PHYSICAL REVIEW VOLUME 166, NUMBER 4 20 FEBRUARY 1968

Mass-Spectrometric Measurement of Lithium, Beryllium, and Boron Isotopes Produced in ^{16}O by High-Energy Protons, and Some Astrophysical Implications

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(Received 13 June 1967; revised manuscript received 27 September 1967)

A special mass-spectrometric method has been developed to determine the isotopic ratios and isotopic absolute cross sections of Li, Be, and B, formed by spallation of oxygen irradiated by protons of energy 135, 550, and 19 GeV. A brief description is given of the ion source of the mass spectrometer—of an ion sputtering type—which permits a very efficient ionization of beryllium and boron. The performances are given; they allow the analysis of 10^{-12} to 10^{-13} g of these elements. The oxygen target is made of very high purity water, and special handling techniques have been worked out to reduce contamination to an acceptable level. The determination of each of the isotopic ratios $^7Li/^6Li$, $^9Be/^7Be$, $^{10}Be/^9Be$, and $^{11}B/^{10}B$ requires a specific method, which is described. Isotope dilution techniques are used to obtain the absolute cross sections; values in millibarns for 135-MeV protons are as follows: ^6Li: 10±2, ^7Li: 8.0±2.5, ^9Be: 1.7±0.5, ^{10}Be: 0.35±0.20, ^{10}B: 11±3, and ^{11}B: 25±8. There are no significant changes at the higher energies. The above cross sections give support to conclusions based previously on calculated values concerning the spallation origin of Li, Be, and B in the solar system, in stars, and in the cosmic radiation.

I. INTRODUCTION

This work is part of a more extended study concerning the spallation production of Li, Be, and B in light elements (C, N, O). The results on lithium isotopes have already been published.[1,2] The motivation of these studies is mostly astrophysical. Indeed, the knowledge of the Li, Be, and B isotopic production cross sections is fundamental in at least three respects:

(1) *The origin of Li, Be, and B in the solar system.* The solution of this question depends essentially on the spallation ratios $^7Li/^6Li$ and $^{11}B/^{10}B$, and their values gave rise to different interpretations. In 1962, Fowler, Greenstein, and Hoyle[3] presented their well-known model, modified in 1965 by Burnett, Fowler, and Hoyle.[4] An alternative model proposed by Gradsztajn[5] in 1965 was based on experimental[1] and calculated[6] cross sections. This model has since been developed by Bernas, Gradsztajn, Reeves, and Schatzman.[7] For more details, the reader may refer to the original literature.

(2) *The understanding of the lithium and beryllium abundances observed at the surface of some stars.*[8–11] Assuming the hypothesis of Fowler *et al.*[12] of a spallation origin of Li, Be, and B at the surface of stars, the comparison of the spallation quantities Li/Be and $^7Li/^6Li$ to the observed ones is expected to give some information on the evolution of stars.[8,13]

(3) *The chemical composition of the galactic cosmic rays at the source.* In cosmic rays, the abundance of Li, Be, and B relative to C, N, O, and Ne is 10^6 times greater than in the solar system. Their presence is interpreted as a product of the spallation and fragmentation of C, N, O, Ne, Si, and Fe on the interstellar hydrogen. The determination of the spallation cross sections would then allow one to determine accurately the amount of matter traversed by the cosmic rays and their composition at the source, knowing the exact composition near the earth.

II. EXPERIMENTAL METHOD

Beryllium and boron in contrast to lithium, cannot be efficiently ionized by the mass-spectrometer thermionic ion source. We were thus led to set up a very sensitive sputtering ion source, similar to that developed by Castaing and Slodzian.[14] Our arrangement allows us, at the present time, to analyze the 10^{-13} to 10^{-12} g of beryllium and boron produced by the spallation of carbon and oxygen in a few μA h proton bombardment.

The spallation products are extracted from the irradiated targets and deposited on the metallic sample plate, which is bombarded by the primary ion beam. Our apparatus is a simplified version of that of Castaing and Slodzian, since we need produce only a secondary ion beam from the deposited sample and not an image of the bombarded spot.

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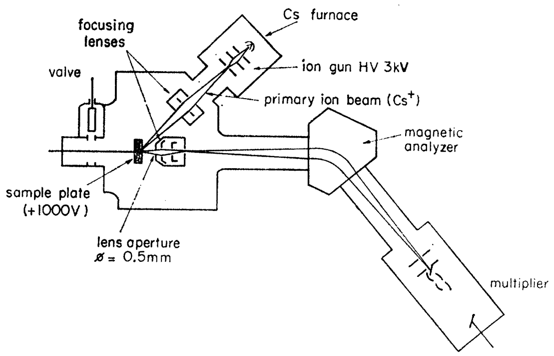

FIG. 1. Ion sputtering mass spectrometer built for the analysis of extremely small samples of lithium, beryllium, and boron.

