

analysis of (6b) shows that for $\alpha \neq 0$, a continuous displacive transition will occur at some $\lambda_c(\alpha)$ with $\lambda_c(\alpha)$ an increasing function of α . An interesting feature of this instability-driven transition is that the transition becomes increasingly sharp as $\alpha \rightarrow 0$. Thus, if this were used to model a real physical system, a case could occur in which it would be impossible experimentally to determine that the transition was not first order. The results for (6b) are as shown in Table I.

In conclusion, a clearer picture of the transition found in Ref. 4 is now available. In the absence of a symmetry-breaking term there should be no transition. In the presence of one (which could be implicitly introduced through the MFA), there should be a second-order transition. These results are for an idealized model. In a real, physical system, there should be at least a weak symmetry-breaking term. Then, since the transition is of the instability-driven type (with the instability of the Coulomb lattice providing an effective "hump" for the TO mode) there would be a sharp second-order transition with almost a first-order character. If this were accompanied by, for example, a lattice distortion, the transition could be a real, first-order transition.

Thus, the first-order character of either the MFA or SPA treatment of the instability-driven transition should not be considered spurious, but rather a strong indication of the inherent instability of the system.

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¹E. Pytte and J. Feder, *Phys. Rev.* **187**, 1077 (1969).

²H. Thomas, in *Proceedings of the Advanced NATO Study Institute on Structural Phase Transitions and Soft Modes*, Geilo, Norway, April, 1970 (to be published).

³N. S. Gillis and T. R. Koehler, *Phys. Rev. B* **4**, 3971 (1971).

⁴N. S. Gillis and T. R. Koehler, *Phys. Rev. B* **5**, 1925 (1972).

⁵E. Pytte, *Phys. Rev. Lett.* **28**, 895 (1972).

⁶The full details of this study will be given in a subsequent publication.

⁷Any quartic Hamiltonian with a single degree of freedom can be scaled to the form (6).

⁸This is also the region where $k_B T$ is of the order of the depth of one of the minima of the effective double-well potential associated with the long-wavelength optic mode.

Half-Life of ^{10}Be

F. Yiou and G. M. Raisbeck

Centre de Spectrométrie de Masse du Centre National de la Recherche Scientifique, 91-Orsay, France

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The half-life of ^{10}Be has been measured to be $(1.5 \pm 0.3) \times 10^6$ yr, a value in significant disagreement with the previously accepted value of $(2.7 \pm 0.4) \times 10^6$ yr. We discuss several implications of the revised half-life.

Cosmogenic radionuclides having very long half-lives can be useful dating tools in a number of astrophysical, geophysical, and cosmochemical problems. One of the most prominent of such species is the nuclide ^{10}Be , which has significant applications in studying the history of cosmic rays,¹ meteorites,² lunar samples,³ marine sediments,⁴ and polar deposits.⁵

The generally accepted⁶ half-life of ^{10}Be is $(2.7 \pm 0.4) \times 10^6$ yr, a value based on two separate experiments carried out more than 20 years ago.^{7,8} We have become interested in the question of the accuracy of this number because of its importance in trying to "date" cosmic-ray ages.¹ Although we had no reason to suspect a large error

in the accepted value, we felt its widespread use warranted an attempt to reduce the error limits. While we have not yet succeeded in that goal, we have found a value which is in strong disagreement with the older number, and which should have important implications in a number of fields of study.

The two essential quantities in the determination of any half-life which is too long to be followed directly are the decay rate and the number of atoms of the decaying species. Usually the former can be measured directly, while the latter is obtained by a mass-spectrometric comparison of the radioactive nuclide and a stable isotope. The main problem with the second step is the

difficulty in having a sufficiently small stable radioactive ratio for a good spectrometric determination, while still having enough stable species for an accurate analytical analysis.

In the present work we have taken a somewhat different approach, avoiding the stable isotope. Essentially, our method consists of measuring, by two separate techniques, the relative nuclear cross sections for production of the two radioactive species ${}^7\text{Be}$ and ${}^{10}\text{Be}$. One of these techniques is directly dependent on the half-life of ${}^{10}\text{Be}$, while the other is independent of this factor. Thus, by comparing the two determinations, we are able to extract the correct half-life.

