Nuclear Pair Emission from the 7.656-Mev Level in C^{12}†

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The 7.656-Mev nuclear pair transition from the 0+ second excited state of C^{12} has been observed in the $Be^9(\alpha,n)C^{12}$ reaction by means of an intermediate-image pair spectrometer. With a beam of 5.81-Mev alpha particles incident on a 0.7-Mev thick Be foil target the observed intensity ratio of the 7.656-Mev pair line to the 4.433-Mev pair line from the 2+ first excited state of C^{12} was $(5\pm1.5)\times10^{-4}$. Approximately the same intensity ratio was found with both 5.38- and 5.81-Mev alpha particles incident on thick (6 mg/cm^2) Be targets. By applying the appropriate factors for the spectrometer efficiency and for the internal pair conversion coefficient of the 4.433-Mev transition the derived ratio of pair to total widths of the 7.656-Mev level is $\Gamma_{e\pm}/\Gamma=8.2\times10^{-7}\times R$ where $R=N_{4.433}/N_{7.656}$, the ratio of neutron populations in the $Be^9(\alpha,n)C^{12}$ reaction. As a rough estimate R is assumed to be ~8 based on the only information available. This leads to $\Gamma_{e\pm}/\Gamma\sim7\times10^{-6}$ which is a factor of ~15 smaller than estimates by Cook et al. in which the width Γ_α for the alpha-particle decay of the level was taken as $\frac{1}{10}$ of the Wigner limit. The most plausible explanation of the data is that Γ_α is close to the Wigner limit.

INTRODUCTION

A BRIEF report[1] has been given on the observation of the 7.656-Mev nuclear pair transition from the second excited state of C^{12}. The existence of this state was predicted by Hoyle[2] after it had been proposed by Öpik[3] that energy generation and element synthesis in red giant stars result from a 3 $He^4\rightarrow C^{12}$ fusion process. In their late stages of evolution the cores of these stars are believed to consist largely of helium. Under the conditions of temperature and density thought to exist in the core helium can combine into unstable Be^8 with an equilibrium ratio of Be^8 to He^4 of $\sim10^{-9}$. Hoyle suggested that in order to explain the reaction rates and the relative isotopic abundances the absorption of an alpha particle by Be^8 is a resonance reaction which occurs at an energy of ~0.33 Mev corresponding to a level in C^{12} at ~7.7 Mev. Helium burning occurs if such a state decays partially to the ground state of C^{12} rather than breaking up by alpha-particle emission. Subsequent investigations making use of a variety of nuclear reactions established the presence of a state in C^{12} whose energy is 7.656 ± 0.007 Mev[4] above the ground state. Cook et al.[5] summarized the experimental evidence and presented arguments for the very probable spin-parity assignment of 0+ to this level. They also observed the alpha-particle spectrum from the 7.656-Mev state occurring in the beta decay of B^{12} thus proving, according to the reversibility of nuclear reactions, that the level can be formed in the manner proposed by Hoyle.

The question of the various possible modes of decay of the 7.656-Mev state has also been considered by Cook et al. and the thermonuclear reaction rates of the helium-fusion process have been calculated by Salpeter.[6] Aside from the alpha-particle decay of the 7.656-Mev level to Be^8 there are two possible paths to the ground state of C^{12}, namely the emission of a 3.2-Mev $E2$ gamma ray to the 2+ first excited state followed by a (4.433 ± 0.005)-Mev gamma ray, and a direct transition to the 0+ ground state which would be an electric monopole transition if the 7.656-Mev state is indeed 0+. Such a transition would proceed almost entirely by the emission of positron-electron nuclear pairs. On the basis of a 0+ spin-parity assignment Cook et al. suggested the following partial widths for the decay of the 7.656-Mev state:

$$\Gamma_\alpha\sim0.5 \text{ ev},$$
$$\Gamma_{3.2\gamma}\sim0.0014 \text{ ev},$$
$$\Gamma_{e\pm}\sim5\times10^{-5} \text{ ev}.$$

Γ_α is based on the "reasonable estimate" that the dimensionless reduced width for alpha-particle emission is $\sim\frac{1}{10}$ of the Wigner limit. $\Gamma_{3.2\gamma}$ is a single-particle estimate and $\Gamma_{e\pm}$ is calculated from a measurement[7] of the cross section for inelastic scattering of electrons by C^{12}.

Many attempts have been made to detect the 3.2-Mev gamma ray and the 7.656-Mev nuclear pair transition from this level. In all cases when one of these transitions has been reported other experimenters have failed subsequently to confirm the previous results. The pertinent references are summarized by Ajzenberg-Selove and Lauritzen[4] and we mention below only the most sensitive of the searches for these transitions.

In the beta decay of B^{12} Kavanagh[8] has shown by gamma-gamma coincidence studies using NaI scintillation counters that the $\gamma_{3.2}/\gamma_{4.4}$ intensity ratio is $<0.1\%$. Since the 7.656 and 4.433-Mev levels are fed

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8 R. W. Kavanagh, Bull. Am. Phys. Soc. 3, 316 (1958).

TABLE I. Theoretical estimates and previous best experimental limits on the ratios of widths for the 7.656-Mev level in C^{12}.

