

Measurement of the excitation energy of the 7.654 MeV state of ^{12}C and the rate of the 3α reaction*

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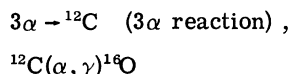
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The excitation energy of the second excited state of ^{12}C has been measured to be 7654.00 ± 0.20 keV. Combining this result with the recent precise measurement of the atomic mass of ^4He by Smith and Wapstra, the Q value for the reaction $3\alpha \rightarrow ^{12}\text{C}(7654.00)$ is determined to be 379.31 ± 0.21 keV. This result implies a change of +9%, at a stellar temperature of 10^8 K, in the 3α reaction rate quoted in the recent compilation of Fowler, Caughlan, and Zimmerman.

NUCLEAR REACTIONS ^{12}C , ^{15}N , $^{16}\text{O}(p, p')$, $E = 34.75$ MeV; magnetic spectrograph; measured excitation energies of $^{12}\text{C}(7.65$ MeV), $^{15}\text{N}(7.56$ MeV), and $^{16}\text{O}(6.13$ MeV) states with 200 eV uncertainties. Measurement related to astrophysical helium burning (3α) rate.

I. INTRODUCTION

In helium burning stars ^4He is converted into ^{12}C and ^{16}O by the successive reactions



with reaction rates $P_{3\alpha}$ and $P_{\alpha^{12}\text{C}}$, respectively.¹ Since the ratio of ^{12}C to ^{16}O at the completion of helium burning depends strongly on the relative values of the two reaction rates, they must be known accurately if one wishes to determine the initial conditions for calculations of subsequent stages of stellar evolution. Such calculations are presently an active research area in astrophysics.

The rate of the 3α reaction depends on the nuclear parameters according to

$$P_{3\alpha} \propto \Gamma_{\text{rad}} e^{-Q/kT}, \quad (1)$$

$$Q = (M_{12}c^2 + E_x) - 3M_{\alpha}c^2, \quad (2)$$

where k is Boltzmann's constant, c is the velocity of light, T is the temperature, M_{12} and M_{α} are the atomic mass excesses of ^{12}C and ^4He , and Γ_{rad} is the radiative width of the 7.65 MeV, $J^{\pi} = 0^+$ state of ^{12}C . The exponential dependence of $P_{3\alpha}$ on the excitation energy E_x of the 7.65 MeV state, coupled with the fact that $kT \approx 10$ keV in helium burning stars means that E_x must be known rather well if one wishes to predict $P_{3\alpha}$ accurately.

The more recent measurements of E_x are all in good agreement,²⁻⁵ the most precise of them having an uncertainty of 1.1 keV, and the weighted average an uncertainty of perhaps 0.8 keV, depending somewhat on the extent to which the various measurements are regarded as uncorrelated. Adding

E_x to $-3M_{\alpha}c^2$, which was known to within ± 0.75 keV,⁶ one obtained Q with an uncertainty of 1.1 keV. This yielded an uncertainty of 14% in $P_{3\alpha}$ at $T = 10^8$ K.

There had been little incentive to measure E_x with more precision because of the limiting uncertainty in M_{α} . A direct measurement of the breakup energy avoids this problem, but it appears that it would be difficult to achieve an accuracy better than the ± 2.0 keV obtained by Barnes and Nichols.⁷ Recently, however, the situation has changed. There has been a measurement of M_{α} with an accuracy of 15 eV⁸ and techniques for the measurement of excitation energies have evolved,⁹ so that it is now possible to determine Q with a standard deviation of 200 eV, essentially eliminating the contribution of the uncertainty in Q to that in $P_{3\alpha}$. The results of such a determination are described in this paper.

II. EXPERIMENTAL METHOD

The excitation energy of the 7.65 MeV state was measured by using a magnetic spectrograph to compare the momentum of protons inelastically scattered from this state with the calibration momenta of protons scattered from states of accurately known mass. Sufficient known lines were recorded *simultaneously* with the unknown to permit the determination from a single exposure of the beam energy, the scattering angle, and the spectrograph calibration parameters.⁹ Such a procedure eliminates many of the uncertainties (such as beam energy and scattering angle variations from run to run) which limit the accuracy of *sequential* calibration-measurement techniques.

