Inelastic Events Induced by 32-Mev Protons on Helium

JACK BENVENISTE AND BRUCE CORK
Radiation Laboratory, University of California, Berkeley, California
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A study of inelastic events induced by the bombardment of helium nuclei with 32-Mev protons has been made.

No low energy group of protons has been found which would indicate the existence of an excited, heavy-particle stable state of the helium nucleus. The absence of a bump on the continuum of protons from the reactions

$$p+\mathrm{He}^4\rightarrow \mathrm{He}^3+n+p',$$
$$p+\mathrm{He}^4\rightarrow \mathrm{H}^3+p''+p',$$

indicates a maximum cross section of 0.1 mb/sterad at 45° c.m. for the production of a single excited helium level as high as 23.3 Mev above the ground state. Further, the observed rather flat shape of the continuum requires the existence of at least two levels of about 1-Mev half-width, separated by about 1 Mev.

The angular variation of the differential cross section for the production of deuterons from the reaction

$$p+\mathrm{He}^4\rightarrow \mathrm{He}^3+d$$

has been measured. When the principle of detailed balancing is invoked, good agreement with the prediction of the theory of Butler is obtained for r_0 (range of nuclear forces plus radius of He^3 nucleus) $=4.2\times 10^{-13}$ cm.

INTRODUCTION

EVIDENCE for the nature of nuclear forces comes from several sources. From binding energy measurements, especially of light nuclei, and from nucleon-nucleon scattering experiments, one may arrive at some conceptions of the character of the forces acting between elementary particles. Application of these ideas to the solution of more complex systems should then serve as a check on the accuracy of these concepts.

Thus, in 1936, it was shown theoretically[1] that, on the basis of what was then known about nuclear forces, the He^4 nucleus could be expected to have at least one excited level which was stable to particle disintegration. However, the analysis was based on sum rules valid only for neutron-proton potentials of the ordinary type. It was pointed out that if new sum rules are set up considering Majorana forces, the upper limit on the excitation energies which can be deduced from them are too high to be useful.

Bethe and Bacher[2] deduced the probable existence of two stable excited levels, a ^1P state at ≈ 16 Mev and a ^3P state at ≈ 10 Mev, and concluded that this was compatible with the experimental data available in 1936.

More recently, King and Goldstein,[3] pointed out that the observed $1/v$ behavior of the He^3 slow neutron absorption cross section indicated the absence of a bound excited state of the compound He^4 nucleus not too far below 20.5 Mev; furthermore, in view of the large (≈ 5000 barns) $\mathrm{He}^3(n,p)\mathrm{H}^3$ cross section[4] in the $1/v$ region, the existence of a broad unstable level in He^4 above 20.5 Mev is in the realm of possibility.

One of the more provocative recent experiments in this field is that of Argo *et al.*,[5] at Los Alamos. The yield of γ-rays from the reaction $T(p,\gamma)\mathrm{He}^4$ was observed as a function of the incident proton energy. A cross section which increased more rapidly than could be explained by barrier penetration was interpreted as evidence for the existence of an excited level in the He^4 nucleus. Only the rising portion of the yield curve was observed in this work, since 2.5 Mev was the highest proton energy available. However, this curve was fitted with a Breit-Wigner single level resonance formula, and the constants evaluated. The presumed level was assigned an excitation energy of 21.6 Mev and a half-width ≈ 1 Mev.

Shortly after the first announcement[6] of these results, Professor W. K. H. Panofsky suggested that the 32-Mev proton linear accelerator was ideal equipment to resolve the question of the existence of an excited He^4 level by the method of inelastic scattering of protons from helium.

The inelastic scattering process may be described in the following way. As illustrated in Fig. 1, a proton incident on a target nucleus (A, Z) in its ground state E_0, may be considered to form a compound nucleus $(A+1, Z+1)$ in an excited state. One of the ways in which the excited compound nucleus might decay is by re-emitting a proton. If the proton (P) comes off with energy equal to that of the incident proton, the target nucleus is left in its ground state; however, if the target nucleus possesses an excited level E_n, the compound nucleus may decay to this level by emitting a proton (p') of less energy than that of the incident proton. This proton is said to be inelastically scattered. Thus, to search for excited levels of the target nucleus, one

[1] E. Feenberg, Phys. Rev. **49**, 328 (1936).
[2] H. A. Bethe and R. F. Bacher, Revs. Modern Phys. **8**, 147 (1936).
[3] L. D. P. King and L. Goldstein, Phys. Rev. **75**, 1366 (1949).
[4] J. H. Coon and R. A. Nobles, Phys. Rev. **75**, 1358 (1949).
[5] Argo, Gittings, Hemmendinger, Jarvis, and Taschek, Phys. Rev. **78**, 691 (1950).
[6] R. F. Taschek, Phys. Rev. **76**, 584 (1949).

requires detectors with which to analyze the energy spectrum of scattered particles.