	Theoretical estimate[a]	Exp. result	Exp. method
$\Gamma_{3.2\gamma}/\Gamma$	2.8×10^{-3}	$<10^{-3}$	B^{12} decay-scint. spec.[b]
		$<10^{-3}$	$C^{12}(\alpha,\alpha')C^{12*}$-recoil[c]
$\Gamma_{e\pm}/\Gamma$	10^{-4}	$<1.7\times10^{-5}$	$Be^9(\alpha,n)C^{12}$-scint. spec.[d]
		$<2.6\times10^{-5}$	$Be^9(\alpha,n)C^{12}$-mag. pair spec.[e]

[a] See reference 5.
[b] See reference 8.
[c] See reference 12.
[d] See reference 9.
[e] See references 10 and 11.

by beta-ray branches of almost equal intensity (about 1.4%) this limit corresponds to $\Gamma_{3.2\gamma}/\Gamma<10^{-3}$, where Γ is the total width of the 7.656-Mev level.

Both scintillation and magnetic spectrometers have been used to search for the 7.656-Mev nuclear pair transition. Goldring et al.[9] have shown by means of a G.M. and NaI coincidence counter arrangement that the number of 7.656-Mev positron-electron pairs is $<1/600$ of the 4.433-Mev pairs in the $Be^9(\alpha,n)C^{12}$ reaction at $E_\alpha=5.3$ Mev on a thick Be target. Under the assumption that the ratio of neutron populations $R=N_{4.433}/N_{7.656}$ is ~8 their result, taking into account the internal pair conversion coefficient of the 4.433-Mev transition, leads to $\Gamma_{e\pm}/\Gamma<1.7\times10^{-5}$. Bent et al.[10] employed this same reaction at $E_\alpha=4.3$ Mev together with an intermediate-image magnetic pair spectrometer in a search for the 7.656-Mev pair line. Their experiment set an upper limit of 5% on the ratio of 7.656- to 4.433-Mev pair lines intensities but this has been reduced to an upper limit of 0.2% in unpublished work[11] making use of similar but improved techniques. The latter data were taken at $E_\alpha=4.5$ Mev on a 2.8-mg/cm^2 thick Be target backed by 12 mg/cm^2 of Ag. By using the above mentioned estimate of relative neutron populations and by applying the spectrometer efficiency and internal pair conversion factors, as will be described later for the present work, Bonner's result corresponds to $\Gamma_{e\pm}/\Gamma<2.6\times10^{-5}$.

An indirect method for detecting the presence of gamma-ray transitions from the 7.656-Mev level is to search for C^{12} recoil nuclei corresponding to the emission of gamma radiation. Thus far the most sensitive experiment of this type has been carried out by Eccles and Bodansky[12] using the $C^{12}(\alpha,\alpha')C^{12*}$ reaction. They were able to set a limit on Γ_γ/Γ of 10^{-3} which is the same as Kavanagh's limit on the 3.2-Mev gamma ray discussed above. However, the recoil experiment also places a limit on the emission of 7.656-Mev gamma radiation.

Table I summarizes the limits on the gamma-ray and pair decays of the 7.656-Mev level prior to the present work.

There are several types of nuclear reaction[4] which may be used to excite the 7.656-Mev level in C^{12}. The experimental situations involving most of these reactions suffer from one or more of the following disadvantages: (a) gamma rays from competing reactions, (b) gamma rays from reactions in other isotopes, (c) low total yield due to beam or target limitations, (d) small population of the 7.656-Mev level relative to the population of the 4.433-Mev level, (e) excitation of higher states in C^{12}, (f) contaminants on the target, and (g) contamination of the beam with other components. Even if there were no gamma-ray lines produced in the neighborhood of 7.6 Mev due to any of the foregoing effects a reaction should be chosen for the magnetic pair spectrometer which results in as small a singles counting rate as possible at the 7.656-Mev pair-line position in order to minimize the random coincidence counting rate. This last consideration rules out the use of B^{12}, which decays by beta-ray emission with a 97% branch of $E_{\beta\max}=13.4$ Mev to the ground state of C^{12} and a branch of 1.3% to the 7.656-Mev level, or the use of N^{12}, which decays by positron emission with an 85% branch of $E_{\beta\max}=16.5$ Mev to the ground state of C^{12} and a branch of 2.5% to the 7.656-Mev level.

The $Be^9(\alpha,n)C^{12}$ reaction is known to populate only the ground and first two excited states of C^{12} at alpha-particle energies of up to 5.3 Mev. The ground-state Q of the reaction is[4] 5.7 Mev which implies a threshold of somewhat less than 3 Mev for forming the 7.656-Mev state. At a bombarding energy of 5.3 Mev the yield[13] of the neutron group to the 7.656-Mev level is $\frac{1}{8}$ of that to the 4.433-Mev state as measured in the forward direction with a thin target. No other data on relative neutron populations have been reported. If this ratio were to represent the relative populations integrated over all angles the $Be^9(\alpha,n)C^{12}$ reaction would be at least as favorable as any other in respect to the population of the 7.656-Mev level. No other reaction resulting in the emission of gamma radiation is expected from the bombardment of Be with alpha particles. One likely target contaminant is carbon. Above $E_\alpha=5.05$ Mev the $C^{13}(\alpha,n)O^{16}$ reaction is known to excite the 6.05-Mev first excited state of O^{16} but this is below the energy of the C^{12} level in question and the associated background under the 7.656-Mev line should be negligible. A substantial reaction rate can be expected in the bombardment of Be with alpha particles because the cross section for the $Be^9(\alpha,n)C^{12}$ reaction is 0.4b at $E_\alpha=5.3$ Mev and because Be foil targets can withstand high beam currents. The energetic neutrons from this reactions, which cause serious background problems in scintillation counter measurements, should not be a major difficulty in a magnetic spectrometer measure-

[9] G. Goldring, Y. Wolfson and R. Wiener, Phys. Rev. **107**, 1667 (1957).
[10] R. D. Bent, T. W. Bonner, J. H. McCrary, and W. A. Ranken, Phys. Rev. **100**, 771 (1955).
[11] T. W. Bonner (private communication).
[12] S. F. Eccles and D. Bodansky, Phys. Rev. **113**, 608 (1959).
[13] W. H. Guier, H. W. Bertini, and J. H. Roberts, Phys. Rev. **85**, 426 (1952).

