

C¹²(He³,n)O¹⁴ Reaction at 19, 22, and 25 MeV*

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With the aid of neutron detectors having thresholds at 1, 5, and 12 MeV, the angular distributions and average cross sections of the C¹²(He³,n)O¹⁴ reaction have been investigated at 19-, 22-, and 25-MeV He³ energy. All angular distributions show strong forward peaking suggestive of direct interactions. One case, involving only transitions to the ground state, exhibits a type of structure similar to that of protons from the C¹²(He³,p)N¹⁴ reaction to the first excited state, the $T=1$ analog of the O¹⁴ ground state. However, the momentum transfer at a reasonable radius is such that plane-wave theory would predict a minimum at 0° instead of the strong forward peak observed.

INTRODUCTION

REACTIONS of the type (T,p) and (He³,n) provide a means of adding two paired identical particles to a given target in what might be a simple extension of the mechanism of single-nucleon stripping reactions. The selection rules, if the particles remain paired, are more restrictive than those for similar reactions which deposit a deuteron, and they may, therefore, provide a better insight into the mechanism for two-nucleon transfer.

Most previous work on two-nucleon stripping has been done with incident energies of ≤ 6 MeV and with agreement between experimental and predicted angular distributions which leave much to be desired. There are, for example, the angular distributions obtained at 5.5 MeV from the (t,p) reaction by Jaffe *et al.*,¹ with fits to the theory of Bhatia² and Newns.³ It is concluded therein that the simplest form of the theory usually provides the best comparison and that the He³ form factor introduced by Newns reduces the large-angle cross section excessively. Towle and Macefield⁴ have examined the angular distribution of the ground-state neutrons in the C¹²(He³,n)O¹⁴ reaction. They were able to secure an approximate fit to Newns' theory at 4.65 MeV but not at 4.98 and 5.26. Fulbright *et al.*,^{5,6} in similar measurements, give results to 10.5-MeV He³ energy and claim good fit to Newns' calculations if the He³ form factor is included. Since these results and others suggest some doubt about the adequacy of the interpretation of the processes involved, the present experiment on the C¹²(He³,n) reaction was undertaken as part of program of examination of the general features of two-proton capture as might be revealed through

observation of neutron groups of differing energies as produced by bombarding targets with He³ particles in the 19–25-MeV range.

EXPERIMENTAL

Threshold neutron detectors were employed for angular-distribution measurements in this experiment, utilizing broadband energy sensitivity as consistent with examination of the general features of reactions of this type.

Two types of activation detectors, Cu and Si, in the form of metallic buttons $\frac{1}{2}$ and $\frac{3}{8}$ in. in diameter, respectively, were employed. A third type of detector was a U²³⁸ fission chamber. The relative neutron sensitivity of these detectors is shown in Fig. 1. The dashed portion is an extrapolation. Six activation detectors could be irradiated simultaneously and then counted approximately 2 min later in a battery of six calibrated methane flow counters equipped with automatic time and count recording. Decay data were analyzed with a computer code providing background corrections, calibration factors, dead-time corrections, and a least-squares fit to multiple exponential decays. Detector exposures were customarily 20 min for Cu, 10 min for Si and counting periods at least 1000 and 600 sec, respectively. It was found that a two-parameter decay curve provided a good fit for both Cu and Si data. In the case of Cu, the activity of interest, 9.8-min Cu⁶³(n,2n)Cu⁶² activity, was nearly 98% of the total for most 0° irradiations, decreasing to 50% near 90°. The contaminant was 5.1-min Cu⁶⁵(n, γ) activity. For Si, the 2.2-min Si²⁸(n,p)Al²⁸ activity of interest was about 90% of the total, relatively independent of angle. The contaminant appeared to be a mixture of Si²⁹(n,p)Al²⁹ and Si³⁰(n, α)Mg²⁷. From the code analysis, irradiation, and lapse time information the desired activity corresponding to saturation (infinite irradiation) was computed. In addition, a similar procedure was carried out following exposure of each detector to a known 14-MeV neutron flux from the Los Alamos Cockcroft-Walton generator. These data combined with cross-section information from the literature provided a neutron-flux calibration for the detectors.

The third detector, a U²³⁸ fission chamber in the form of a 1-in. right cylinder, contained approximately 1 g of

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¹ A. A. Jaffe, F. de S. Barros, P. D. Forsyth, J. Muto, I. J. Taylor, and S. Ramavatharain, Proc. Phys. Soc. (London) **A76**, 914 (1960).