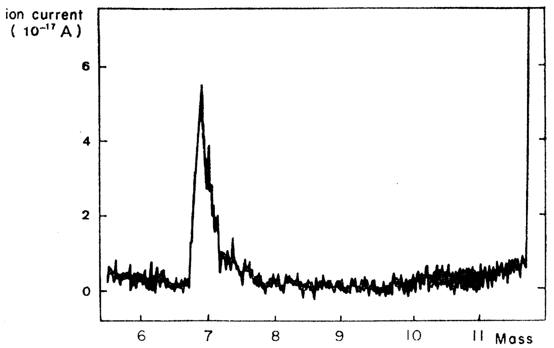

FIG. 2. Mass spectrum obtained with a clean sample plate and no deposit. (Peaks at mass 12 and above are always present and most likely due to hydrocarbons.) Note that the ^{7}Li peak present is 0.1% of the ^{7}Li peak obtained after irradiation (Fig. 4).

Figure 1 represents the apparatus, with its primary ion source yielding a focused beam impinging on the sample plate from which secondary ions are emitted. This secondary ion beam is accelerated to 1000 V, focused by a 3-element lens, and directed towards the magnetic analyzer. The isotopic beams at the collector are measured by an electron multiplier. For the sake of simplicity, the ion gun is essentially composed of a tantalum furnace, emitting Cs$^+$ ions by surface ionization, and of a focusing lens. The cross section of the 2.000-eV ion beam is about 2 mm^2, and its density at the sample plate is 10^{-9} A/mm.^{2} The sample plate is a polished stainless-steel disk, 2 cm in diameter, on which first chromium and then high-purity gold are vacuum-evaporated.

Instead of the adopted values of 4.0±0.1 [15] for the natural ^{11}B/^{10}B ratio and 12.27±0.03 [16] for the natural ^{7}Li/^{6}Li ratio, we obtain, respectively, 3.5±0.1 and 11.4±0.1. Hence a correction factor has been used for all our measurements.

Figure 2 shows the mass spectrum obtained without any deposit on a clean sample plate, and Fig. 3 shows the over-all sensitivity (mass spectrum corresponding to a water sample containing 10^{-12}-g enriched ^{10}B and 1.3 $\times 10^{-12}$-g enriched ^{6}Li. At this point, it must be stressed that the efficiency of the sputtering process varies very significantly from one element to another, and this will be made use of later. It appears from many measurements that, under our experimental conditions, a solution containing equal amounts of ^{7}Li, ^{9}Be, and ^{11}B will give rise to peaks with relative heights approximately equal to 80:1:0.1.

The oxygen targets consisted of 30 g of very pure water contained in a platinum vessel 2 cm in diameter and 7 cm long. Proton irradiations were performed in external beams at 155 MeV[17] (ORSAY), 600 MeV, and 19 GeV (CERN). After bombardment, 10-g fractions were concentrated to 100 mg, which were then deposited on the metallic sample plate of the ion source, evaporated to dryness, and analyzed.

After many unsuccessful attempts involving various methods of preparation, water, containing less than 10^{-12} g/g of either lithium or beryllium and less than 10^{-11} g/g of boron, was prepared by fractional crystallization.

The handling of the water samples was performed in an argon atmosphere. While no contamination with natural beryllium could ever be detected during a complete manipulation, contamination with natural boron usually reached 10^{-10} g/g. Contamination with lithium was of the order of 10^{-12} g/g. As a consequence, the proton flux in our experiments was always such that the amount of boron produced by spallation would be several times greater than that due to contamination. In Table I we have summarized the situation with respect to natural and spallation-produced boron in a 20-h bombardment (135 MeV).