The system we have chosen for this study is the high-energy proton spallation of ${}^{12}\text{C}$. The details of the two experimental techniques have been described elsewhere, and we only briefly summarize them here. A high-purity graphite target was irradiated in the internal beam of the Orsay (150 MeV) or CERN (600 MeV) synchrocyclotron for ~ 30 h. The carbon was then burned in air, with the resulting spallation products being deposited on a gold disk (with $\sim 90\%$ efficiency in the case of Be). After heating under vacuum to remove Li, the cross-section ratio $\sigma({}^{10}\text{Be})/\sigma({}^7\text{Be})$ can be determined directly from this deposit with our sputtering mass spectrometer.⁹ Thus one has

$$\frac{\sigma({}^{10}\text{Be})}{\sigma({}^7\text{Be})} = \frac{H({}^{10}\text{Be})}{H({}^7\text{Be})} \gamma^3 e^{-\lambda t}, \quad (1)$$

where H is the directly measured peak height, γ is the discrimination factor of the mass spectrometer, λ is the decay constant for ${}^7\text{Be}$, and t is the time since irradiation.

Using similar deposits, we then applied a technique which we have recently developed for measuring ${}^{10}\text{Be}$ from ${}^{11}\text{B}$ targets.¹⁰ The Be isotopes were dissolved off the gold disks in a dilute HCl

solution containing several milligrams of ${}^9\text{Be}$ carrier. The Be was then precipitated with $\text{NH}_4(\text{OH})$, converted to the oxide and introduced into the isotope separator SIDONIE.¹¹ The two isotopes ${}^7\text{Be}$ and ${}^{10}\text{Be}$ were collected simultaneously as BeCl^+ ions and their disintegration rates determined by γ -ray and low level β -particle counting, respectively. With this procedure one determines the same ratio as

$$\frac{\sigma({}^{10}\text{Be})}{\sigma({}^7\text{Be})} = \frac{(dN/dt)_{10\text{Be}} t_{1/2}({}^{10}\text{Be})}{(dN/dt)_{7\text{Be}} t_{1/2}({}^7\text{Be})} e^{-\lambda t}, \quad (2)$$

where dN/dt and $t_{1/2}$ are the disintegration rates and half-lives, respectively, for the different species. Combining Eqs. (1) and (2) one thus has immediately that

$$t_{1/2}({}^{10}\text{Be}) = \frac{(dN/dt)_{7\text{Be}}}{(dN/dt)_{10\text{Be}}} t_{1/2}({}^7\text{Be}) \frac{H({}^{10}\text{Be})}{H({}^7\text{Be})} \gamma^3. \quad (3)$$

The results of our two experiments are summarized in Table I, where the various quantities entering into Eq. (3), and their estimated errors, are given.

Values for the ratio $\sigma({}^{10}\text{Be})/\sigma({}^7\text{Be})$ from carbon targets have previously been published.⁹ They were remeasured here using the same targets as utilized in the separator experiment in order to avoid any systematic errors which might arise from different irradiations. The values of $H({}^{10}\text{Be})/H({}^7\text{Be})$ listed in Table I, when corrected for γ^3 , are (within experimental error) in agreement with the published values. In all previous Be work the value for γ has been interpolated^{9,12} because Be has only a single stable isotope. Recently we have undertaken to make a direct measure of this factor, and the value given in Table I is a preliminary result of that work.¹³ Unfortunately such a direct determination poses several difficulties, and the uncertainty in this factor remains the largest source of error in the present

TABLE I. Measured parameters for the determination of ${}^{10}\text{Be}$ half-life as given in Eq. (3).

Run No.	Count rate ^a (${}^{10}\text{Be}$)	$(dN/dt)_{10\text{Be}}$ ^b (10^{-3} dis/sec)	$(dN/dt)_{7\text{Be}}$ ^c (10^6 dis/sec)	$\frac{H({}^{10}\text{Be})}{H({}^7\text{Be})}$ ^d	γ^3	$t_{1/2}({}^{10}\text{Be})$ (10^6 yr)
1 (150 MeV)	0.15 ± 0.01	8.6 ± 0.8	1.02 ± 0.05	0.070 ± 0.007	1.3 ± 0.2	1.6 ± 0.3
2 (600 MeV)	0.52 ± 0.03	29 ± 3	1.06 ± 0.05	0.21 ± 0.02	1.3 ± 0.2	1.5 ± 0.3

^aNet count rate per minute; background subtracted as indicated in text.

^bCalculated disintegration rate per second with efficiency of β counter of 0.30 ± 0.02 (Ref. 10).

^cDisintegration rate per second corrected to time of irradiation using branching ratio of 10.3% and half-life of 53.5 days.

^dCorrected to time of irradiation.

half-life measurement.