A. Calibration lines

A set of calibration lines appropriate for the present measurement has only recently become available.¹⁰ In Ref. 10 the excitation energies of several states in ¹⁵N were determined with increased precision by reanalyzing existing ¹⁴N-(*n*, γ) data.¹¹ This was made possible by an improved ¹⁴N-¹⁵N mass-difference determination,¹² which yielded a value of 10833.64 \pm 0.13 keV for the *Q* value of the (*n*, γ) reaction. The resulting excitation energies for the states in ¹⁵N which were used as calibration lines in the present experiment are listed in the left hand column of Table I.

However, a still more precise value of the ¹⁴N-¹⁵N mass difference was reported by Smith and Wapstra⁸ after the reanalysis of Ref. 10. This more recent mass measurement gives a *Q*-value or separation energy $S_n = 10\,833.395 \pm 0.043$ keV, i.e., a change $\Delta S_n = -0.245$ keV. Using this new determination of S_n it is, in principle, possible to determine the excitation energies E_x of a state in ¹⁵N seen in the (*n*, γ) cascade with an uncertainty in E_x given by:

$$\sigma_{E_x} = (E_x/S_n)\sigma_{S_n},$$

where $\sigma_{S_n} = 43$ eV. In the absence of new (*n*, γ) data or additional information on the previous data it seems that the excitation energies of the states of ¹⁵N should be corrected by an amount

$$\Delta E_x = (E_x/S_n)\Delta S_n.$$

Near $E_x = 7.6$ MeV this represents a change of -170 eV, which is barely significant in the present experiment. There is probably not sufficient

information, however, to justify decreasing the contribution of the uncertainty in S_n to the overall error and thus decreasing the uncertainties given in Ref. 10. Hence, the excitation energies, corrected in this manner, are given in the right hand column of Table I with the uncertainties unchanged.

Also listed in Table I is a recent determination¹³ of E_x for the 6.13 MeV state of ¹⁶O which has an uncertainty of 43 eV. The energy scale of this measurement was also based on the mass measurements of Smith and Wapstra.⁸ This very precise value provides a useful check of the present experiment, as is discussed below.

It is important to note that the excitation energies listed in Table I used as standards in the present experiment are tied directly to the mass measurements of Smith and Wapstra,⁸ which are also the source of the new more accurate mass measurements of ⁴He. Hence, any future change in the conversion factor between the energy scales (in MeV) and mass scales (in amu) will not affect the current value of the mass difference *Q* from Eq. (1) if it is expressed in atomic mass units. The value of E_x for the 7.65 MeV level determined in the present experiment does depend directly on the conversion factor used here¹⁴:

$$1u = 931\,501.6 \pm 2.6 \text{ keV.}$$

B. Experimental procedures

A Uracil (C₄H₄N₂O₂) target 50 $\mu\text{g}/\text{cm}^2$ in thickness, and enriched to 95% in ¹⁵N, was made by vacuum evaporation of Uracil onto a 5 $\mu\text{g}/\text{cm}^2$ carbon foil reinforced by a single layer of Formvar. The targets were bombarded with 34.75 \pm 0.01 MeV protons from the Michigan State University sector focused cyclotron. The reaction products were momentum analyzed in an Enge split-pole magnetic spectrometer with a circular acceptance aperture 0.5° in diameter and were detected in 51 cm long Ilford L4 nuclear track plates with emulsion layers 25 μm thick. Stainless steel absorbers, 0.25 mm thick were used to stop most reaction products other than protons. Dispersion matching¹⁵ and kinematic compensation techniques were used to obtain resolutions of typically 5 keV full width at half-maximum. Data were taken at scattering angles of 12°, 13°, and 14°. Following a run at a particular angle, the magnetic field was changed substantially so the particle trajectories sampled a somewhat different region of the spectrograph field and fell on a somewhat different region of the plate.

The nuclear emulsions were scanned by a human scanner using a computer linked microscope with

TABLE I. Excitation energies of calibration lines.

Nucleus	E_x (keV)	E_x corrected ^a (keV)
¹⁵ N ^b	5270.35 \pm 0.10	5270.23 \pm 0.10
¹⁵ N	5299.16 \pm 0.12	5299.04 \pm 0.12
¹⁵ N	6323.85 \pm 0.12	6323.71 \pm 0.12
¹⁵ N	7155.36 \pm 0.11	7155.20 \pm 0.11
¹⁵ N	7301.09 \pm 0.17	7300.92 \pm 0.17
¹⁵ N	8312.79 \pm 0.14	8312.60 \pm 0.14
¹⁶ O ^c	6130.430 \pm 0.043	

^a These energies were obtained from the results of Greenwood and Helmer as described in the text.