² A. B. Bhatia, K. Huang, R. Huby, and H. C. Newns, Phil. Mag. **43**, 485 (1952).

³ H. C. Newns, Proc. Phys. Soc. (London) **A76**, 489 (1960).

⁴ J. H. Towle and B. E. F. Macefield, Proc. Phys. Soc. (London) **A77**, 399 (1961).

⁵ H. W. Fulbright, J. W. Verba, and V. K. Deshpande, Bull. Am. Phys. Soc. **6**, 380 (1961).

⁶ H. W. Fulbright, W. Parker Alford, O. M. Bilanink, V. K. Deshpande, and J. W. Verba, USAEC Report NYO 10034.

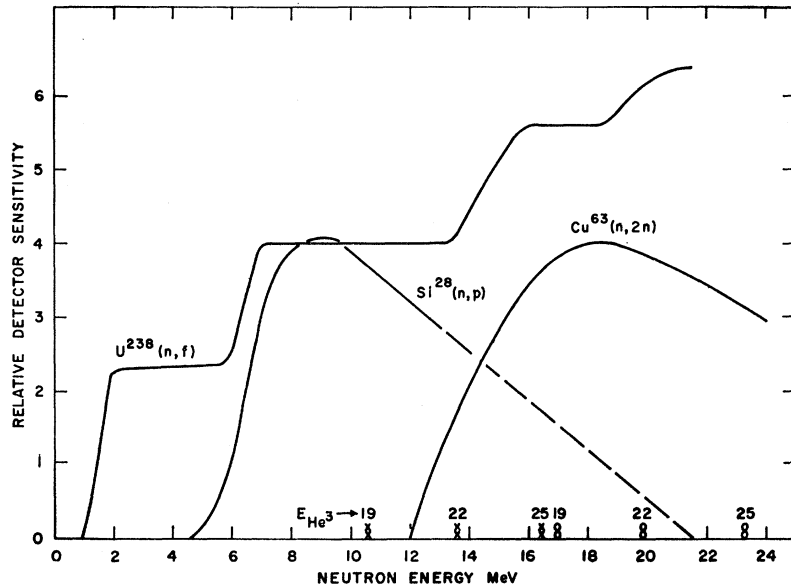


FIG. 1. Relative detector sensitivity curves. Data from BNL 325 with extrapolations shown as broken lines.

material highly depleted in U^{235} on a rather thick foil. As a result, the counting-rate bias curve was not flat. Its operation was checked periodically with a PuBe source in a fixed position as a safeguard against unwanted electronic drifts.

The Los Alamos variable-energy cyclotron was used as a source of He^3 particles of energy between 18 and 25 MeV. This machine is equipped with a long external beam tube containing collimation and electric and magnetic focusing and beam positioning. It also contains a provision for beam-energy determination by comparison with an alpha standard. The beam tube terminated in a

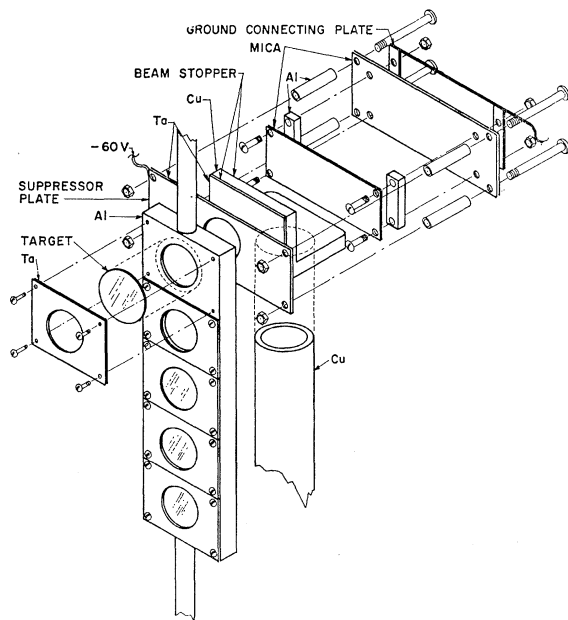
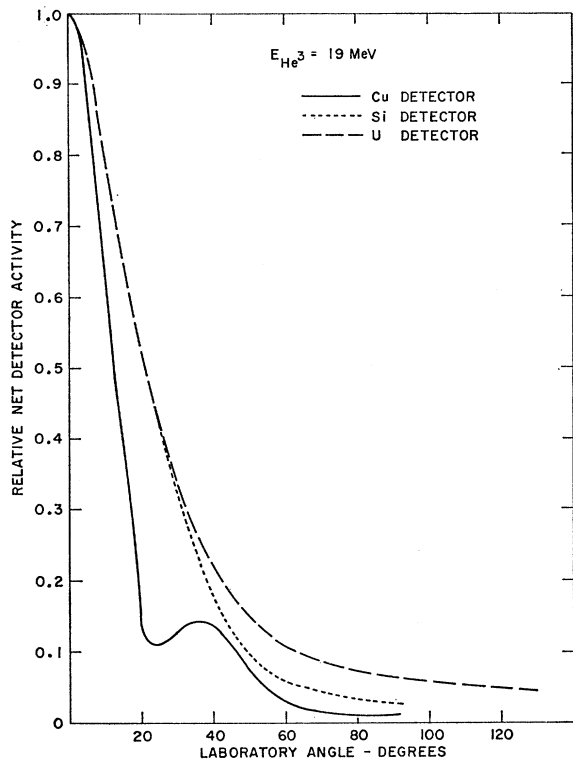