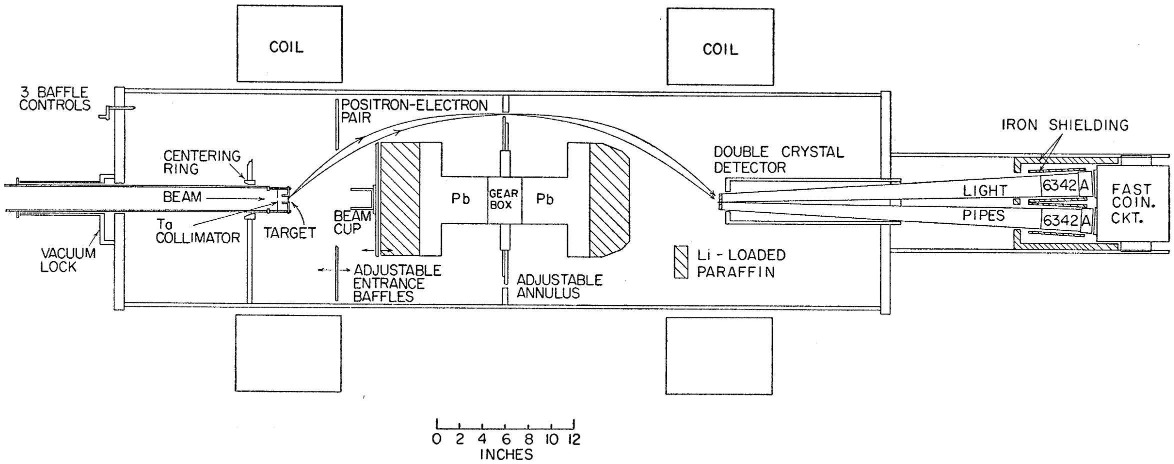

FIG. 1. Intermediate-image pair spectrometer showing the locations of Li-loaded paraffin neutron shields.

ment. In view of these considerations the $Be^9(\alpha,n)C^{12}$ reaction was selected as being the most promising one for the 7.656-Mev pair-line search.

EXPERIMENTAL

The intermediate-image pair spectrometer used in this work has been described previously.[14,15] Preliminary evaluation tests on the $Be^9(\alpha,n)C^{12}$ reaction were made with a He^+ beam of up to 3 Mev from the Brookhaven Van de Graaff accelerator. From the spectrometer yield of the 4.433-Mev pair line and the background at 7.656 Mev it was concluded that if a beam of 5- to 6-Mev alpha particles were available the chances were favorable for detecting a 7.656-Mev pair line several times weaker than the upper limit set by Goldring et al.[9]

Through the cooperation of Oak Ridge National Laboratory the search for the 7.656-Mev nuclear pair transition was carried out with their large Van de Graaff accelerator. This High Voltage Engineering Company vertical machine is nominally rated at 5.5 Mev but it will operate well up to 6 Mev. The intermediate-image spectrometer, all of the associated electronics and a 15-kw motor-generator set were moved to Oak Ridge and installed in the Van de Graaff experimental area. It had been determined beforehand at Brookhaven that the current output of the 15-kw motor-generator set was sufficient to reach a momentum of focused electrons just above the expected position of the 7.656-Mev pair line.

Two modifications were made in the spectrometer prior to these investigations. Because of the possible background from the scattering of neutrons into the detector the shielding inside the spectrometer was improved by the installation of two $3\frac{1}{2}$-in. thick by 10-in. diameter lithium-loaded paraffin disks between the target and the detector as shown in Fig. 1. The paraffin shield at the detector end of the instrument is tapered in order to avoid cutting off some of the focused electrons, since at high transmission settings the envelopes of electrons are not symmetrical on both sides of the annulus. The paraffin was found to out-gas at first but a satisfactory vacuum could be achieved after a time. As may be seen from Fig. 1 with the help of a straight-edge neutrons from the target cannot singly-scatter from the vacuum chamber or coils into the detector without going through one or the other of the neutron shields.

As a precaution against the scattering of neutrons from the spectrometer base plate into the detector the region below the vacuum chamber and between the two coil housings was filled with blocks of Li-loaded paraffin (not shown in Fig. 1). Unfortunately there was insufficient time to test the relative background rates in the $Be^9(\alpha,n)C^{12}$ reaction with and without the Li-loaded paraffin disks and the above-mentioned blocks.

A further modification was the substitution of RCA-6342A photomultiplier tubes which were cemented onto the light pipes in place of the type-6342 tubes formerly used. The 6342A tube has a curved photo surface which increases the photoelectron collection efficiency and decreases the transit time spread. All of the pair-line spectra were taken at a coincidence resolving time of $\tau = 1.0\times10^{-9}$ sec since it was found that the coincidence efficiency for the pair line of the 4.433-Mev gamma ray was close to 100% at that time resolution setting.