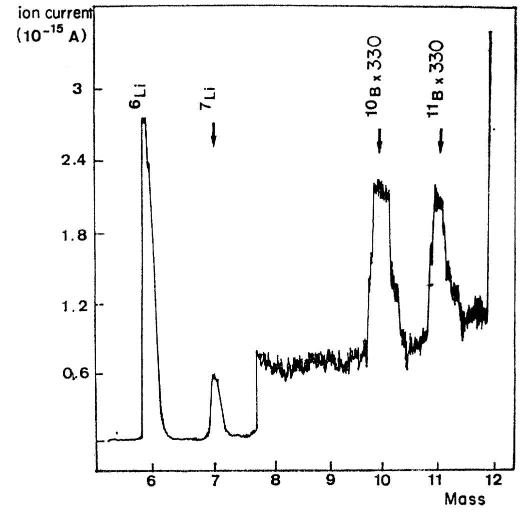

FIG. 3. The sensitivity of the ion sputtering mass spectrometer: mass spectrum of a water sample containing 10^{-12} g of enriched ^{10}B and 1.3×10^{-12} g of enriched ^{6}Li. Note the absence of peaks at masses 9 and 8.

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[17] Because of the energy loss in the thick target, the mean bombarding energy will be given as 135 MeV.

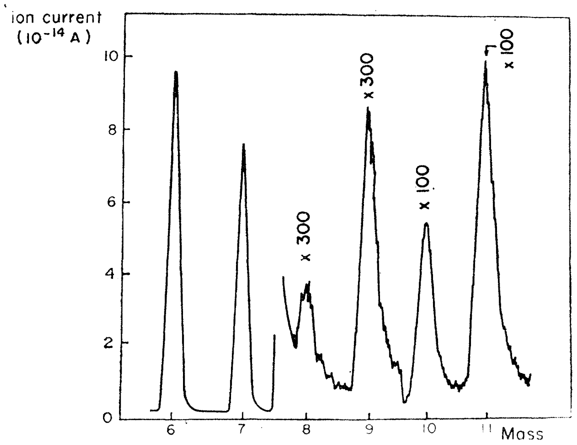

FIG. 4. Mass spectrum obtained from a proton-irradiated water sample. See Fig. 5 for the various contributions to each mass peak.

III. ISOTOPIC RATIO MEASUREMENTS

A typical spectrum obtained after a 20-h bombardment is shown on Fig. 4, while Fig. 5 represents schematically the contribution of the isotopes of Li, Be, and B to the different peaks; that neither mass 8 nor mass 9 is present in unirradiated water samples containing only lithium and boron was already apparent from Fig. 3.

The presence of a mass-8 peak in irradiated water samples is due to a peculiarity of beryllium: As observed also by Collins and McHugh,[18] the sputtering of beryllium gives rise to a BeH^+ peak. Thus the apparent mass 8 is due to $^7BeH^+$ ions. For the same reasons, ^9Be, either natural or spallation produced, will give a peak at mass 10 due to $^9BeH^+$.

The existence of a BeH peak being due to a chemical process, we have made the assumption that $^7BeH/^7Be$ is not different from $^9BeH/^9Be$, but it was not quite certain that spallation ^7Be or ^9Be would behave in the same way as natural ^9Be. Hence a method was devised which permitted a direct measurement of $^7BeH/^7Be$ ratio. This method will be described below. The fact that we do not observe any peak at mass 8 in nonirradiated water shows that LiH^+ is not formed. As to the presence of BH^+, we have shown by comparing peaks at masses 11 and 12 that, if any BH^+ is formed, the ratio BH/B is smaller than 1/40.

TABLE I. Results of the analysis of H_2O targets after proton bombardment (135 MeV). (Numbers in columns 2 to 6 are in units of 10^{12} atoms/g.)

Target	Total number of incident protons	Boron pollution n_{11}	Boron pollution n_{10}	Amounts produced by spallation: Boron N_{11}	Boron N_{10}	Beryllium N_7
A	$\simeq 10^{16}$	1.0	0.24	2.34	1.13	...
B	1.4×10^{17}	5.5	1.3	27.7 ± 2.6	12.3 ± 1	6.0 ± 0.2

1. Determination of the $^7Li/^6Li$ Ratio

As we have pointed out earlier, lithium ions are emitted approximately 80 times more efficiently than beryllium ions from equal amounts of the elements. Hence, the contribution of ^7Be to the peak at mass 7 is only about 1% of the contribution of ^7Li, and we have taken the ratio $^7Li/^6Li$ to be that of the ion currents at these two mass values except for this small correction of the mass-7 peak which we base on the height of the mass-8 peak.

2. Determination of the $^9Be/^7Be$ Ratio

Two possibilities exist of obtaining this ratio: One consists in relying on the BeH/Be value to deduce from the mass-8 peak the height of the peak at mass 7 due to ^7Be and compare it to the peak at mass 9 due only to ^9Be. The other method is based on the possibility of eliminating selectively the lithium from the deposit made on the sample plate by the water sample. As this last method turned out to be easily performed, we preferred to use it. Thus after having deposited on the sample plate the 100 mg of irradiated water and having dried and analyzed it, we added a 10-mg drop of very pure water, which easily dissolved the lithium present and only a small fraction of the beryllium, and was then pipeted out.