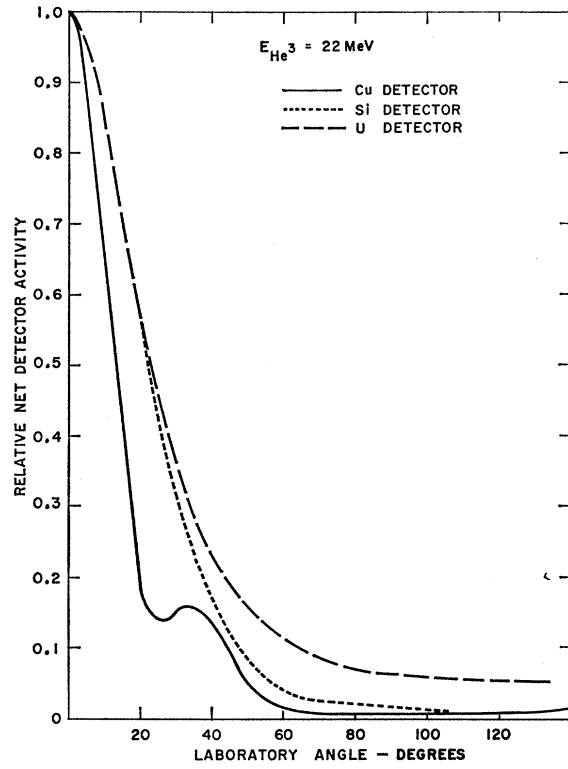
FIG. 2. Target assembly detail.

6-in.-diam, $\frac{1}{16}$ -in. wall, 20-in.-long Al cylinder the axis of which was vertical and perpendicular to the He^3 beam. The details of the target assembly and beam stopper are shown in Fig. 2 in an exploded view. The target slide with provision for mounting four targets and retaining one blank for background measurements is centered in the cylinder and externally movable along the cylinder axis. The target for this experiment was a disk of graphite 22 mg/cm² thick mounted between two thick Ta collimator plates each with a $\frac{3}{8}$ -in.-diam hole. External indices provide accurate positioning of target or blank. Approximately $\frac{5}{16}$ in. beyond the target the beam is stopped by thick Ta soldered to an air-cooled Cu tube to which the current integrator is connected. This arrangement permitted angular-distribution measurements including 0° with small geometrical separation of target source and beam-stopper background source. Ta was chosen for collimation and beam-stopping after a preliminary experiment showed that its neutron yield at 0° when bombarded by 25-MeV He^3 particles and measured with a Cu detector was less than 2% of that from the C target. The absence of a conventional Faraday cup in this geometrical arrangement of target and stopper can cause erroneous beam-current measurement. In order to minimize such an effect, an insulated Ta sheet with a $\frac{1}{2}$ -in.-diam hole concentric with the $\frac{3}{8}$ -in. collimators of the target slide was mounted between the latter and the beam stopper as an electron suppressor. A curve of beam current vs suppressor voltage, with neutron yield monitored by the fission chamber, showed saturation at -40 V. Accordingly, data were taken with a fixed -60 V on this suppressor plate.