^b All ¹⁵N energies from Greenwood and Helmer, Ref. 10.

^c Reference 13, using Smith's mass doublet as a reference.

a stepping motor driven stage. The stage permitted position measurements with an rms uncertainty of about $2.5 \mu\text{m}$. The spectra were contained in bands 3 mm tall on the plates, which were each scanned in strips 0.5 mm tall in steps of $50 \mu\text{m}$. Six such strips were added to obtain the final spectrum for each of the three scattering angles. The plates were also scanned independently in steps of $100 \mu\text{m}$ and strips 1 mm tall to check the reproducibility of the scanning procedure. An expanded portion of one of these spectra is shown in Fig. 1.

Centroids of the calibration and unknown lines were extracted by fitting the peaks with asymmetric Gaussian shapes. The summed spectra in $50 \mu\text{m}$ steps contained a constant background of about 2 counts per channel in the 5–8 MeV excitation region. This background was subtracted before peak fitting. The centroid of the peak resulting from scattering from hydrogen in the target was determined via a different procedure. This peak shifts very rapidly with angle and a comparison of its position with that of other calibration peaks provides a measurement of the scattering angle θ accurate to 0.01° or better. Although the acceptance aperture was made small to minimize the kinematically induced width of the hydrogen peak, this peak was still $\approx 150 \text{ keV}$ wide and its shape was not well described by a Gaussian. Its position was, for reasons discussed in Sec. IID, taken to be the numerical centroid of the counts in the peak.

C. Fitting procedure and results

In addition to the beam energy and the scattering angle, one must determine the relationship between the location (centroid) D of a peak on the plate and the effective radius of curvature ρ in the spectrograph's magnetic field. To fit a region of the focal plane covering an 8 MeV range of excitation energies (a 26 cm section in this case) the following cubic expansion is adequate:

$$\rho(D) = \rho_0 + \alpha(D - D_0) + \beta(D - D_0)^2 + \gamma(D - D_0)^3 .$$

Here D_0 is the (arbitrary) point from which all distances are measured, $\rho_0 = \rho(D_0)$ and α , β , and γ define the linear, quadratic, and cubic dependence of ρ on the distance from D_0 .

The values of the constants E , θ , ρ_0 , α , β , and γ are determined for each run by a weighted least-squares fit to the calibration lines as discussed in Refs. 9 and 16. Because of the increased accuracy of the present results a detailed discussion of the possible uncertainties of the fitting procedure as well as other possible sources of systematic errors is given in the next section.

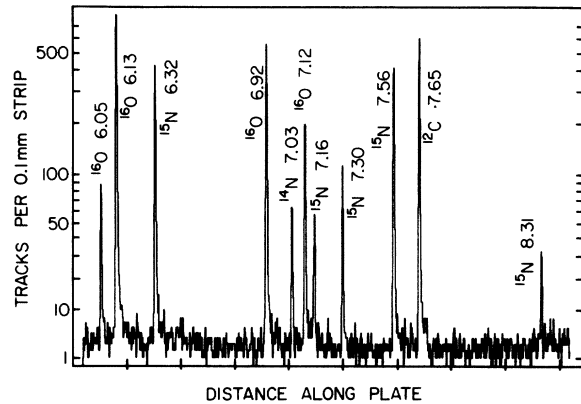


FIG. 1. A portion of a (p, p') spectrum resulting from the 35 MeV proton bombardment of a Uracil target enriched in ^{15}N . The laboratory scattering angle was 14° . The peaks are identified and labeled with their excitation energies in MeV. To improve the readability of the figure, 10 counts have been added to each channel before plotting and the vertical scale adjusted accordingly. Hence, the ordinate is not strictly logarithmic.

In the present analysis the fitting of each spectrum was actually done in steps, the first a fit over the entire 8 MeV of excitation energy to determine the beam energy and scattering angle, and the second over the region from 5.2 to 8.3 MeV to determine the excitation energy of the 7.65 MeV level of ^{12}C . The first fits utilized only the stronger calibration lines and were done on the spectra which were scanned with the $100 \mu\text{m}$ step size. The second fits used only the seven calibration lines listed in Table I with the beam energy and scattering angles fixed at the values determined previously.