In this way, the lithium contribution to mass 7 is reduced to less than 5% of the peak height, and this can be checked and corrected by the corresponding residual ^6Li peak at mass 6. The ratio $^9Be/^7Be$ is thus given by the ratio of the ion currents at masses 9 and 7. The time sequence for these operations must, however, be followed carefully, since the decay of ^7Be to ^7Li as the sample is being analyzed yields a non-negligible increase in peak height at mass 7 due to the 80-fold higher efficiency of emission of ^7Li with respect to ^7Be. Indeed, during a 2-h analysis the mass-7 peak increases by about 9%. We shall see in the next section that a value of the ratio BeH/Be is needed to measure $^{10}Be/^9Be$. The method just described allowed us to measure directly the $^7BeH/^7Be$ ratio; the value obtained is 0.085 ± 0.015, which is the average of the measurements of seven deposited samples taken from two irradiated water targets.

3. Determination of $^{10}Be/^9Be$ and $^{11}B/^{10}B$ Ratios

The ^{10}Be contribution to mass 10 is not directly measurable: as shown before (Fig. 5), three different types of ions contribute to mass 10: ^{10}B, (^9BeH), and ^{10}Be. However, even at concentrations as low as 10^{-10} g/g, Be and B may be discriminated, owing to their different rates of crystallization from aqueous solutions. Thus if we deposit and dry a solution containing equal amounts of Be and B on an area of about 30-mm,[2] the quantities of ^9Be and ^{10}B at any one point of that surface will generally not be the same.

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[18] T. L. Collins and J. A. McHugh, *Advances in Mass Spectrometry* (Elsevier Publishing Company, Inc., Amsterdam, 1966), Vol. III, p. 169.

As the area from which ions emitted from the sputtered sample enter the first lens of the ion source is quite small, much smaller than the 2 mm^2 of the incoming Cs$^+$ beam, slight displacements (as small as 0.2 mm) of the sample plate will lead to significant variations in the ratio of beryllium to boron at each position, and this can be used to give indications on the Be and B isotopic composition of our mixture.

Let us refer again to Fig. 5, and represent the height of the peak at mass M ($M=6$, 7, 8, 9, 10, 11) by the symbol M, the contribution at mass M of an individual isotope, for example ^{10}Be at mass 10, by (^{10}Be), and the absolute concentration in the solution (proportional to the formation cross section) by ^{10}Be. We can write

$$r_1=(10)/(9)=[(^{10}\mathrm{B})+(^{10}\mathrm{Be})+(^{9}\mathrm{BeH})]/(^{9}\mathrm{Be}),$$

$$r_2=(11)/(9)=[(^{11}\mathrm{B})+(^{10}\mathrm{BeH})]/(^{9}\mathrm{Be}).$$

If γ represents the discrimination factor of the mass spectrometer for Be and B ($\gamma=1.15$) and $1/R_0=$ (BeH)/(Be), the following relation between r_1 and r_2 is obtained:

$$r_1=\frac{1}{\gamma}\frac{^{10}\mathrm{B}}{^{11}\mathrm{B}}\times r_2+\frac{1}{R_0}+\gamma\frac{^{10}\mathrm{Be}}{^{9}\mathrm{Be}}\left[1-\frac{1}{\gamma}\frac{^{10}\mathrm{B}}{^{11}\mathrm{B}}\times\frac{1}{R_0}\right],$$

which is represented by a straight line the slope of which gives the ratio $(1/\gamma)^{10}\mathrm{B}/^{11}\mathrm{B}$ and the intercept the quantity

$$\frac{1}{R_0}+\gamma\frac{^{10}\mathrm{Be}}{^{9}\mathrm{Be}}\left[1-\frac{1}{\gamma}\frac{^{10}\mathrm{B}}{^{11}\mathrm{B}}\times\frac{1}{R_0}\right].$$

Since $^{10}\mathrm{B}/^{11}\mathrm{B}<1$, the second term

$$(1/\gamma)(^{10}\mathrm{B}/^{11}\mathrm{B})\times 1/R_0<\tfrac{1}{10}$$

can be neglected in a first approximation, so that we obtain the very simple relation

$$r_0=(1/\gamma)\times(^{10}\mathrm{B}/^{11}\mathrm{B})\times r_2+1/R_0+\gamma\times{}^{10}\mathrm{Be}/^{9}\mathrm{Be}. \quad (1)$$

The ratios ^{10}Be/^{9}Be and ^{10}B/^{11}B can then be determined, hence the corresponding cross sections.