The position chosen for the spectrometer in the Oak Ridge Van de Graaff experimental area was 25 feet from the 90° deflecting magnet on the opposite side of a 3-foot thick water-filled shielding wall having a hole for the beam pipe to pass through. A special rf ion-source tube, used only when helium is accelerated so as to minimize proton or deuteron contamination of the beam, was installed in the terminal of the machine at the beginning of these experiments. Singly-ionized helium was accelerated and then gas-stripped to He^{++} so that the deflecting magnet could bend the beam through 90°

[14] D. E. Alburger, Rev. Sci. Instr. **27**, 991 (1956).
[15] D. E. Alburger, Phys. Rev. **111**, 1586 (1958).

into a horizontal beam pipe. The maximum gas-stripping efficiency was $\sim 75\%$. An electrostatic strong-focusing lens located 10 feet from the target made it possible to put ~ 10 μamp of He^{++} through a 4.5-mm diameter tantalum defining aperture just in front of the target inside the spectrometer. In order to illuminate the target more uniformly the beam was actually defocused by adjusting the voltage on the lens so that the current through the defining aperture was reduced to one-half of its intensity at the best focus condition. The current intensity on the target was then controlled by varying the rate of gas flow in the stripper tube so as to change the stripping efficiency.

The target materials used in this work consisted of Be foils 1.3 mils (6.0 mg/cm^2) thick and 0.1 mil thick. R. Benenson of Columbia University kindly supplied the 0.1-mil Be foils which had been made by H. Bradner at the University of California in Berkeley. 6-mg/cm^2 Be is infinitely thick for 6-Mev alpha particles whereas the energy loss in 0.1-mil Be is ~ 350 kev. Two pieces of the 0.1-mil Be were clamped together in the water-cooled target holder for the "thin"-target runs.

Because of the difficulties in being sure of the beam current reading owing to the effects of secondary electron emission from the target or the beam cup the number of target reactions was monitored by detecting neutrons in a BF_3 long counter located 12 feet from the target. The monitor rate in the thick target runs was a few hundred counts per second.

All of the data on pair spectra were taken at the highest transmission setting of the instrument (annulus width 17 mm) where the pair-line resolution for a point source in the absence of Doppler broadening is 2.5% and the absolute spectrometer transmission for $E0$ transitions is 1 count per 10^4 pairs. Before taking each point the pulse-height analyzers were adjusted so as

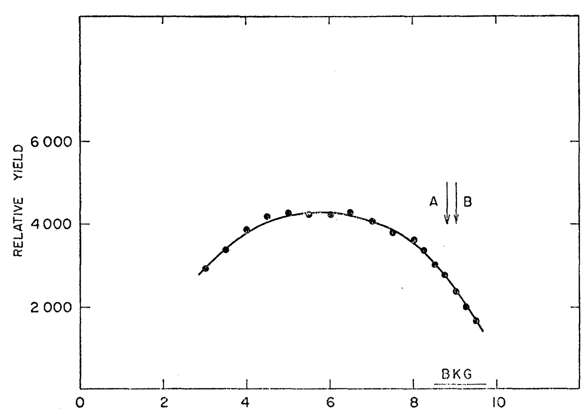

FIG. 2. Spectrum of electrons from a 6-mg/cm^2 Be target bombarded with 3-Mev alpha particles as detected in one of the two crystals of the intermediate-image pair spectrometer. Arrow A is at the expected momentum position of the 7.656-Mev pair line and B is at the expected end point of the continuum of the 4.433-Mev positron-electron internal pairs.

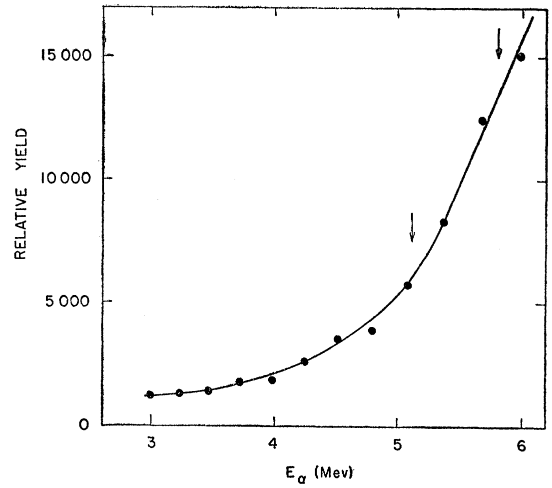

FIG. 3. Intensity of the 4.433-Mev internal pair conversion line from the $Be^9(\alpha,n)C^{12}$ reaction, using a 6-mg/cm^2 thick Be target, versus alpha-particle energy. The arrows indicate the entrance and exit energies corresponding to the thin-target run of Fig. 5.

to include only the full-energy-loss peak whose position varies according to the electron energy being focused. (See reference 15, Fig. 3, for a plot of the pulse-height spectrum from one of the crystals.)

