

By

Sakae Shimizu, Shinjiro Yasumi, Yoshio Saji and Jiro Muto

(Received July 7, 1950)

ABSTRACT

The (γ, n) reaction of molybdenum produced by the irradiation of Li (p, γ) γ-rays was observed. The radioactivities induced in molybdenum samples were measured precisely by an end-window type β-ray counter with a thin mica window. While we determined the absolute number of γ-ray quanta by a specially constructed Geiger-Mueller counter with a thick lead wall, of which counting efficiency was computed theoretically. By measuring the intensities of activities produced in a set of molybdenum samples of various thickness and the number of γ-ray quanta concerned, we could determine the apparent cross section of this reaction for the sample with different thickness. Extrapolating the curve of apparent cross section *versus* the thickness of irradiated sample to zero thickness, we could determine the absolute cross section of the Mo^{92} (γ, n) Mo^{91} reaction for the Li (p, γ) γ-rays as 9.9×10^{-26} cm². Some discussions on the results obtained are also given.

1. Introduction

The (γ, n) type photo-disintegration of heavier nuclei can be induced only by the high energy quanta having the energy larger than neutron binding energy of a neutron in these nuclei. In recent years many works on thresholds and excitation functions of these reactions by the high energy X-rays from betatrons and synchrotrons were reported. Since, however, the continuous energy spectra of the X-rays used have been scarcely known, it is very difficult to determine the absolute cross sections for the photo-induced reactions by these experiments. And there are so far a few works concerning the experimental determination of the absolute cross sections with high energy γ-rays produced by the Li (p, γ) reaction (1).

One of the authors has recently reported the experimental determination of the cross section for Cu^{63} (γ, n) Cu^{62} reaction produced by these γ-rays (2). After this work, with the intention to find out any relation, if it exist, between cross sections and nuclear structures, we

have further continued the studies on (γ, n) reactions of heavier elements by the irradiation of Li (p, γ) γ-rays. The present report describes a similar work with molybdenum.

2. Apparatus

The apparatus and experimental arrangements were essentially the same as those of the previous work. But it is profitable to give some simple descriptions about the apparatus used.

For the production of Li (p, γ) γ-rays we used a high voltage machine of Cockcroft-Walton type equipped in our laboratory. The proton beam of about 460 Kev was directed on the target of metallic lithium placed on the bottom of a thin brass tube 0.2 mm thick. It was focused to a spot of about 1.5 mm in diameter.

The number of γ-ray quanta was counted by a Geiger-Mueller counter with a lead wall 6.5 mm thick, the inside diameter and the effective length of which are 2 cm and 2 cm respectively. The counting efficiencies of this lead counter for high energy quanta were recently computed theoretically by M. Sonoda (3) by taking the knowledges of the shower theory into account. Since the rigorous determination of the counting efficiency by accurate experiments is yet difficult, we used presumably these theoretical values. In actual arrangements the lead counter was placed at a fixed position 29.4 cm apart from the Li-target and usually shielded by a lead cylinder 1.05 cm thick. The practical absorption coefficient of this lead cylinder was measured as $\mu_\gamma = 0.53$ cm^{-1}. The absolute number of γ-ray quanta concerned was easily obtained from the data of net counts as described in the following section.

The samples irradiated were a set of molybdenum circular discs of various thicknesses. The diameter of all the discs was precisely equal to 22.8 mm, while the thickness was chosen as listed in Table I. The purity of molybdenum was examined spectroscopically and the density of samples was measured and was found to be $\rho = 9.67$ g/cm^3.

TABLE I.   Thickness of molybdenum samples.
(Diameter of each sample = 22.8 mm)

Sample No.	Thickness		
	in mm (δ)	in mg/cm^2 (w)	in mg/cm^2/$Z^{\frac{1}{4}}$
1	0.05	48.0$_5$	7.4$_1$
2	0.08	77.0$_3$	11.8$_9$
3	0.12	117.8$_5$	18.1$_8$
4	0.16	153.1$_7$	23.6$_3$

In the present experiment we used also copper activation monitors of 40 mm in diameter and 801.3 mg/cm² in thickness in order to avoid the effects due to the fluctuation of γ-ray irradiation, which may introduce some errors into the final results.