In order to check this method, we have applied it to the analysis of a mixture of natural beryllium and boron added to some purified water. The concentration of the solution was 10^{-9} g/g of Be and B. Figure 6 shows the variations of r_1 as a function of r_2 in the analysis of different points of the sample.

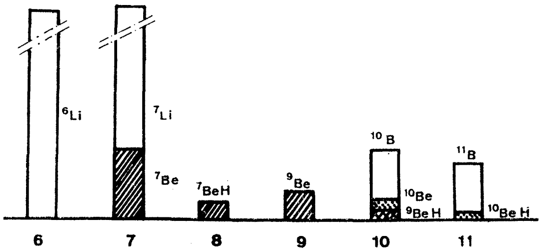

FIG. 5. Schematic mass spectrum showing the contributions to the various peaks in irradiated water.

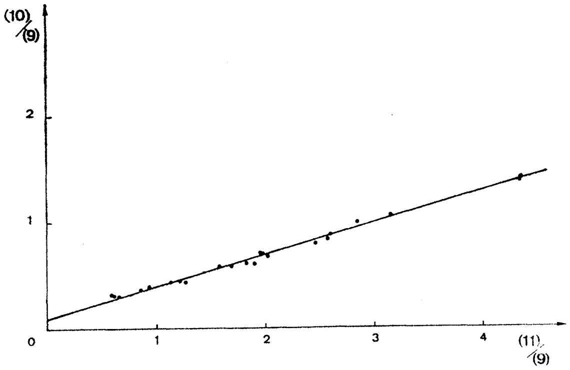

FIG. 6. Variation of the ratio (11)/(9) as a function of (10)/(9) for different impact points of the cesium ion beam on the sample plate, for a mixture of natural beryllium and boron.

Since there is no ^{10}Be present in this water, the ordinate at the origin is $1/R_0={}^{9}\mathrm{BeH}/^{9}\mathrm{Be}$. As we can see on Fig. 6, the value obtained is 0.09±0.015, and it can be compared with the value of ^{7}BeH/^{7}Be obtained by the direct method previously described (0.085±0.015).

In the course of preliminary measurements of the ^{9}BeH/Be ratio using a solution of natural beryllium made with water purified only by ion exchange, we could observe that this ratio had a different value ($1/R_0=0.12\pm0.02$). Hence we took care to determine BeH/Be both in irradiated and unirradiated water targets purified by crystallization. The fact that we find identical results for ^{7}BeH/^{7}Be (in irradiated water) and ^{9}BeH/^{9}Be (nonirradiated water) allows us to use the 0.09 value in the method just described, where

TABLE II. Values of the (10)/(9) and (11)/(9) ratios corresponding to the points on straight lines I and II in Fig. 7. The uncertainties vary from 2 to 5%.

Curve I		Curve II	
(10)/(9)	(11)/(9)	(10)/(9)	(11)/(9)
0.46	0.36	0.73	0.34
0.52	0.36	0.99	0.60
0.43	0.38	3.01	2.23
0.50	0.48	3.24	2.36
0.49	0.49	3.28	2.49
0.51	0.51	3.28	2.54
0.59	0.59	3.57	2.65
0.60	0.67	3.95	3.01
0.57	0.68		
0.66	0.78		
1.44	2.58		
1.53	2.59		
1.56	2.72		
1.65	2.77		
1.71	2.93		
1.71	3.02		
1.83	3.21		
1.82	3.36		
1.91	3.42		
2.13	3.92		
2.17	3.94		

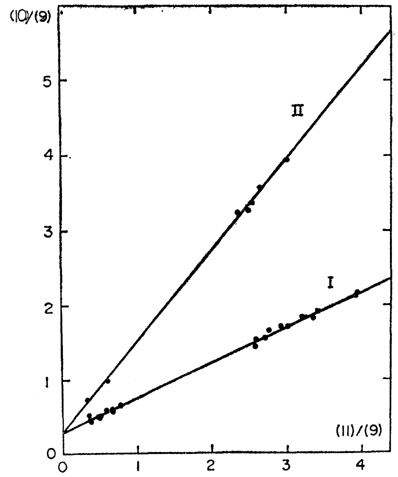

FIG. 7. Variation of the ratio (10)/(9) as a function of (11)/(9) for different impact points of the cesium ion beam on the sample plate on which was deposited the irradiated water: Curve I: (10)/(9) as a function of (11)/(9) for 10 g of irradiated water. Curve II: (10)/(9) as a function of (11)/(9) for 10 g of irradiated water to which are added 4×10^{-9} g of ^{10}B.

the ordinate at the origin gives $1/R_0+\gamma\,^{10}\mathrm{Be}/^{9}\mathrm{Be}$. From this, we can deduce the value of ^{10}Be/^{9}Be.