EXPERIMENTAL RESULTS

The spectrum of electrons from a 6-mg/cm^2 thick Be target bombarded with a 3-Mev He^{++} beam is shown in Fig. 2. Data were taken up to the maximum safe current output of the motor-generator set ($\sim 20\%$ current overload). The arrow marked A indicates the expected momentum position of the 7.656-Mev pair line while the one marked B is the expected endpoint of the positron-electron pair continuum associated with the 4.433-Mev transition. Most of the yield can be ascribed to Compton electrons produced by 4.433-Mev gamma rays since the extrapolation of the spectrum agrees with the expected Compton end-point within 100 kev. Since the Compton electrons are presumably produced in the target material the data suggest that in order to keep the singles rate in thick-target pair spectrum runs as low as possible the target should be held to a minimum thickness consistent with the range of the alpha particles. 6-Mev alpha particles have a range of 5 mg/cm^2 in Be.

A run was then made, as shown in Fig. 3, on the yield of the 4.433-Mev pair line as a function of the energy of alpha particles incident on the 6-mg/cm^2 Be target. Although there is some uncertainty about the shape of this curve, owing to the previously mentioned difficulty in obtaining a reliable current reading, the curve indicates that the yield at 6 Mev is 3 times greater than at 5 Mev and 12 times greater than at 3 Mev. The arrows in Fig. 3 are drawn at entrance and exit energies of 5.81 and 5.1 Mev, respectively, of alpha particles incident on the "thin" target.

Four runs were made on the pair-line spectrum occurring in the $Be^9(\alpha,n)C^{12}$ reaction. The first was carried out at $E_\alpha=5.38$ Mev on a 6-mg/cm² thick Be target and it gave positive evidence for the existence of the 7.656-Mev pair line. Using 6-mg/cm² thick Be targets at $E_\alpha=5.81$ two runs were made, one of which is shown in Fig. 4. It is the average of the total coincidence counting rates taken during a 15-hour period. The beam intensity was held close to 4 μamp of He^{++} (~12 watts dissipated in the target) and the number of pair counts was recorded for intervals of 500×64 neutron monitor counts.

At the peak of the 4.433-Mev line in Fig. 4 the counting rate was 4000 per min. The 3.3% width of this line may be compared with a calculated window width of 2.9% based on the point-source, non-Doppler-broadened width of 2.5% folded together with the 1.4% width resulting from the 4.5-mm diameter of the target spot. When the beam energy was increased from $E_\alpha=3$ Mev to $E_\alpha=5.81$ Mev the 4.433-Mev peak position shifted upward in momentum by approximately 0.5%. Both the peak shift and the broadening of line are ascribed to the Doppler effect. The calculation of the expected position of the 7.656-Mev line was made by using the 4.433-Mev line position for calibration but making no Doppler-shift corrections, inasmuch as the 7.656-Mev line is also Doppler-shifted. Although this procedure is not quite exact the error in the predicted position of the 7.656-Mev line is much smaller than the line width.

Above the 4.433-Mev line in Fig. 4 the sloping background results largely from random coincidences. At a current setting of 7.5 the random rate, when measured with a long delay line in one side of the coincidence circuit, was $\frac{3}{4}$ as large as the corresponding point on the curve. The shape of the sloping coincidence background is consistent with the shape of the singles curve shown shown in Fig. 2. However, the calculated random rate was only half as great as the measured rate. A measured random rate larger than the calculated rate could occur either through a malfunction of the coincidence circuit

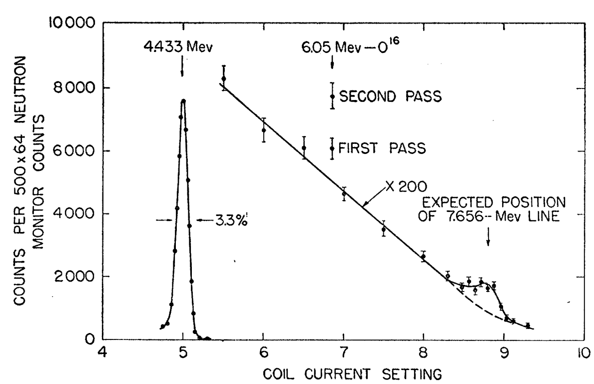

FIG. 4. Pair-line spectrum from a 6-mg/cm² thick Be target bombarded with 5.81-Mev alpha particles.

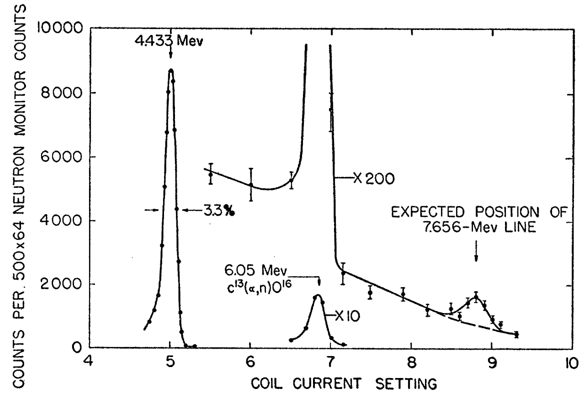

FIG. 5. Pair-line spectrum from a 0.2-mil thick (0.7-Mev thick for alpha particles) Be target bombarded with 5.81-Mev alpha particles.

or because of fluctuations in beam intensity which result in *effective* singles counting rates that are higher than the measured average rates. The beam spot when observed at low currents on a quartz viewer exhibited a considerable amount of rapid jitter in intensity which may have been responsible for this effect. Another possibility is that there may be an rf intensity modulation of the beam because of the type of ion source used in this machine. Neither of these possibilities was investigated because of time limitations, but it would be of general interest to understand the factor of 2 discrepancy (if it is real) between the calculated and measured random rates for the sake of other coincidence experiments to be carried out using the beam from this accelerator.