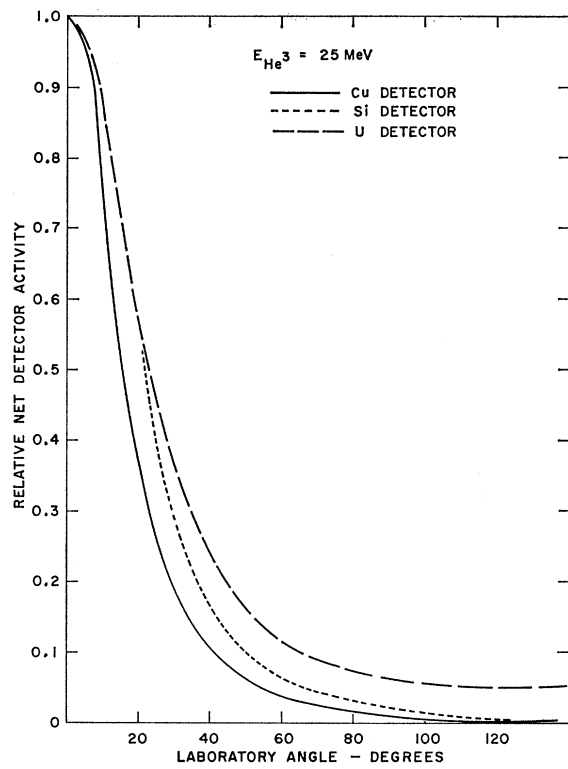
Since the energy loss in the target is not negligible (4.5 MeV at 25 MeV), the appropriate target-out background is not that at the incident He^3 energy but at an energy lower by the target loss. Consequently, in addi-



(a)



(b)



(c)

FIG. 3. (a), (b), (c) Experimental relative angular distributions for three detectors at three He^3 energies as indicated.

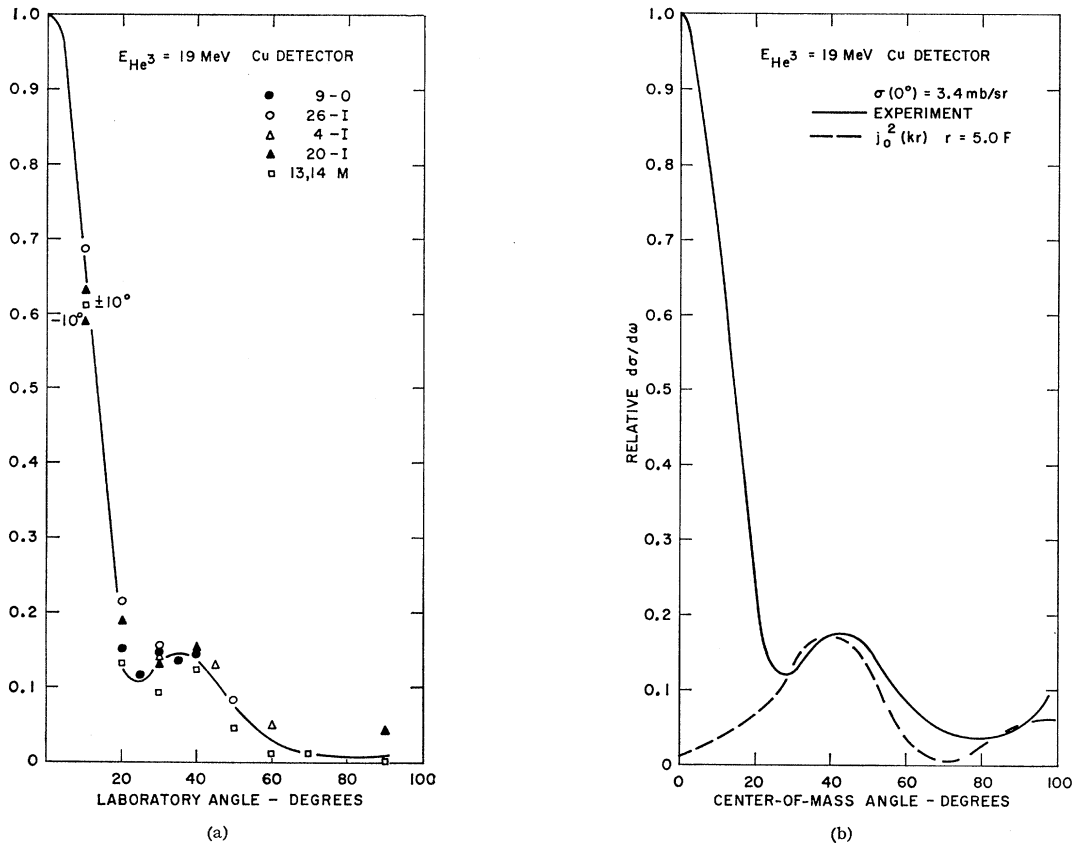


FIG. 4. (a) Sample experimental angular distribution illustrating reproducibility in different runs at different distances, I , M , O . 10° points each side of 0° shown. (b) Corrected experimental angular distribution with simple theoretical comparison.