The slope of the straight line of Fig. 6 leads to ^{10}B/^{11}B $=0.29\pm0.02$. This value is in good agreement with the result 0.28 ± 0.02, obtained by a direct measurement of ^{10}B/^{11}B, which justifies the use of our method for the determination of the ^{10}B/^{11}B spallation ratio in the irradiated water target.

Figure 7 represents the variation of (10)/(9) versus (11)/(9) and Table II contains the corresponding ratios for each of the points on the sample where a mass-spectrometric analysis has been performed. We may remark that, at some points of the sample, beryllium is almost totally absent so that the peak at mass 10 is pure boron, thus giving the ^{11}B/^{10}B ratio. Inversely, for some other points, the boron quantity is low compared to the beryllium one, thus giving the (^{10}Be+^{9}BeH)/^{9}Be ratio. However, the method described above, the results of which appear on Fig. 7, confirms these values and has the advantage of a greater accuracy. The final results obtained from the different irradiations are presented in Tables III and IV.

IV. ABSOLUTE CROSS SECTIONS (TABLE IV)

1. ^{7}Be-^{9}Be-^{10}Be

These cross sections are normalized to a radiochemically determined ^{7}Be absolute value which we have obtained in the irradiation conditions discussed above (7-cm-long water target). This value does not differ from the ones obtained previously with a thin target.[19,20]

The ^{7}Be cross section which we have measured has been obtained by reference to the $^{27}\mathrm{Al}(p,3p3n)^{22}\mathrm{Na}$ monitor cross section, for which the value of 18 mb at 150 MeV has been taken from Brun *et al.*[21] We have taken the branching ratio for the decay of ^{7}Be into the 0.477-MeV level of ^{7}Li to be 0.10.[22]

2. ^{10}B and ^{11}B

The determination of the absolute amount of boron produced in the irradiated water has been done using a classical isotope dilution method. Ten grams of this water were analyzed and the ratio $R_1=(N_{11}+n_{11})/(N_{10}+n_{10})$ was measured, N_{11} and N_{10} being the number of atoms of the boron isotopes produced by the irradiation, n_{11} and n_{10} those present in the water before irradiation or added through contamination during the experiment. (n_{11} and n_{10} are determined in a separate measurement: see below under contamination.)

Another 10-g sample of the irradiated water was analyzed after the addition of a spiking solution containing s_{11} and s_{10} atoms of enriched boron. The measured ratio

$$R_2=(N_{11}+n_{11}+s_{11})/(N_{10}+n_{10}+s_{10})$$

combined with R_1 gave N_{11} and N_{10}. Then the comparison with N_7, the quantity of ^{7}Be produced during the same irradiation, leads to the ^{11}B and ^{10}B production cross sections, which include, of course, the contributions due to the decay of ^{11}C and ^{10}C, respectively.

The ratios R_1 and R_2 were determined by the point analysis described earlier (see Fig. 7), and it is to be noted that the ^{10}Be/^{9}Be ratios measured in these two analyses are equal.

3. Contamination

We have done a careful study of boron contamination due to the container and atmospheric air, and determined its increase as a function of time. In order to include possible accidental contamination, a blank target, identical to the irradiated one, was prepared together with it, placed in the accelerator room during the time of the bombardment, and handled exactly in the same way and in the same glove-box as the active

TABLE III. Isotopic ratios obtained by proton bombardment of H_2O targets.

Energy	Target	^{7}Li/^{6}Li[a]	^{9}Be/^{7}Be	^{10}Be/^{9}Be	^{11}B/^{10}B
135 MeV	*A*		0.31 ± 0.05	0.18 ± 0.06	2.1 ± 0.6
135 MeV	*B*	0.8 ± 0.09	0.32 ± 0.04	0.21 ± 0.05	2.3 ± 0.4
600 MeV	*C*		0.37 ± 0.10	0.24 ± 0.05	2.1 ± 0.6
19 GeV	*D*		0.34 ± 0.08	0.29 ± 0.08	<2.9

[a] Without any contribution from the decay of ^{7}Be.