The agreement in the half-life determined by the two experiments is very good, despite the significant difference in the $^{10}\text{Be}/^7\text{Be}$ ratio at the two energies. This can be taken as indirect evidence against systematic contamination by some other long-lived species. For the present we adopt a half-life value of $(1.5 \pm 0.3) \times 10^6$ yr. Using samples with a higher specific activity along with an improved measure of γ we hope to be able to increase the precision of this number in the future.

Accepting the half-life for ^{10}Be given here, there are a number of interesting implications, a few of which we discuss briefly below.

First it is obvious that any cross section which has been based on ^{10}Be activity measurements will be reduced by almost a factor of 2. This includes, of course, our own recent measurements for the production of ^{10}Be from a ^{11}B target.¹⁰ The corrected cross section for $^{11}\text{B}(p, 2p)^{10}\text{Be}$ will thus be 11 ± 4 and 14 ± 5 mb at 150 and 600 MeV, respectively. While these values are significantly lower than have been estimated,¹ they are still within the range expected on the basis of other $(p, 2p)$ cross sections.¹⁴

The modification of cross sections may help clear up a long-standing discrepancy between fragmentation cross sections for ^{10}Be production from ^{16}O targets as measured by mass spectrometry,¹² compared with those inferred by radiochemical measurements.¹⁵ As we have pointed out elsewhere,¹⁶ cosmic-ray workers have generally adopted the directly measured mass-spectrometer values¹ while those doing meteorite¹⁷ and lunar sample¹⁸ studies have preferred the larger radiochemical values. The present change in ^{10}Be half-life will tend to bring the radiochemical values more in line with the mass-spectrometric results.

Similar comments will, of course, apply to other measured ^{10}Be cross sections, such as those in iron and nitrogen,¹⁵ which are the most important sources for production in iron meteorites and the atmosphere, respectively. It is interesting to note that even the previously accepted uncertainty in the ^{10}Be half-life has seldom been included when giving radiochemical cross-section errors.

The ^{10}Be activity in meteorites is generally assumed to be in secular equilibrium, and thus the decay rate is equal to the production rate, independent of the half-life. There have been a few attempts to calculate such production rates,

starting from basic nuclear cross-section data.^{18,19} Although the calculations necessarily rely on a number of simplifying approximations, the agreement with observed activities in the case of ^{10}Be is relatively satisfactory. It would therefore appear that use of significantly smaller cross sections would alter that agreement. However, as has been emphasized elsewhere,¹⁶ such a conclusion is dependent on the assumption that the effects of secondary reactions have been correctly taken into account. For high energies the cross sections for neutron-induced reactions are generally taken as equal to those for protons.^{18,19} While often a quite satisfactory assumption, we would like to point out that a more critical examination of the nuclear physics involved in the case of an O target suggests that the major source of ^{10}Be will arise from secondary high-energy neutrons via $^{16}\text{O}(n, 4p3n)^{10}\text{Be}$, which is statistically more favorable than the corresponding reaction $^{16}\text{O}(p, 5p2n)^{10}\text{Be}$. Such an explanation would also be consistent with the otherwise puzzling observation that stone meteorites generally have a ^{10}Be specific activity an order of magnitude greater than irons, despite the fact that for the primary proton spectrum the production in iron is probably larger than in oxygen.¹⁶

Another consequence arises in those cases in which ^{10}Be has been used, in conjunction with another cosmogenic product, to infer exposure or terrestrial ages.²⁰ In cases of short radiation ages, or very long terrestrial ages, the revised half-life will have a direct bearing on the calculated result. Similar remarks will apply to lunar samples, where material can be turned over in times comparable to the ^{10}Be half-life.

Observed ^{10}Be activity in ocean sediments has also been used to calculate sedimentation rates over extended periods of time.²¹ The chronology is again, of course, directly dependent on the ^{10}Be half-life, as well as the assumed nitrogen production cross section mentioned earlier (the neutron-induced reaction probably being the dominant process in this case also).

Because it was the first, and is still one of the few reliably identified unique, second-forbidden β transitions, the decay rate of ^{10}Be is also interesting from a theoretical point of view. Warburton, Garvey, and Towner²² have made transition-matrix calculations for several such nuclides, using both shell and collective models. Although the uncertainties are fairly large, their tentative evidence for the necessity of allowing for collective suppression effects would seem to

be now removed in the case of ^{10}Be .