tion to background data at 19, 22, and 25 MeV on the angular distribution with target out, data were also taken at 14.7 MeV. From these four energy points an excitation curve for the background at each angle was constructed. With such a family of curves and the average energy-loss in the target obtained from the tables of UCRL-2301, the appropriate background subtraction was made. Since the emergent cyclotron beam was collimated to $\frac{1}{2}$ -in. diam at a point 48 in. from the target and checks made of alignment and focus, it was assumed that no background originated at the full beam energy from the Ta collimator supporting the target. The correction, if this assumption were not true, is second order for the target-out position would contribute an identical background.

The activation detectors were supported at known angles by several thin, detachable Al rings concentric with the target cylinder. Target-detector distances were 3, 4.5, and 9 in., identified as I , M , O . Constancy of the activity-distance squared product established confidence that any general room background was negligible. Symmetry was checked by exposures on both sides of an optically determined 0° position. The fission detector was mounted on a milling head for rotation about the target axis at a radius of 44 cm. During exposures of the

activation detectors, this chamber was used as a monitor at the 0° position. One activation detector was also always exposed at 0° in order to obtain relative measurements at other angles in the same exposure.

RESULTS

Relative angular distributions as observed at three different bombarding energies are shown for each of the three detectors in Figs. 3(a), (b), and (c). These curves are shown with experimental points eliminated since their number is sufficiently large to obscure the general trend observed with different detectors. A sample angular distribution with experimental points from a number of different runs with varying experimental conditions (slight differences in ${}^3\text{He}$ energy and cyclotron adjustment, exposures at different distances indicated by I , M , O over a period of several months) is shown in Fig. 4(a). Data taken at higher ${}^3\text{He}$ energies were generally even more reproducible.

In only one case can the experimental data be corrected properly for center-of-mass motion and for detector sensitivity. This can be understood by reference to Fig. 1, the data for which were obtained from the compilation of BNL-325 and from the article by Allen

and Henkel.⁷ From the ground state $Q = -1.15$ MeV for the reaction and the lowest excited state reported by Towle and Macefield⁴ as 5.905 MeV above the ground state, the 0° laboratory neutron energy of these two groups can be computed for the three He³ energies of 19, 22, and 25 MeV. The expected position is indicated by the symbols ○ and ✕, respectively. Higher excited states at 6.30 and 6.586 MeV⁴ are not shown, though their location will be just below the ✕ positions. It is clear from this figure that at $E_{\text{He}^3} = 19$ MeV the Cu⁶³ reaction will detect only the ground-state (g.s.) group. In this case the effect of detector sensitivity on the angular distribution can be calculated and the conversion of center-of-mass angle accomplished. This has been done for the smoothed data of Fig. 3(a) and is shown as the solid curve of Fig. 4(b). The rise beyond 80° may not be significant since a correction factor between 3 and 10 for the detector sensitivity quite close to an uncertain threshold has been applied to low-intensity experimental data.

For this particular case also the absolute cross-section scale can be established from the sensitivity curve of the detector and the known Cockcroft-Walton calibration flux. The results of three separate runs are: $\sigma(0^\circ) = 3.2, 3.4, 3.5$ mb/sr. A weighted average of 3.4 mb/sr is chosen with an estimated cumulative error of all factors of 20% for the reaction C¹²(He³, n)O¹⁴ g.s. at 19-MeV He³ bombarding energy. The integrated cross section to 90° is 2.5 mb.

Although no absolute cross-section calculation can be made for the other cases, it is possible in each to obtain an average cross section for "Si" or "Cu" neutrons which gives a relative measure of the 0° laboratory yield. If the Si and Cu sensitivity functions of Fig. 1 are integrated from threshold to the maximum possible neutron energy and divided by that energy interval, an average detector sensitivity \bar{S} is obtained. If R is defined as the saturated activity per incident He³ particle for a cyclotron exposure, C the similar quantity for a Cockcroft-Walton exposure in a known neutron flux $F/\text{cm}^2 \text{ sec}$ for which the detector sensitivity is S_{14} , then the cyclotron flux φ over this energy interval is given by

$$\varphi = \frac{R S_{14}}{C \bar{S}} F \text{ neutrons/cm}^2 \text{ He}^3 \text{ particle.}$$

This procedure is equivalent to the assumption of a constant neutron flux per unit energy interval from detector threshold to ground-state group energy. With this flux value, detector solid angle, and target areal density, a cross section for the reaction is calculated in the usual fashion. It should be emphasized that this cross section, operationally so defined, can be very spectrum-dependent. An illustration is provided by comparison between the σ given above for a Cu detector

TABLE I. Average 0° laboratory cross sections in mb/sr for Si and Cu detectors determined as described in the text, and relative U²³⁸ fission detector yield per microCoulomb.