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[19] G. Albouy, J. P. Cohen, M. Gusakow, N. Poffe, H. Sergolle, and L. Valentin, Phys. Letters 2, 306 (1962).

[20] J. P. Cumming, Ann. Rev. Nucl. Sci. 13, 261 (1963).

[21] C. Brun, M. Lefort, and X. Tarrago, J. Phys. Radium 23, 371 (1962).

[22] T. Lauritsen and F. Ajzenberg-Selove, Nucl. Phys. 78, 49 (1966).

TABLE IV. Absolute cross sections (in mb) obtained by bombardment of H_2O targets.

Proton energy	Target	^{6}Li[a]	^{7}Li[b]	^{7}Be	^{9}Be	^{10}Be	^{10}B[c]	^{11}B[c]
135 MeV	*A*[d]				1.9±0.6	0.3 ±0.2		
135 MeV	*B*	10±2	8.0±2.5	5.4±1	1.7±0.5	0.35±0.20	11±3	25±8
600 MeV	*C*			6.5±2	2.4±1.2	0.6 ±0.4	12±5	25±12
19 GeV	*D*			6.5±2	2.2±1.1	0.64±0.5		

[a] E. Gradsztajn, M. Epherre, and R. Bernas, Phys. Letters **4**, 257 (1963).
[b] With no contribution from ^{7}Be decay.
[c] ^{10}B and ^{11}B include contributions from ^{10}C and ^{11}C, respectively.
[d] Because of the absence of monitoring only relative isotopic abundances have been obtained (see Table III) with this target. ^{9}Be and ^{10}Be cross sections are deduced by combining ratios from Table III and the ^{7}Be cross section from target *B*.

one. Finally, the ^{11}B and ^{10}B present were measured by isotope dilution. The proton flux incident on the target and the values of n_{11}, n_{10}, N_{11}, N_{10}, and N_7 are given in Table I for the 135-MeV irradiations.

4. ^{6}Li-^{7}Li

These cross sections have been measured previously.[1] However, despite the great care used in handling the samples at that time, some slight residual contamination amounting to approximately 10^{-14} g must have occurred. Our new experimental technique, and the longer bombardments performed presently, lead to an amount of spallation-produced lithium several times larger than the residual natural amount, and thus to a much higher accuracy. The new ^{7}Li/^{6}Li ratio of Table III associated with the former ^{6}Li cross section[1] yields the new ^{7}Li absolute cross-section value given in Table IV. This value does not include any contribution from the decay of ^{7}Be.

5. Remarks Concerning the Proton Bombardment

As a consequence of the target thickness, the incoming protons will give rise to a number of secondary neutrons which cannot be neglected. If we consider that neutrons capable of producing Li, Be, and B from ^{16}O must have an energy greater than 40 MeV, we can estimate that their abundance is about 5% of that of the incident protons [this results from the fact that energetic neutrons are essentially produced by (p,pn) reactions]. From the work of Fuller[23] and Kellog,[24] the production of Be and B from ^{16}O and ^{12}C bombarded by neutrons of 300 and 90 MeV, respectively, corresponds to cross sections which are similar to those measured here. Thus, although it may be difficult to make an accurate correction to our measured values, it is most likely smaller than 10%. In any case, it must be realized that the true cross sections should not be larger than those obtained here.

V. CONCLUDING REMARKS

Let us refer again to the implications already mentioned in the Introduction:

(a) The present measurements confirm the values of ^{11}B/^{10}B and Li/^{9}Be calculated by the Monte Carlo method or estimated in a previous paper[5] for the spallation of carbon and oxygen at 150 MeV (see Table V).

Now, if one assumes (as in Refs. 3-5, 7) that the Li, Be, and B present in the solar system originate from the spallation of C, N, and O, one has to take into account the energy spectrum of the solar protons assumed to be responsible for the spallation of C, N, and O; the variation of the cross sections with energy, from the thresholds to the GeV region[25]; and the production due to nitrogen.

These points have indeed been considered by Bernas, Gradsztajn, Reeves, and Schatzman,[7] and their conclusion confirms that, in any spallation model of the nucleosynthesis of Li, Be, and B, the initial formation ratios ^{11}B/^{10}B and Li/^{9}Be will not have to be significantly altered by subsequent nuclear processes in order to yield the corresponding values observed on the earth and in the meteorites,[26,27] as was already stated in Ref. 5.