Near the high-momentum end of Fig. 4 there is a bump at the expected position of the 7.656-Mev line. When the dashed extrapolated background is subtracted the net peak has approximately the same shape and percentage width as the 4.433-Mev line and the peak position agrees with the calculated position with an accuracy of 1%. Both the net peak height of the 7.656-Mev line and the extrapolated background at the peak position are smaller than the 4.433-Mev peak intensity by a factor of ~2000, i.e., each is about 2 counts per minute. Approximately the same ratio of 7.656- to 4.433-Mev peak heights had been observed in the run at $E_\alpha=5.38$ Mev but the relative background under the 7.656-Mev line was somewhat higher than at $E_\alpha=5.81$ Mev.

Points in Fig. 4 at a peak position corresponding to a transition of 6 Mev showed the presence of a line of that energy whose intensity increased during the run. As discussed below the 6-Mev line is ascribed to carbon contamination.

Figure 5 shows the results of one run taken with a 0.2-mil thick Be target at $E_\alpha=5.81$ Mev. In this run the current readings in the beam cup were completely unreliable because of secondary electron effects and it could only be estimated that the beam intensity on the target was 1–2 μamp of He^{++}. The neutron detector was used not only to monitor the number of target reactions but its counting rate was referred to for maintaining a reasonably steady beam current. The data of Fig. 5 were obtained during a period of 36 hours of counting and the right-hand portion of the curve is the average of four passes over the spectrum. At the peak of the 4.433-Mev line the rate was 1000 counts per minute. The first background point above the 4.433-Mev line is down by a factor of 300 and the highest momentum point on the curve is less intense by a factor of 4000.

Although the statistics in Fig. 5 are not as favorable as in the thick target runs the 7.656-Mev line seems to show up more clearly than in Fig. 4 because of the lower sloping background. The amplitude of the 7.656-Mev peak after subtraction of an extrapolated background is $(5\pm1.5)\times10^{-4}$ relative to the 4.433-Mev peak height.

The presence of a line at 6 Mev which is 2% as strong as the 4.433-Mev line may be seen in Fig. 5. Its origin was investigated by cutting off the flow of gas to the stripping tube in the accelerator which reduced the He^{++} beam intensity to a very low value. Had the 6-Mev line been caused, for example, by proton contamination of the beam together with fluorine contamination on the target [the $F^{19}(p,\alpha)O^{16}$ reaction] the line intensity would have remained unchanged when the stripping gas was cut off. The virtual disappearance of the line proved that it was associated with alpha-particle bombardment and the probable reaction is $C^{13}(\alpha,n)O^{16}$ which forms[4] the 6.05-Mev nuclear pair-emitting state in O^{16} at $E_\alpha>5.05$ Mev. If the assignment is correct the number of 6.05-Mev transitions is $\sim3\times10^{-5}$ as great as the number of 4.433-Mev transitions. It is not known why the amount of carbon on the thin target was relatively so much greater than on the thick targets.

DISCUSSION

The evidence that the 7.656-Mev pair line has been observed in the $Be^9(\alpha,n)C^{12}$ reaction is contained in Figs. 4 and 5. By using the 4.433-Mev pair-line intensity as a measure of the number of reactions, which in turn gives the number of 7.656-Mev states formed, the relative branching of the 7.656-Mev level by pair emission to the ground state may be calculated by applying the appropriate factors to the ratio $(5\pm1.5)\times10^{-4}$ of the 7.656- to 4.433-Mev pair-line intensities. The absolute pair transmission of the spectrometer is not involved in the calculations.

In order to derive the relative total numbers of pairs from the ratio of peak heights a correction must be made for the efficiency of the spectrometer in detecting pairs of the 4.433-Mev $E2$ transition as compared with $E0$ pairs from the 7.656-Mev transition. In a previous calculation[16] an $E2/E0$ pair-line efficiency ratio of 1.26 was derived for an $E2$ transition of 5.3 Mev ($E0$ efficiency is nearly independent of energy). Although the $E2$ pair detecting efficiency is energy dependent the ratio 1.26 holds within a few percent for a 4.433-Mev $E2$ transition and it is accurate enough for the present calculations. The ratio of 7.656- to 4.433-Mev total pair intensities is therefore 6.3×10^{-4} which is a factor of 2.6 smaller than the upper limit set by Goldring et al.[9] and a factor of 4 smaller than the limit from Bonner's[11] magnetic spectrometer measurement. (Corrections have not been made for possible slight differences in the percentage widths of the two pair lines as a result of Doppler-broadening effects nor have corrections been made for the possible effects of angular distribution of the pairs relative to the alpha-particle beam.)