For the measurements of the positron activities induced in these samples we used two Geiger-Mueller counters of 2π-type (end-window type) with same dimensions except that the thickness of mica window of the sample counter is 5.2 mg/cm² while that of the monitor counter is 6.1 mg/cm². The details of these counters were described in the previous paper (2). The pulses of the counters, after amplification, were counted by a decade scaler and two scale-of-4 scalers.

3. Experimental procedures

Cross section

When the irradiation of γ-rays of constant intensity is made for one minute in the geometry shown in Fig. 1, the number of the active nuclei produced in the sample, N, is given by the following expression:

$$ N = \frac{\sigma m N_\gamma (1 - e^{-\lambda})}{\lambda} \left[\frac{1}{4} \int_a^{a+\delta} \log\left(\frac{x^2 + r^2}{x^2}\right) dx \right], \qquad (1) $$

where

σ : reaction cross section,

m : number of the parent Mo-nuclei per c.c. in molybdenum sample,

λ : decay constant of the active nuclei produced,

N_γ : total number of γ-ray quanta emitted from the Li-target for one minute,

a : distance between the γ-ray source and the upper surface of the sample disc,

δ : thickness of the sample disc,

r : radius of the sample disc.

Since, in actual experiments, γ-ray intensity fluctuated gradually while the sample was irradiated during about $20 \sim 50$ minutes, γ-ray counts were measured for each one minute, for which the intensity was assumed to be constant. The activities of the active Mo-nuclei produced were measured from just one minute after the end of irradiation for the following 15 minutes. Then the net counts, N_s, obtained by this procedure can be given, according to Eq. (1),
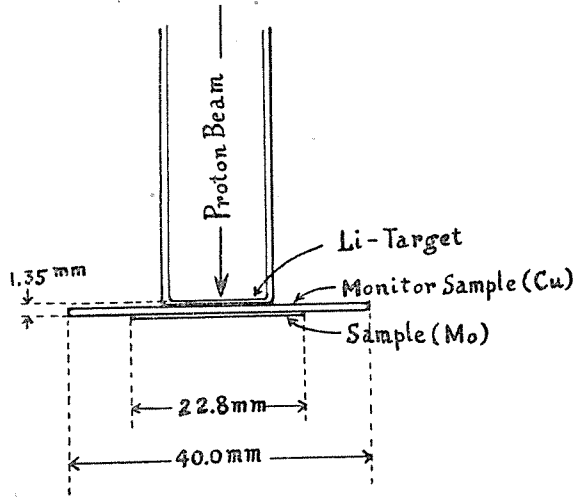
Fig. 1.   Standard geometry of irradiation of samples.

$$N_s = \frac{\sigma A_s m}{2 C_g C_\beta} \frac{1 - e^{-\lambda}}{\lambda} \left[\frac{1}{4} \int_a^{a+\delta} \log\left(\frac{x^2 + r^2}{x^2}\right) dx \right]$$
$$\times (1 - e^{-15\lambda}) \sum_{i=1}^{T} N_{\gamma_i} e^{-(T-i+1)\lambda}, \qquad (2)$$

where

T : time of irradiation in min.,

N_{γ_i}: absolute number of γ-ray quanta emitted from the Li-target during each one minute interval,

C_g : correction factor concerning the geometrical condition of activity measurements of the sample,

C_β : correction factor for the absorption of β-particles in a mica window of the β-ray counter,

A_s : factor concerning the self-absorption in the sample.

From Eq. (2) we obtain

$$A_s \sigma = \frac{2\lambda C_g C_\beta}{(1 - e^{-\lambda})(1 - e^{-15\lambda}) \sum_{i=1}^{T} N_{\gamma_i} e^{-(T-i+1)\lambda}}$$
$$\times \frac{N_s}{\frac{1}{4} \int_a^{a+\delta} \log\left(\frac{x^2 + r^2}{x^2}\right) dx} \frac{1}{m}. \qquad (3)$$

The value of $A_s\sigma$ is the apparent cross section for the sample of thickness δ. We prepared four kinds of molybdenum samples as listed in

Table I, and for these samples the values of $A_s\sigma$ were obtained. Now the true cross section means the value of $A_s\sigma$ when the sample thickness tends to zero. Hence we could easily obtain the true cross section, σ, by extrapolating the curve of $A_s\sigma$ *versus* w (weight of sample per unit area) to zero thickness $w = 0$.