Detector \ He ³ energy (MeV)	19	22	25
Si	32	51	72
Cu	5.6	8.4	20
Fission	58	87	100

at $E_{\text{He}^3} = 19$ MeV which refers to one group and that given in Table I below obtained by the \bar{S} procedure. Table I lists the cross sections as obtained by this method for all the cases examined. Since no calibration of the fission chamber was made, only relative results are included. A crude estimate of its efficiency yielded the somewhat surprising result that it does not detect many more neutrons than the Si detector in spite of its lower threshold.

DISCUSSION

As mentioned above, the cleanest situation occurs for the Cu detector at 19-MeV He³ energy. Although these data could be contaminated with neutrons from the 1% C¹³ in the normal graphite target, the cross section for the ground-state group in pure C¹³ is only of the order of 1 mb/sr at 0°. Furthermore, the angular distribution [Fig. 4(b)] appears as a reasonable extension of lower energy investigation. According to plane-wave stripping theory the angular distribution for the ground-state transition, $\Delta L = 0$, should be of the form

$$d\sigma/d\omega \sim |j_0(kr)|^2.$$

This function is also shown as the dashed curve in Fig. 4(b). Here k is the momentum transfer of the "diproton lump" to the target and r , in accord with Fulbright,^{5,6} is taken as 5 F. The ordinate of this curve has been normalized to the experimental data at 40°. Although a slightly smaller radius could improve the fit at larger angles, the main point is that no expression of this type with or without multiplicative form factor can account for the sharp rise at 0°. At this energy the 0° momentum transfer, $\sim 0.64 \text{ F}^{-1}$, is such that for a reasonable radius $j_0(kr)$ is very near its first zero at $kr = \pi$. This is thus an exceptionally clear case of failure of the plane-wave approximation. Fortunately, the modification of using wave vectors inside the nucleus with a potential ~ 50 MeV as suggested by Rodberg⁹ yields a momentum transfer at 0° nearly twice as large which places the argument near the second zero of j_0 . Furthermore, the next maximum would occur at smaller angles than observed. Decreasing the well depth by a factor two will give a 0° peak but not one at 40°. Hence, this method of allowing for distortion is inadequate in this case.

⁷ D. W. Allen and R. L. Henkel, Progr. Nucl. Energy, Ser. I 2, 1 (1958).

⁸ H. C. Bryant, E. R. Flynn, and W. T. Leland (private communication).

⁹ L. S. Rodberg, Nucl. Phys. 21, 270 (1960).

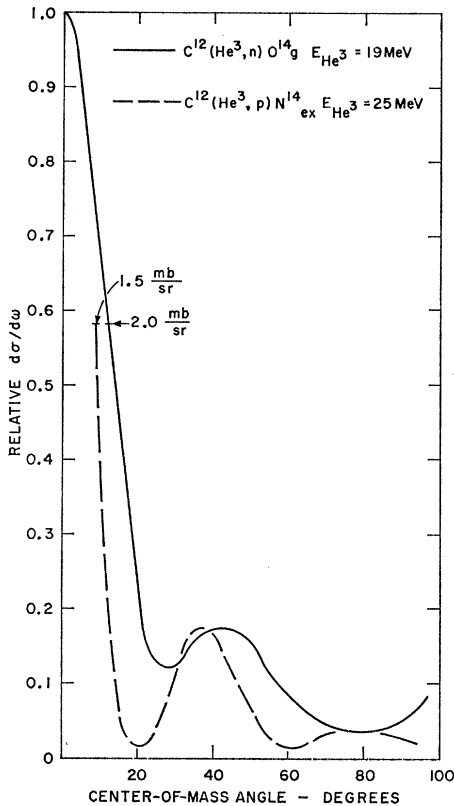


FIG. 5. Comparison of angular distributions for transitions to the $C^{12}(\text{He}^3, p)N^{14}$ first excited state and the $C^{12}(\text{He}^3, n)O^{14}$ ground state.