The limited region covered by the lines listed in Table I did not require a cubic calibration curve, a quadratic fit yielding an average χ^2 per degree of freedom of 2. The uncertainties associated with the calibration lines were those listed in Table I plus the statistical uncertainties of their centroids. The latter, determined by the experimental resolution ($\sim 5 \text{ keV}$) and the number of counts in the peaks (100–4000), varied from 0.04 to 0.22 keV for the peaks in a given spectrum.

A weighted quadratic least-squares fit using the seven calibration lines, with momentum uncertainties and centroid uncertainties input separately, was made to determine the best value of the excitation energy of the ^{12}C 7.65 MeV state and its associated uncertainty.¹⁷ This procedure utilized the error matrix associated with the fitting parameters ρ_0 , α , and β . The standard deviation found for the excitation energy extracted in this way was about 150 eV for each spectrum fit, with roughly equal contributions from the statistical

TABLE II. Results of the separate runs.

	Run 1	Run 2	Run 3	Average
Scattering angle (deg)	12.19	13.16	14.11	
χ^2 of fit	2.4	0.4	3.1	2.0
$E_x(7.65)$	7653.94	7654.20	7653.87	7654.00 keV
Standard deviation				0.13 keV ^a

^a Standard deviation of the mean of the three measured values.

and calibration line uncertainties.

The values for the excitation energy of the 7.65 MeV level of ^{12}C and the χ^2 values of the three fits are listed in Table II. The final value is

$$E_x = 7654.00 \pm 0.20 \text{ keV.}$$

The standard deviation of the mean of the three values is 0.13 keV. The various contributions to the over-all standard deviation of 200 eV are listed in Table III and discussed in the next section.

Using the value of E_x and the mass excess of ^4He ⁸

$$M_\alpha c^2 = 2424.898 \pm 0.016 \text{ keV}$$

the Q value for the $3\alpha \rightarrow ^{12}\text{C}$ reaction [Eq. (2)] is 379.31 ± 0.21 keV.

The state of ^{15}N at 7.56 MeV is the closest one to the 7.65 MeV state of ^{12}C (see Fig. 1). However, it was not seen in the (n, γ) work, and its energy was not well enough known so it could serve as a calibration line. The excitation energy of this state has been determined in the present work to be 7563.68 ± 0.20 keV, compared with the previously reported value of 7566 ± 3 keV.¹⁸ The difference in excitation energy between these two states is determined somewhat better than the separate values:

$$E_x(^{12}\text{C}, 7.65) - E_x(^{15}\text{N}, 7.56) = 90.32 \pm 0.16 \text{ keV.}$$

As an empirical check on the present methods the fitting procedure was also performed without using the ^{16}O 6.13 MeV state as a calibration line. The excitation energy of this state was then found to be 6130.27 ± 0.16 keV in good agreement with the value indicated in Table I. Omitting this line from the calibration caused a shift of only -10 eV in the extracted value for the excitation energy of the ^{12}C 7.65 MeV level.

The values of the excitation energies reported above supercede the recently reported preliminary values,¹⁶ which were higher by 0.3 to 0.4 keV. The change is due to three effects. Firstly, the present analysis used the corrected calibration energies as listed in Table I and discussed above. This lowered the value for the ^{12}C 7.65 state by 170 eV.

An additional 70 eV reduction was due to an erroneous constant in the relativistic kinematics program which was used in the preliminary analysis. The remaining change was caused by detailed changes in the fitting procedure. For example, the bin size was 100 μm throughout the earlier analysis. The present analysis used all seven calibration lines listed in Table I in a weighted least-squares fit, whereas the previous analysis used only four lines from the ^{15}N spectrum in an unweighted fit. The change was consistent with the statistical uncertainties of the fits.

D. Uncertainties

Beam Energy. The cyclotron beam energy was determined by the momentum matching method described in Refs. 19 and 9. The deuteron line resulting from the $^{12}\text{C}(p, d)$ reaction to the first excited state of ^{11}C was used for this purpose. The well known Q value for the reaction leading to this state,^{8,9} and the fact that it was conveniently located in the (p, p') spectrum, permitted a determination of the beam energy with an uncertainty of less than ± 10 keV.