The relative numbers of 7.656- to 4.433-Mev transitions may be found by multiplying the ratio of total pair intensities by the internal pair conversion coefficient of the 4.433-Mev transition, which has a value of 1.3×10^{-3} according to Rose.[17] The 7.656-Mev transition is assumed to proceed 100% by nuclear pair emission. In order to derive $\Gamma_{e\pm}/\Gamma$ the ratio of 7.656- to 4.433-Mev transition intensities must be multiplied by the ratio R of neutron populations to the two levels, i.e., $R=N_{4.433}/N_{7.656}$. We thus obtain for the fractional decay of the 7.656-Mev level by nuclear pair emission, or in other words the ratio of the pair width to the total width:

$$\begin{aligned}\Gamma_{e\pm}/\Gamma&=(5\times10^{-4})\times1.26\times(1.3\times10^{-3})\times R\\&=8.2\times10^{-7}\times R.\end{aligned}$$

A precise number for R under the conditions of either the thick or thin target runs of this work is not available at present. As mentioned previously the ratio[13] of neutron group intensities at 0° and $E_\alpha=5.3$ Mev on a thin Be target is 8. Consequently we may derive only an approximate value for the ratio of widths by assuming $R\sim8$, namely

$$\Gamma_{e\pm}/\Gamma\sim7\times10^{-6}.$$

For a more reliable value of the ratio of widths a proper evaluation of the neutron population ratio R will be required. The thin-target run was made partly with aim of obtaining data which could be correlated later with neutron measurements. In that run a compromise target thickness of 0.7 Mev was chosen so as to result in a high enough pair spectrum yield but to allow the neutron groups to be resolvable. Time-of-flight or photographic-plate techniques are possible ways of making the neutron population measurements. Relative

[16] D. E. Alburger, A. Gallman, and D. H. Wilkinson, Phys. Rev. **116**, 939 (1959).

[17] M. E. Rose, Phys. Rev. **76**, 678 (1949).

intensities of the neutron groups will have to be integrated over all angles, since it has been shown by the work of Risser et al.[18] that strong angular distributions of the neutrons leading to the 4.433-Mev state occur in the $Be^9(\alpha,n)C^{12}$ reaction.

The ratio $\Gamma_{e\pm}/\Gamma \sim 7\times10^{-6}$ is a factor of $\sim$15 smaller than the estimate given by Cook et al.[5] There are three possible explanations for the discrepancy: (a) the width Γ_α has a value which is actually close to the Wigner limit rather than to the estimate of $\frac{1}{10}$ of that limit, (b) the ratio R is incorrectly assumed to be $\sim$8, or (c) the width $\Gamma_{e\pm}\sim5\times10^{-5}$ ev was derived incorrectly from the results of the $C^{12}(e,e')C^{12*}$ electron scattering experiment. Alernative (a) would seem to be the most plausible way of explaining the data.

If the estimated width of 0.0014 ev for $\Gamma_{3.2\gamma}$ is assumed to be valid, a value of Γ_α close to the Wigner limit would explain why attempts to observe the 3.2-Mev gamma ray have not yet met with success. Thus the partial decay of the 7.656-Mev level by 3.2-Mev gamma-ray emission would also be $\sim$15 times smaller than estimated by Cook et al., or $\sim$0.02% per decay of the level. This is 5 times smaller than the best experimental upper limit listed in Table I.

Although the observation of the 7.656-Mev pair line provides experimental support for the helium fusion mechanism of energy generation and element synthesis in red giant stars the stellar reaction rates depend on the dominant mode of decay to the ground state of C^{12} which is probably via the 3.2- to 4.433-Mev gamma-ray cascade. Until this branch has been measured the present results give only a rough indication of what the relative gamma-ray branching intensity might be.

It can be expected that improved techniques will eventually result in the detection of the 3.2-Mev gamma ray from the 7.656-Mev level in C^{12}. No attempt was made to do this with the magnetic pair spectrometer during the work at Oak Ridge for the following reasons. If it be assumed that $\Gamma_{3.2\gamma}\sim0.0014$ ev and $\Gamma_{e\pm}\sim5\times10^{-5}$ ev the 3.2-Mev gamma ray would be $\sim$30 times more intense than the 7.656-Mev nuclear pair transition. However, the internal pair conversion coefficient of the 3.2-Mev gamma ray is 10^{-3} which would make its pair line $\sim$30 times weaker than the 7.656-Mev pair line. Furthermore the 3.2-Mev pair line lies not only in the low-energy tail of true coincidences from the 4.433-Mev pair line but it is in a region of high random coincidence background. To observe the line using the techniques described in this paper would be exceedingly difficult. Improvements in the gamma-gamma coincidence or C^{12}-recoil methods may eventually solve this problem.

The present results give a strong indication, although not positive proof that the 7.656-Mev level in C^{12} has a spin-parity of 0+. According to the work of Eccles and Bodansky[12] the maximum fractional gamma-ray decay of the level is 0.1%. If we assume that $R\sim8$ in the $Be^9(\alpha,n)C^{12}$ reaction, as discussed above, an upper limit for the ratio of 7.656 to 4.433-Mev internal pair conversion line intensities may be derived for the alternative assignment[5] of 2+ to the 7.656-Mev state, i.e., $E2$ emission to the ground state. By taking the relative theoretical internal pair conversion coefficients into account the upper limit for the ratio of 7.656- to 4.443-Mev pair-line intensities would be $\sim1.8\times10^{-4}$. Since this limit is almost 3 times smaller than the experimentally observed ratio the measured intensity of the 7.656-Mev pair line would constitute proof of the $E0$ nature of the transition if it were not for the uncertainty in the knowledge of R. A smaller upper limit on $\Gamma_{7.6\gamma}/\Gamma$ would strengthen the assignment of 0+ to the 7.656-Mev level whereas an accurate measurement of R might make the assignment certain.