Finally we return to the subject which originally interested us in this work. We have recently emphasized that we believe it will be absolutely essential to have separation of individual isotopes in cosmic-ray measurements before ^{10}Be can be reliably used as a cosmic-ray "clock."¹⁰ The present work would seem, if anything, to strengthen that view. Firstly, the reduction in several ^{10}Be cross sections, especially that from ^{11}B as noted earlier, will mean that the production ratio $^{10}\text{Be}/\text{Be}(\text{total})$ in cosmic rays will be even smaller than presently estimated. A revised estimate corresponding to Table II of Ref. 10, for example, leads us to predict $^{10}\text{Be}/\text{Be}(\text{total})$ without decay will be 0.15 ± 0.03 .

Also, if one accepts nuclear fragmentation reactions in the interstellar medium as the source of the light elements Li, Be, and B in cosmic rays, the lower limit on their age can be estimated as roughly 3×10^6 yr. Thus with the shorter half-life given here it seems probable that at least a portion of the ^{10}Be will have decayed even in the case of confinement of the cosmic rays to the galactic disk.

Allowing for the uncertainty involved in both cross-section data and cosmic-ray observations, it would seem that the possibility of distinguishing between "young" cosmic rays and those in which all the ^{10}Be has decayed will remain inaccessible by such indirect techniques as measuring Be/B ratios.²³

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¹M. M. Shapiro and R. Silberberg, *Annu. Rev. Nucl.*

Sci. **20**, 323 (1970).

²M. Honda and J. R. Arnold, in *Handbuch der Physik*, edited by K. Sitte (Springer, Berlin, 1967), Vol. 46/2.

³J. Shedlovski, M. Honda, R. Reedy, J. Evans, D. Lal, R. Lindstrom, A. Delany, J. Arnold, H. Loosli, J. Fruchter, and R. Finkel, *Geochim. Cosmochim. Acta* **2**, Suppl. 1, 1503 (1970).

⁴D. Lal and B. Peters, in *Handbuch der Physik*, edited by K. Sitte (Springer, Berlin, 1967), Vol. 46/2.

⁵R. McCorkell, E. L. Fireman, and C. C. Langway, *Science* **158**, 1690 (1967).

⁶T. Lauritsen and F. Ajzenberg-Selove, *Nucl. Phys.* **78**, 104 (1966).

⁷D. J. Hughes, C. Egger, and C. M. Huddleston, *Phys. Rev.* **71**, 269 (1947).

⁸E. M. McMillan, *Phys. Rev.* **72**, 591 (1947).

⁹P. Fontes, C. Perron, J. Lestringuez, F. Yiou, and R. Bernas, *Nucl. Phys.* **165**, 405 (1971).

¹⁰G. Raisbeck and F. Yiou, *Phys. Rev. Lett.* **27**, 875 (1971).

¹¹J. Camplan, R. Meunier, and J. L. Sarrouy, *Nucl. Instrum. Methods* **84**, 37 (1970).

¹²F. Yiou, *Ann. Phys. (Paris)* **3**, 169 (1968).

¹³G. Raisbeck, J. Lestringuez, and F. Yiou, to be published.

¹⁴R. F. Schall, Jr., and A. A. Caretto, Jr., *Phys. Rev. C* **2**, 1924 (1970).

¹⁵M. Honda and D. Lal, *Nucl. Phys.* **51**, 363 (1964); B. S. Amin, S. Biswas, D. Lal, and B. L. K. Somayajula, to be published.

¹⁶G. M. Raisbeck and F. Yiou, *Nature (London), Phys. Sci.* **228**, 73 (1971).

¹⁷P. S. Goel, *Nature (London)* **223**, 1263 (1969).

¹⁸R. C. Reedy and J. R. Arnold, *J. Geophys. Res.* **77**, 537, (1972).

¹⁹J. R. Arnold, M. Honda, and D. Lal, *J. Geophys. Res.* **66**, 3519 (1961).

²⁰C. Chang and H. Wänke, in *Meteorite Research*, edited by P. M. Millman (D. Reidel Publishing Co., Dordrecht, Holland, 1969), p. 397.

²¹B. S. Amin, D. P. Kharkar, and D. Lal, *Deep-Sea Res.* **13**, 805 (1966); Y. Yokoyama, *Nature (London)* **216**, 569 (1967).

²²E. K. Warburton, G. T. Garvey, and I. S. Towner, *Ann. Phys. (New York)* **57**, 174 (1970).

²³F. W. O'Dell, M. M. Shapiro, R. Silberberg, and C. H. Tsao, in *Proceedings of the Twelfth International Conference on Cosmic Rays*, Hobart, Tasmania, Australia, August, 1971 (to be published), Paper OG-59.