The effect of this uncertainty on the excitation energy determination was determined by carrying out the calibration procedure with the beam energy fixed at different values. A beam energy uncertainty of 10 keV was found to contribute an uncertainty of 8 eV to the extracted excitation energy.

Scattering angle. The position of the peak re-

TABLE III. Contributions to the uncertainty in the excitation energy measurement.

Source of uncertainty	Contribution to uncertainty (eV)
Beam energy (± 10 keV)	8
Scattering angle ($\pm 0.01^\circ$)	40
Angular distribution effects	20
Centroid determination	144
Calibration lines	120
Target thickness effects	50
Total standard deviation of the mean	199 eV

sulting from proton scattering on hydrogen in the target was used to determine the scattering angle. At $\theta = 13^\circ$ the lines from (p, p') on ^{12}C shift kinematically 4 keV per degree faster than those from ^{15}N , so that a scattering angle uncertainty of 0.01° contributes 40 eV uncertainty to the excitation energy determination of a line in ^{12}C relative to calibration lines in ^{15}N .

It is important to know the cross section variation of the hydrogen scattering across the angular range (0.5°) of the spectrograph aperture, since a rapid variation could bias the centroid and, thereby, cause an erroneous angle determination. Experimental measurements at 31.8²⁰ and 39.4 MeV²¹ show that the laboratory cross section variation at scattering angles between 12° and 14° is 4% per degree or less, and, therefore, produces a negligible effect in this case.

Since the cross section variation is too small to produce the observed peak asymmetry, it must be due to small variations in the angle of incidence of the beam on the target during the run, and/or nonuniform illumination within the angular divergence of the incident beam. In this case it is the numerical centroid of the counts within the peak which properly determines the average scattering angle of the exposure. The statistical significance of the centroids was adequate to determine the scattering angles with uncertainties of 0.01° .

Angular distribution effects. Yield variations across the spectrograph aperture can also be relevant for the other lines in the spectra. From the measured yields an upper limit can be placed on the centroid shifts due to this effect. The cross section for the ^{12}C 7.65 MeV state varied the most rapidly, its yield decreasing by about 10% per 0.5° over the angular range covered. This much variation can cause a maximum centroid shift of < 20 eV for a ^{12}C line relative to a ^{15}N line, the exact magnitude and sign of the shift depending on the focal plane setting for kinematic compensation.

Centroid determination. The accuracy of the centroid determinations is limited mainly by two factors: the statistical uncertainty due to the finite resolution and number of counts in each peak, and small fluctuations associated with the accuracy of the microscope lead screw. The statistical uncertainties from the peak fitting, assuming perfect peak shapes and no background, vary from 40 to 220 eV. Using these numbers in the calibration routine yields for each run a contribution of about 100 eV to the uncertainty of the energy of the ^{12}C 7.65 MeV level. Using these numbers in combination with the calibration line errors yielded the average $\chi^2 = 2$ mentioned above. This uncertainty should be increased in practice,

however, to account for such effects as finite scanning-bin size and nonideal peak shapes.

The microscope lead screw has fine scale deviations from linearity which have an rms value of $2.5 \mu\text{m}$ or about 80 eV in the present work. Combining this effect with a realistic estimate of the other uncertainties gives an over-all contribution to the excitation energy uncertainty of about 250 eV per run or 144 eV after averaging over the three independent runs.

A measure of whether this is a reasonable estimate is the actual sample standard deviation, 224 eV, of the measured values listed in Table II. This indicates that the actual experimental reproducibility is consistent with the above estimates of the statistical uncertainties, and that there are no fine scale fluctuations in the magnet calibration curve which are significant at this level of accuracy. All other lines within the region of interest in these spectra show similar reproducibility.

Calibration lines. The calibration line uncertainties of from 40–170 eV listed in Table I would contribute an uncertainty of 100 eV to the excitation energy determination if these errors were entirely uncorrelated.¹⁷ We have increased this uncertainty slightly, to 120 eV, to allow for partial correlations in the errors.

Target thickness effects. Since the present measurements involve only a comparison of the relative positions of like particles in the spectra, the only target thickness effect is due to differential energy losses. An upper limit on the magnitude of this effect can be obtained by assuming the ^{15}N and ^{12}C are on opposite sides of the target, separated by $20 \mu\text{g}/\text{cm}^2$ of another substance. This gives a differential energy loss between incoming 35 MeV protons and outgoing 28 MeV protons of about 50 eV. (In the beam energy determination the position of a deuteron group was measured, but in this case the target thickness effect was included explicitly.)