(b) The cross-section values obtained in the present work have been used in a detailed calculation of the production rate of Li, Be, and B in galactic cosmic rays.[28,29] The comparison of these calculated results with

TABLE V. Comparison between the values calculated by Gradsztajn (Ref. 5) and the present measurements for some isotopic or elemental ratios obtained by 135-MeV proton spallation of ^{16}O and ^{12}C.

Target	Oxygen ^{11}B/^{10}B	Oxygen Li/^{9}Be	Oxygen B/Li	Carbon Li^9/Be
Calculated	2	20	1.5	20
This work[a]	2.3±0.4	14±4	1.5±0.4	30 (preliminary)

[a] The lithium values indicated here include the contribution from ^{7}Be decay.

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the observed elemental ratios of Li, Be, and B relative to C, N, and O in cosmic rays of energies higher than 1.5 GeV/nucleon[30,31] strengthens the hypothesis of a spallation origin of Li, Be, and B in the cosmic radiation and points out to the stability of ^{7}Be in cosmic rays.

Finally, the relatively low value of the production cross section of ^{10}Be in ^{16}O (and the preliminary value in ^{12}C) will make it more difficult than initially expected to use this isotope as a "clock" for the determination of cosmic-ray "ages," at least in connection with the expected difference in the ratio of Be/B between laboratory experiments and cosmic-ray measurements at similar energies.[32–34]

ACKNOWLEDGMENTS

We wish to express our thanks to Professor B. Gregory, director-general of CERN, for the use of the CERN accelerators and to Dr. E. Herz, Dr. E. G. Michaelis, and E. Dahl-Jensen who have been most helpful in arranging for the irradiations. Our thanks are due also to E. Alorent, C. Thouault, and M. Salomé for their skillful help in the setting up and in the careful operation of the mass spectrometer.

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PHYSICAL REVIEW VOLUME 166, NUMBER 4 20 FEBRUARY 1968

Intrinsic Matrix Elements in the *s*-*d* Shell

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(Received 20 September 1967)

A new table of the intrinsic matrix elements of a two-body scalar Hamiltonian independent of spin is presented. The space spanned is that subspace of the *s*-*d* shell which contains the more deformed intrinsic states. The table is shown to be especially useful for the study of the relative position of bands belonging to the same $SU(3)$ representation.

I. INTRODUCTION

THE aim of this work is to present a new table of the intrinsic matrix elements of a two-body scalar Hamiltonian in the *s*-*d* shell. The table does not contain more information than that in Tables VII and IX of Ref. 1, from which it was derived, but the information is more readily available.

The table will be found useful for the study of the dependence of the intrinsic matrix elements on the two-body interaction. It has already been utilized in our work[2,3] on the order of levels in ^{22}Ne and lately in the work being done on ^{24}Mg.

The usefulness of the table was first suggested by Levinson in 1961, and the table was then computed by Kugler and myself. Unfortunately, we performed this calculation with decimals, and because of the accumulation of round-off errors, the final results were too inaccurate to be used in quantitative work. We have now precisely recomputed the parameters appearing in the table (as quotients of integers) by making use of the Elliott 503 Computer.

II. DEFINITIONS AND NOTATIONS

With the exception of Eq. (1), all the quantities and symbols used in this and the following sections are defined in Ref. 1. Nevertheless, for the sake of readability, we repeat here some of the definitions.

The Hamiltonian of a N-nucleon system in the space of the *s*-*d* shell can be written as

$$H=\sum_{p=1}^{10} M_p H_p\,, \tag{1}$$

if the N nucleons interact through a two-body scalar interaction independent of spin. The matrix elements M_p depend only on the two-body interaction and not on the nuclear configuration. They are defined as follows:

$$\begin{aligned}
&M_1\equiv\langle s^2,0|H|s^2,0\rangle\,, && M_6\equiv\langle d^2,2|H|d^2,2\rangle\,,\\
&M_2\equiv\tfrac{1}{5}{}^{1/2}\langle s^2,0|H|d^2,0\rangle_s\,, && M_7\equiv\langle d^2,4|H|d^2,4\rangle\,,\\
&M_3\equiv\langle d^2,0|H|d^2,0\rangle\,, && M_8\equiv\langle d^2,1|H|d^2,1\rangle\,,\\
&M_4\equiv\langle sd,2|H|sd,2\rangle_s\,, && M_9\equiv\langle sd,2|H|sd,2\rangle_a\,,\\
&M_5\equiv(1/14)^{1/2}\langle sd,2|H|d^2,2\rangle_s\,, && M_{10}\equiv\langle d^2,3|H|d^2,3\rangle\,.
\end{aligned} \tag{2}$$

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