Absolute number of γ-ray quanta

In order to obtain the absolute number of γ-ray quanta, N_{γ_i}, from the observed net counts of the γ-ray Geiger-Mueller counter used, n_{γ_i}, the following relation was used:

$$N_{\gamma_i} \frac{\omega}{4\pi} \varepsilon_{ff} e^{-\mu\gamma x} = n_{\gamma_i}, \qquad (4)$$

where ω is the solid angle subtended by the effective volume of the γ-ray counter at the γ-ray source, and $e^{-\mu\gamma x}$ is the factor for the absorption of γ-rays by the lead cylinder (thickness $x = 1.05$ cm) shielding the γ-ray counter. The efficiency of this counter used, ε_{ff}, was calculated theoretically by M. Sonoda (3), obtaining the result that $\varepsilon_{ff} = 0.23_8$ for 17.6 Mev γ-ray quanta. By numerical computation we obtained

$$n_{\gamma_i}/N_{\gamma_i} = 8.28_4 \times 10^{-5} . \qquad (5)$$

Correction factors concerning the β-ray measurements

The geometrical correction, C_g, was carefully determined by a series of experiments using Ag-sample activated by a neutron source (50 mg Ra + Be), and was found to be $C_g = 1.89$. In the case of Cu^{62}, as one of the authors previously reported (2), the correction factor C_β for the absorption of β-rays in a mica window of the sample counter was determined by inserting a set of thin mica sheets between the sample and the counter. Since the maximum energy of β-rays of the active molybdenum seemed to be about 2 Mev, as described in the following section, we could also in the present case adopt the same value as that in the case of Cu^{62}, namely $C_\beta = 1.06_6$.

Copper monitor

The irradiation was monitored by the measurements of Cu^{62} activities induced in the monitor sample of copper, mounted directly under the bottom of the Li-target together with Mo-samples in the geometry shown in Fig. 1. After the irradiation the similar measurements were made for the Cu-monitor as for the Mo-samples. For each run of measurements, the value

$$\frac{N_M}{\sum_{i=1}^{T} N_{\gamma_i} e^{-(T-i+1)\lambda_{Cu}}} \qquad (6)$$

was computed, where N_M is the total net counts of Cu-monitor for 15 minutes. We regarded the value of (6) as an index for the condition of γ-ray irradiations. In practice, however, throughout whole series of measurements data of only one run was excluded according to this point of view.

Stability of the β-ray counter

Stability of the β-ray counters used was daily examined by a standard KCl-sample placed at the fixed position in front of a mica window of the counter. The errors which may be introduced by small day-to-day variations in counter sensitivity were excluded by measuring the ratio of the net counts of KCl-sample in every day.

4. Results

Cross section

The apparent cross section for each sample is shown in Table II and Fig. 2. The observed value corresponding to the No. 1-sample could not be adopted, since the intensity of the β-activities induced in this sample was much weaker than the others. By the calculation using the least square method, we obtained the following empirical relation between the apparent cross section, $A_s\sigma$ (in cm^2), and the thickness of samples, w (in mg/cm^2):

$$A_s\sigma = (-0.0125_2 w + 15.7_6) \times 10^{-27}/\zeta , \qquad (7)$$

where ζ is the abandance ratio of the parent Mo-isotope concerned. Extrapolating w to zero, then A_s tends to 1 (cf. Fig .2). Hence, we obtained a value of the true cross section as

$$\sigma = (15.7_5 \times 10^{-27})/\zeta \text{ cm}^2 . \qquad (8)$$

Therefore we obtained the following results:

(a) If the observed reaction is assigned to $Mo^{92}(\gamma, n) Mo^{91}$, then
$$\sigma = 9.9 \times 10^{-26} \text{cm}^2 \text{ for Li}(p, \gamma) \text{ } \gamma\text{-rays,}$$
since ζ of Mo^{92} is 0.1586 (4).

(b) If the observed reaction is assigned to $Mo^{94}(\gamma, n) Mo^{93}$, then
$$\sigma = 17.3 \times 10^{-26} \text{cm}^2 \text{ for Li}(p, \gamma) \text{ } \gamma\text{-rays,}$$
since ζ of Mo^{94} is 0.0912 (4).

(9)

But, we can affirm that the reaction observed in the present experiment is assigned to $Mo^{92} (\gamma, n) Mo^{91}$, since the recent work of Duffield and Knight[5] indicated that the activity with a half-life of 15.5 minutes was observed by the use of molybdenum oxide enriched in Mo^{92} under the irradiation of 23 Mev betatron X-rays.