The narrow 0° peaking is not particularly associated with this reaction on C^{12} for it has been observed for a number of targets in a survey of (He^3, n) reactions now in progress. It is believed to be the effect of distortion, primarily of the neutron waves, by the nuclear potential as has previously been suggested.¹⁰⁻¹² This effect has been tested to a degree¹³ by performing distorted-wave calculations for a number of cases with and without a potential acting on the neutrons. There is invariably a 0° peaking when the neutron potential is included. Although a number of trials have been made to fit the observed angular distribution,¹⁴ no real success has yet been achieved nor is the difficulty transparent. Consequently, the hope that this reaction with capture of two protons to fill the p shell would provide insight into the mechanism has not been realized.

Before leaving this case it is interesting to make a comparison with the angular distributions of protons in the $C^{12}(\text{He}^3, p)N^{14}$ reaction to the first excited state, a member of the $T=1$ triad with the ground state of O^{14} .

The distribution has been measured in this Laboratory¹⁵ at $E_{\text{He}^3} = 25$ MeV for which the 0° momentum transfer k is only about 30% larger than that in the (He^3, n) reaction discussed above. Both the He^3 particles and the protons have sufficient energy to be only slightly perturbed by Coulomb effects. Hence, if the angular distribution is indeed a function of k but modified by nuclear distortion, the two distributions should show considerable similarity. There is, of course, the difference that in deuteron capture neither particle completes the shell. The comparison is illustrated by the two curves of Fig. 5 in which the similarity of appearance is rather striking in spite of the poorer resolution and more difficult background problems in the neutron experiment. Although the absolute cross sections indicated in this figure do not have the ratio 2 found by Fulbright^{5,6} over a wide energy range, they are not at the same energy. Since Table I shows that the Cu detector response increases by more than a factor three between 19 and 25 MeV, the factor two would easily be obtained and leave a balance for transitions to higher states.

It should not escape notice that all previous work on the angular distributions to the states of this $T=1$ triad^{1,5,6} really shows no structure that can be fit with a form of $|j_0(kr)|^2$ except for the 0° peak. If, as seems likely from these recent higher energy (He^3, n) and (He^3, p) experiments, the 0° peak arises mostly from wave distortion, then doubts about conclusions wholly dependent on theoretical interpretation of angular distributions are warranted.

Discussion of the remaining experimental data is limited by ignorance of several factors. In comparison to the 19-MeV Cu detector data, if either the He^3 energy is raised or the detector threshold lowered (as with Si and U^{238}) other neutron groups can be detected. A group associated with transitions to the first excited state of O^{14} would have an energy shown at \times in Fig. 1 for the He^3 energy designated. It is clear that the Si and U^{238} detectors will respond to these neutrons even at $E_{\text{He}^3} = 19$ MeV as well as to lower energy neutrons corresponding to transitions to higher, unknown states of O^{14} . It may be noted that O^{14} becomes proton-unstable at an excitation of 4.6 MeV. At the other extreme, the Si detector should have little or no response to ground-state neutrons at $E_{\text{He}^3} = 25$ MeV. But observable neutrons need not arise from the (He^3, n) reaction. The $(\text{He}^3, n\bar{p})$ reaction with $Q = -5.77$ MeV could produce neutrons with a maximum energy about 1 MeV higher than the position \times . This reaction is quite probable with a total cross section of 60-80 mb between He^3 energies of 14-24 MeV.¹⁶ It is quite possible that this reaction could have a stripping character similar to the roughly equally probable (He^3, d) reaction¹⁷ but involving a dissociated deuteron. Such a direct interaction

¹⁰ I. E. McCarthy, Nucl. Phys. **11**, 574 (1959).

¹¹ N. Austern and S. T. Butler, Phys. Rev. **109**, 1402 (1958).

¹² S. T. Butler, N. Austern, and C. Pearson, Phys. Rev. **112**, 1227 (1958).

¹³ E. M. Henley (private communication).

¹⁴ A. M. Lockett, III (private communication).

¹⁵ A. G. Blair and H. E. Wegner (private communication).

¹⁶ D. R. F. Cochran and J. D. Knight, Phys. Rev. (to be published).