The various contributions to the over-all uncertainty are summarized in Table III. Added in quadrature, they total 199 eV.

III. SUMMARY AND DISCUSSION

The value of E_x obtained here is consistent, within about one standard deviation or better, with all of the recent measurements (see Table IV) and is 1.05 keV, a little over one standard deviation, lower than the weighted average of the earlier results. Because the present result is much more precise than any of the earlier measurements it dominates the weighted average of all the data. The value of Q is obtained from Eq. (2)

TABLE IV. Summary of measurements of E_x and Q . Prior work is listed if the error is less than 3 keV.

Authors	Reaction	E_x^a (keV)	Q^b (keV)
Austin, Trentelman, and Kashy (Ref. 2)	$^{12}\text{C}(p, p')^{12}\text{C}^*$	7656.2 ± 2.1	381.5 ± 2.1
Stocker, Rollefson, and Browne (Ref. 3)	$^{12}\text{C}(p, p')^{12}\text{C}^*$	7655.9 ± 2.5	381.2 ± 2.5
McCaslin, Mann, and Kavanagh (Ref. 4)	$^{15}\text{N}(p, \alpha)^{12}\text{C}^*$	7654.2 ± 1.6	379.5 ± 1.6
Barnes and Nichols (Ref. 7)	$^{12}\text{C}^* \rightarrow 3\alpha$	7654.3 ± 2.0	379.6 ± 2.0
Jolivet <i>et al.</i> (Ref. 5)	$^{12}\text{C}(p, p')^{12}\text{C}^*$	7655.2 ± 1.1	380.5 ± 1.1
Weighted average of prior results ^c		7655.05 ± 0.73	380.35 ± 0.73
Present result		7654.00 ± 0.20	379.31 ± 0.21
Weighted average		7654.07 ± 0.19	379.38 ± 0.20

^a Obtained from direct measurements of E_x , except in the case of Barnes and Nichols, where E_x was obtained from the measured value of Q using Eq. (1) and $3M_\alpha = 7274.69 \pm 0.05$ (Ref. 8).

^b Except for the case of Barnes and Nichols where Q is measured directly, obtained from E_x using Eq. (1) and the value of M_α given in footnote a.

^c Data have been weighted inversely as the square of the quoted standard deviation.

using the weighted mean of Table IV and the value of M_α of Smith and Wapstra⁶ to yield

$$Q = 379.38 \pm 0.20 \text{ keV.} \quad (3)$$

One can write Eq. (1) in the form²²

$$P_{3\alpha} \propto \exp(-Q/86.171 T_9) = \exp(-4.4026/T_9), \quad (4)$$

where Q is given in keV and $T_9 = 10^{-9}T(\text{K})$. In their recent compilation Fowler, Caughlan, and Zimmerman (FCZII)²² obtained $P_{3\alpha} \propto \exp(-4.4109/T_9)$. Thus, the present results imply that the tabulated rates of FCZII should be multiplied by a factor

$$P_{3\alpha} = P_{3\alpha}(\text{FCZII}) \times \exp(0.0083/T_9)$$

to conform to the result of Eq. (3). This corresponds to an increase in the 3α reaction rate of about 9% at 10^8 K. (Had FCZII included the relatively high values of Q from Ref. 5, the percent correction would have been somewhat greater.) The uncertainty in $P_{3\alpha}$ arising from the uncertainty in Q is now less than 2.5% at 10^8 K, en-

tirely negligible compared to the 30% error in Γ_{rad} . (See Ref. 23 for a review of the situation with regard to Γ_{rad} .)

Recently Dyer and Barnes²⁴ have used the reaction rates they had obtained for the $^{12}\text{C}(\alpha, \gamma)^{16}\text{O}$ reaction and the results of calculations by Arnett²⁵ to establish that ^{12}C is likely to be the dominant product of helium burning, at least in stars lighter than about 15 solar masses. The results of the present experiment establish that the $P_{3\alpha}$ reaction is somewhat faster than had been thought, leading to still greater ^{12}C production and, hence, qualitatively strengthening this conclusion.

IV. ACKNOWLEDGMENTS

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¹See D. D. Clayton, *Principles of Stellar Evolution and Nucleosynthesis* (McGraw-Hill, New York, 1968) and references therein.

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