TABLE II. The apparent cross section for each sample.

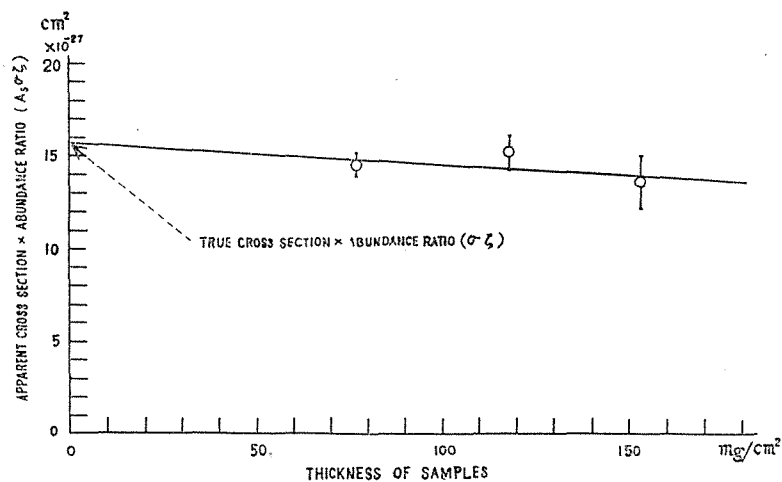| Sample No.	Thickness w in mg/cm^2	Apparent cross section × abundance ratio $A \sigma \zeta$ in cm^2
2	77.0_3	$14.3_9 \cdot 10^{-27}$
3	117.8_5	$15.1_5 \cdot 10^{-27}$
4	153.1_7	$13.3_7 \cdot 10^{-27}$

Fig. 2. Apparent cross section *versus* thickness of samples.

Half-life

In order to obtain half-life, T, or decay constant, λ, of the active Mo-nuclei, Mo-samples of 40 mm in diameter were prepared. Data obtained from the counting measurements of 12 separate runs are plotted in Fig. 3. From these data T and λ were determined by applying the rigorous method of Peierls[6] as follows:

Half-life $T = 14.2$ min., and decay constant $\lambda = 0.0487$ min^{-1}.

These values were used in the calculation of the cross section.

Transmission of β-rays

If we assume that the extrapolated value obtained by the empirical formula (7) corresponding to the transmission of β-rays is 100 per cent, the per cent transmission of the Mo-samples used is obtained as follows:

93.9 % for No. 2-sample (11.8_9 mg/cm^2/$Z^{\frac{1}{4}}$),

90.6 % for No. 3-sample (18.1_8 mg/cm^2/$Z^{\frac{1}{4}}$),

87.8 % for No. 4-sample (23.6_3 mg/cm^2/$Z^{\frac{1}{4}}$).
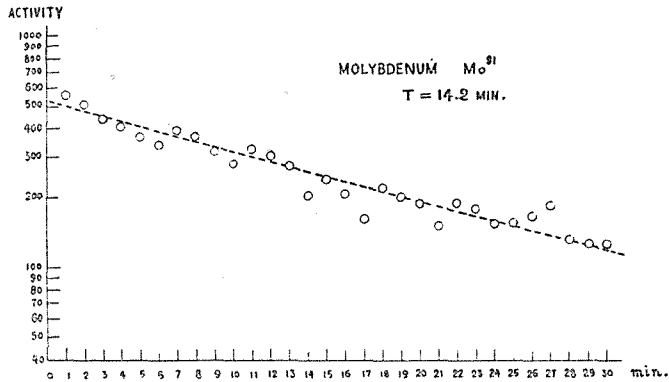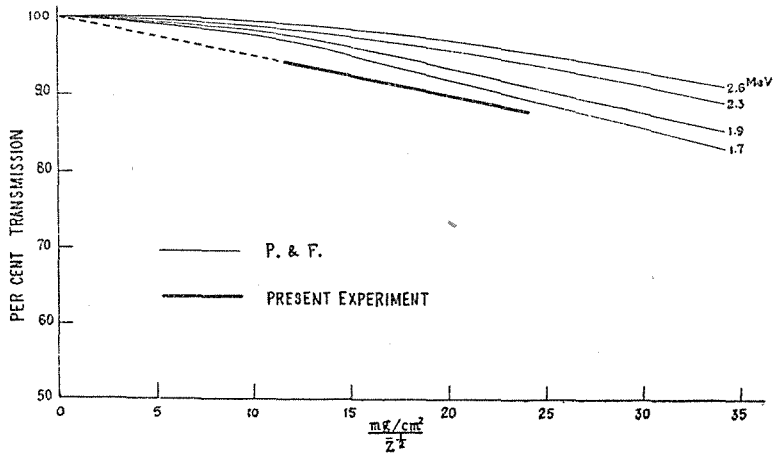

Fig. 3.   Decay curve of active molybdenum.