Because of the importance of the 7.656-Mev level of C^{12} in astrophysics it would be worthwhile to make an independent experimental check of the results described in this paper. The approximately constant ratio of 7.656- to 4.433-Mev pair line intensities in all of the $Be^9(\alpha,n)C^{12}$ runs suggests that the ratio of integrated yields of the two lines between $E_\alpha=5.1$ and $E_\alpha=5.81$ Mev is about the same as the ratio of integrated yields that would occur in a thick target at $E_\alpha=5.1$ Mev. Hence it would appear that as far as the ratio of the line intensities is concerned the choices of the target thickness and of the bombarding energy between 5 and 6 Mev are not critical. On the other hand the yield of the 4.433-Mev line, and presumably also of the 7.656-Mev line, rises rapidly with the energy of the bombarding alpha particles as may be seen from Fig. 3. Thus at a higher bombarding energy a smaller beam current can be used to obtain a desired counting rate. Alpha-particle energies of greater than 6 Mev may be even more favorable in this respect as long as the production of gamma rays from higher excited states in C^{12} does not increase the background at 7.656 Mev. Among the other improvements in the experiment which may be suggested is the use of an intermediate-image spectrometer having a larger annulus opening than the maximum opening in the present instrument and also having detecting crystals of sufficient diameter to match the correspondingly larger final-image size. A spectrometer pair resolution of 4–5% would not be disadvantageous and it might be expected that a factor of 2 or more could be gained in absolute pair transmission at a larger annulus opening. A factor of 2 improvement in the real to random ratio might be achieved if the earlier discussion of the random coincidences corresponds to the true situation and if a steady He^{++} beam were to be produced. It would of course be necessary to retain a coincidence resolving time of 1.0×10^{-9} sec or even to decrease this value if

[18] J. R. Risser, J. E. Price, and C. M. Class, Phys. Rev. **105**, 1288 (1957).

it is possible to do so without loss of coincidence efficiency.

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Activation Cross Sections for 14.8-Mev Neutrons and Some New Radioactive Nuclides in the Rare Earth Region*

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Activation cross sections on 27 stable nuclides of elements Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Er, Yb, and Lu were measured for 14.8±0.8-Mev neutrons. Highly enriched isotopes were used as targets in most cases, and in a few instances radiochemical separations were performed whenever it was necessary and possible in view of the product half-lives. The measured cross sections for $(n,2n)$ reactions were found to agree within an order of magnitude with predictions from statistical evaporation theory. However, experimental values of (n,p) and (n,α) cross sections generally appear to be larger than calculated from continuum theory of the compound nucleus. The cross sections show no significant effects due to the 82-neutron closed shell and, furthermore, the Levkovskii effect, which is quite striking in the low Z region, appears to be negligible for (n,p) and (n,α) reactions in the rare earth region. The $(n,2n)$ cross sections show little variation with mass number at constant Z, and they exhibit a decrease with increasing mass number at $N=82$.

Several previously unreported activities were observed; their half-lives, assignment, and gamma radiations are as follows: 12±3 min Pr^{148} from the $Nd^{148}(n,p)$ reaction; 0.5±0.1 min Sm^{157} from the $Gd^{160}(n,\alpha)$ reaction, 0.57±0.01 Mev gamma; 7±1 min Tb^{163} from the $Dy^{163}(n,p)$ reaction, 0.18±0.05-Mev gamma; 3.3±0.5 min Ho^{168} from the $Er^{168}(n,p)$ reaction, 0.85±0.05-Mev gamma; 40±10 sec Ho^{170} from the $Er^{170}(n,p)$ reaction; 4.4±0.4 min Dy^{167} from the $Er^{170}(n,\alpha)$ reaction; 2.0±0.5 min activity with gammas at 0.18±0.01, 0.25±0.01, and 0.36±0.01 Mev which may be Tm^{176}, Er^{173}, or possibly isomeric Yb^{177m} from enriched Yb^{176} bombardments. Tentative assignment of a 5.5±0.5-min activity to Tm^{174} is suggested from bombardment of enriched Yb^{174}.

INTRODUCTION

In a previous paper[1] cross sections for some samarium isotopes were reported for 14.8-Mev neutrons as part of a larger study in the rare earth region to determine whether the relative variation in (n,p) and (n,α) cross sections, as pointed out by Levkovskii[2] for low Z nuclides, persists in higher Z elements. In the low Z (up to $Z=22$) cases examined by Levkovskii,[2] the relative variation in (n,p) and (n,α) cross-sections at 14 Mev shows an almost integral decrease by factors of 2, 4, or 8 with increasing mass number at constant Z.

It was also of considerable interest in this work to investigate neutron cross sections in the region of the 82-neutron shell closure in order to determine whether shell effects[3] noticed in the region of $Z=20$ and $Z=50$ are present in the vicinity of $N=82$. As may be seen from the present results, both the Levkovskii variation and the shell closure appear to have negligible effect on the 14.8-Mev neutron cross sections in the rare earth region.

EXPERIMENTAL

Activation cross sections on 27 stable nuclides of elements barium, lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, dysprosium, erbium, ytterbium, and lutetium were measured for 14.8±0.8-Mev neutrons from the $H^3(d,n)He^4$ reaction with total fluxes of 10^{10} to 10^{11} neutrons/second from the University of Arkansas 400 kv Cockcroft-Walton accelerator. Samples of natural and enriched rare earths, either as metal or as oxides, were pressed into flat tablets of known weight and area, weighed thin copper monitor foils were placed in front and back of the tablets, and irradiated for periods ranging from a few minutes to about 9 hours.

By absolute beta counting of 9.9 min Cu^{62} (or 12.8 hour Cu^{64} when the length of the bombardment made Cu^{62} a poor monitor) induced in the copper foils, the

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