¹⁷ H. E. Wegner and W. S. Hall, Phys. Rev. **119**, 1654 (1960).

mechanism could produce neutrons heavily concentrated in the forward direction as observed. However, perhaps the most probable energy division between the neutron and proton is near equality in which case the average neutron energy would be more nearly one-half the maximum. Then contamination of the data by neutrons of such origin would be less serious. Although the O¹³ product of a (He³, 2n) reaction is not known, a calculation from the masses of its known isobars yields an estimate of $Q = -25$ MeV. Neutrons from this reaction would then not be observed in this experiment. Finally, it seems unlikely that the tight neutron binding in C¹² would permit any appreciable neutron ejection by inelastic processes. With these considerations in mind, it will be assumed that the neutron angular distributions of Figs. 3(a), (b), (c) and the 0° laboratory cross sections of Table I are primarily associated with the C¹²(He³, n)O¹⁴ reaction, though with an unknown energy spectrum below an energy corresponding to an O¹⁴ excitation of around 6.5 MeV.

The most immediately noticed feature of the angular distributions is the sharp forward peak obtained with all detectors at all energies. Although an increasingly poor signal-to-background ratio makes large-angle points less reliable, numerous runs to 150° failed to show any rise beyond 90°. For very special neutron groups there can be an artificial decrease of the large-angle yield due to the neutron energy approaching a detector threshold. It would be remarkable, however, for such to occur for three thresholds and three bombarding energies. The U²³⁸ detector angular distribution is especially surprising since an appreciable number of evaporated neutrons should be above its 1-MeV threshold if compound nuclear processes are competitive with direct interaction. The similarity of the U²³⁸ and Si distributions is consistent with the crude estimate that both detectors receive about the same total number of neutrons in spite of their rather different energy sensitivity. It is as if only a few relatively high-energy neutron groups were formed in the reaction.

If all neutron groups observed are the result of a stripping process as the ground-state group at 19 MeV certainly seems to be, then the over-all result would be expected to be a superposition of several different angular momentum changes. In simple stripping theory this would mean an angular distribution of the form

$$\sigma(\theta) \sim \sum_l |a_l j_l(kr)|^2,$$

where l is the angular momentum change of each group. Because of the dependence of k on energy and angle, the argument kr varies from about 3 at 19 MeV, 0°, to about 6 at 25 MeV, 60°, for the ground-state group. Groups corresponding to higher excitation would have arguments around 5 at 0° and also reach 6 at 60°, nearly independent of excitation. In view of the character of the functions j_l , especially for odd and even l , it would be expected that a superposition of the form of the sum would result in a much greater broadening of the angular

distributions than is observed for the different detectors and He³ energies. It is as if wave distortion is effective in causing a 0° peak for whatever l may be involved. An alternative explanation might be that of Yoshida¹⁸ which would say that the $l=0$ terms of the sum could be strongly enhanced. However, groups corresponding to transitions to higher 0⁺ states would be expected to have similar angular structure to that of the ground-state group shifted to smaller angle as the result of larger k . Such structure could be masked by the rapid rise toward 0°. No evidence has been seen in the data, though a transition of comparable strength to that of the ground state might not be missed. It is unfortunate that nothing is yet known of the character of the excited states of O¹⁴. That other than the ground-state group is involved is clear from the 0° cross sections obtained, from the difference in the distributions between Cu and the other detectors at 19 MeV, and from the disappearance of the 40° rise of the Cu data at 25 MeV. The persistence of this rise at 22 MeV is understandable from inspection of the sensitivity curve of Fig. 1.

With the exception of the Cu detector results already noted, there is practically no change in the angular distributions between $E_{\text{He}^3} = 19$ and 25 MeV. This is consistent with the small percentage change in momentum transfer for any single neutron group for this energy change and also with a small or negligible change in wave distortion. It is not consistent with new groups of different angular momentum character passing above the detector thresholds.

Table I shows that the 0° laboratory yields increase about 50% between 19 and 22 MeV for all detectors. Between 19 and 25 MeV the Si detector shows a 40% increase, Cu a factor of 2.4 but U²³⁸ only 15%. The large increase for the Cu detector is almost certainly due to the X group approaching the maximum of the Cu sensitivity curve. However, in view of the shapes of the sensitivity curves for Si and U²³⁸ the only consistent explanation is the statement that the ground-state yield must decrease between 22 and 25 MeV while the group or groups in the X region increase with this energy change. Fulbright⁶ has already observed a rather rapid change of the ground-state yield in the 7–11-MeV region. The smaller increase of the U²³⁸ detector between 22–25-MeV than in the 19–22-MeV interval once again appears to indicate the absence of excitation of especially high states in O¹⁴.

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¹⁸ S. Yoshida, Nucl. Phys. 33, 685 (1962).