Fig. 4.   Self-absorption of β-rays of various energies (Mev).
Thin curves are quoted from the results obtained by Perlman and
Friedlander (Phy. Rev. **74** (1948), 442).   Thick curve is obtained by
the present experiment.

In Fig. 4 self-absorption curve—transmission *versus* mg/cm^2/Z$^{\frac{1}{2}}$—obtained by the present experiment is shown together with the curves given by Perlman and Friedlander (7). It seems probable, from Fig. 4, that the gradient of the curve obtained for β-rays of Mo-sample lies between those of 1.9 Mev and 2.3 Mev. For this reason it is concluded that the maximum energy of β-rays emitted from the active molybdenum may be about 2 Mev.

5. Discussions

From the preliminary experiments using a neutron source (50 mg Ra + Be) with and without paraffin, we could ascertain that the neutrons which might be emitted from the Li-target by the natural content of deuterons in the proton beam were incapable of causing any observable activities in the Mo-sample.

Since the decay measurements of active molybdenum were commenced from one minute after the end of irradiation, it could not be concluded whether much shorter half-lives* than that we observed exist or not. We observed 14.2-min. half-life, while other investigators reported that the active molybdenum produced by (γ, n) reaction had 17-min. half-life (8). Recently, however, Duffield and Knight (5) observed 15.5-min. half-life, which is in considerable agreement with the determination of the present experiment.

Since there are many stable isotopes of molybdenum, the assignment of the observed active molybdenum seems to be difficult. It is, however, probable that the present reaction is assigned to either Mo^{92} (γ, n) Mo^{91} or Mo^{94} (γ, n) Mo^{93}. But, the recent work of Duffield and Knight (5) using the molybdenum samples enriched in Mo^{92} and Mo^{94} established the assignment of this reaction, and namely the intensity of the reaction was found to be proportional to the Mo^{92} content of the irradiated samples, confirming its assignment to Mo^{92}. Therefore, as mentioned in the preceding section, the cross section of Mo^{92} (γ, n) Mo^{91} determined by the present experiment is

$$\sigma_{Mo} = 9.9 \times 10^{-26} \text{cm}^2 \quad \text{for Li} (p, \gamma) \gamma\text{-rays.} \qquad (9')$$

---

* Besides 17-min. half-life a shorter period of about one min. was observed by Baldwin and Koch using the X-rays of above 14 Mev (Phys. Rev. **67** (1945), 1) and Duffield and Knight (5) using the X-rays of 23 Mev.

Now comparing this value with the cross section for $Cu^{63}(\gamma, n) Cu^{62}$ reaction, $\sigma_{Cu} = 8.5_0 \times 10^{-26} cm^{2*}$, obtained by the previous work (2), we obtain :

$$\sigma_{Mo}/\sigma_{Cu} = 1.1_6 \text{ for } Mo^{92} \text{ and } Cu^{63} \text{ for } Li(p, \gamma) \gamma\text{-rays.} \qquad (10)$$

Taking into consideration the fact that the gradient of self-absorption curves for β-rays with various energies becomes smaller as the value of $mg/cm^2/Z^{\frac{1}{4}}$ decreases,** as shown in Fig. 4, it appears that the extrapolation of w to zero according to the linear relation (7) may give a little larger value of the cross section than the true one, however, the error introduced by this procedure is not larger than 5 per cent. The probable error of the results obtained by the present work, derived from the whole course of the experiments, is estimated to be about 15 per cent. But the result (10) seems to be more accurate, since the apparatus and experimental arrangements adopted in both works are essentially same.

Now, by the measurements of Perlman and Friedlander (7) it was made clear that the (γ, n) yields for the target nuclei with masses between 60 and 120 were in the same order for 50 Mev X-rays and, further, the same trend was also valid for 100 Mev X-rays. This fact, however, only suggests that the integrated cross sections for these X-rays are nearly equal. But from the present experiment we can conclude that the absolute cross section for copper and molybdenum are in the same order for lithium γ-rays.

The value of cross section above mentioned has been obtained on the assumption that the lithium-γ-rays constitute themselves of the quanta of 17.6 Mev monochromatically. But, according to the recent measurements of Walker and McDaniel (9) using a γ-ray spectrometer there are two lines instead of a single line, and namely a line at 17.6 Mev and another line at 14.8 Mev. And the ratio of intensities of the lower energy line to the higher energy line was found to be approximately 0.50 for the protons of 460 Kev bombarding energy. Then, it is proper that the effect due to the quanta of 14.8 Mev should be taken into account, since the observation by Baldwin and Koch (10) indicated that

---

* This value is, however, about 10 per cent larger than that previously obtained, $\sigma_{Cu} = 7.7_5 \times 10^{-26} cm^2$. This discrepancy is due to the difference of the counting efficiency adopted in both works, and namely $\varepsilon_{ff} = 0.21_7$ in the previous work while $\varepsilon_{ff} = 0.23_8$ in the present work. The difference of two values results from the difference of calculating procedures of efficiency. At present we can not ascertain, from the experiments, which value is more accurate.

** This effect was also observed in the previous work with Cu^{62}.

the threshold for Mo (γ, n) reaction was 13.5 ± 0.4 Mev. By simple considerations, it is clear that all corrections for 14.8 Mev quanta can be made, multiplying by a factor α which is given by

$$\alpha = \left(1 + \frac{\varepsilon_{11}}{\varepsilon_{17}}R\right) \Big/ \left(1 + \frac{\sigma_{14}}{\sigma_{17}}R\right),$$

where R is the ratio of intensities of the line at 14.8 Mev to that at 17.6 Mev, and ε_{17} and ε_{14} are the counting efficiencies of the γ-ray counter for 17.6 Mev and 14.8 Mev quanta respectively, and are given by M. Sonoda(3) theoretically as $\varepsilon_{17} = 0.23_8$ and $\varepsilon_{14} = 0.18_4$ respectively. σ_{17} and σ_{14} are the cross section for 17.6 Mev and 14.8 Mev quanta respectively. Since, however, at present the ratio σ_{14}/σ_{17} has been scarcely known, it is difficult to determine precisely the cross section for each of two lines of lithium-γ-rays.

Further work on the cross sections of photo-nuclear reactions with various nuclei is clearly necessary. In particular, the high yield of $\mathrm{Re}^{187}(\gamma, n)$ reaction observed by Perlman and Friedlander (7) tempts us to investigate (γ, n) reactions for the nuclei with masses above 120. Further studies on the dependence of cross section on nuclear structure are now in progress.

6. Acknowledgments

In connectin with the happy circumstance that this paper will appear in the number of this "Memoirs," issued in memory of Professor Arakatsu's 60th birthday, it is a great pleasure for us to express our most profound admiration for his important contribution to the development of nuclear physics in Japan.

REFERENCES

1. W. Bothe and W. Gentner, Zeits. f. Phys., **106** (1937), 236.
   H. Wäffler and O. Hirzel, Helvetica Phys. Acta, **21** (1948), 200.

2. S. Shimizu, Mem. Coll. Sci., Univ. of Kyoto, A, **25** (1950), 193.

3. M. Sonoda, Journ. Phys. Soc. Japan, now in press.

4. D. Williams and P. Yuster, Phys. Rev., **69** (1946), 556.

5. R. B. Duffield and J. D. Knight, Phys. Rev., **76** (1949), 573.

6. R. Peierls, Proc. Roy. Soc., A, **149** (1935), 467.

7. M. L. Perlman and G. Friendlander, Phys. Rev., **74** (1948), 442.

8. W. Bothe and W. Gentner, Naturwiss., **25** (1937), 191; Zeits. f. Phys., **106** (1937), 236.
   This period was also observed by $(n, 2n)$ reaction of molybdenum, (M. L. Pool, J. M. Cork, and R. L. Thorton, Phys. Rev., **52** (1937), 239; R. Sagane, S. Kojima, G. Miyamoto, and M. Ikawa, Phys. Rev., **54** (1938), 542.)

9. R. L. Walker and B. D. McDaniel, Phys. Rev., **74** (1948), 315.

10. G. C. Baldwin and H. W. Koch, Phys. Rev., **67** (1945), 1.

Nuclear Research Laboratory,
Department of Physics,
Faculty of Science,
University of Kyoto.