EXPERIMENTAL DETAILS

Bombardment Geometry

A diagram of the bombardment geometry is shown in Fig. 2. The 32-Mev proton beam from the linear accelerator was first passed through a "stripping foil" to remove molecular ions and then through the pre-magnet collimator C_1, usually set at about $\frac{1}{8}$ in.$\times\frac{1}{8}$ in. After being deflected into the 20° port by the analyzer magnet M, the beam was further collimated to $\frac{1}{8}$ in. diameter at C_2 and C_3, before it entered the scattering chamber S. Then, the scattering chamber was carefully aligned, and the beam was collected in a Faraday cup and integrated.

Vacuum and Gas-Handling System

Before a bombardment, the chamber was evacuated, flushed thoroughly with the target gas, then pumped down to ≈ 10 microns. After isolating the chamber from the pumps, helium was slowly introduced through the charcoal-liquid air trap; the rate of flow was adjusted at the regulator. The combination of charcoal and the liquid air is capable of filtering out all gases but He, Ne and H_2. Since the producer specifies the bottled He to be 99.95 percent pure, with less than 0.03 percent H_2, target impurity was considered inconsequential. Actually, since the major impurity is hydrogen, a range spectrum of scattered particles would show the contributions from He and H_2 as separate.

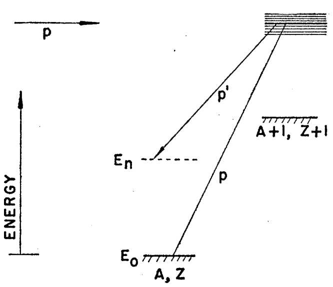

FIG. 1. Schematic description of elastic and inelastic scattering processes.

The chamber was filled with the target gas to a pressure of about 1 atmosphere. This pressure was maintained at a constant differential with respect to atmospheric pressure by means of an oil-filled manometer through which the target gas bubbled continuously during the course of a run. The pressure differential was of the order of 10 in. of oil ("Litton" diffusion pump oil—0.9 g/cm^3), i.e., about 2 percent of the total pressure. The atmospheric pressure was read before and after a run to 0.1 mm on a precision mercury barometer.

Beam Integrator

The beam was collected in a Faraday cup placed in a vacuum chamber. The charge was placed on a low leakage condenser connected to the grid of a 5803 electrometer tube used as a null indicator. The feedback voltage, or a fraction thereof, was applied to a Leeds and Northrup recording voltmeter which was self-calibrating against a standard cell.

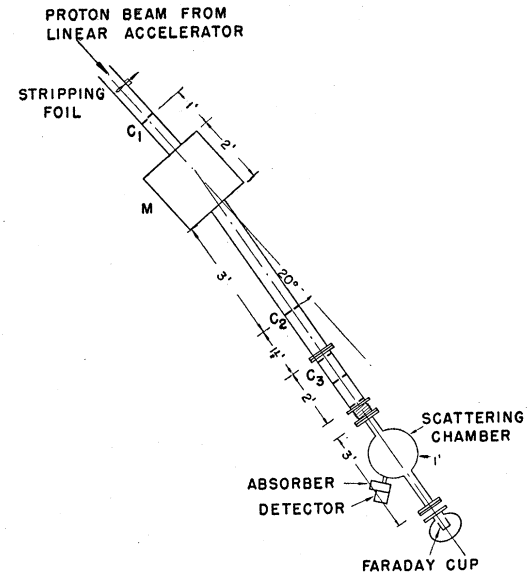

FIG. 2. Bombardment geometry.

The condenser used was compared on a General Radio Company impedance bridge with a G.R. standard condenser whose capacitance was known within 0.1 percent.

Electrons which might be coming down with the proton beam were kept from entering the Faraday cup by two permanent horseshoe magnets placed just beyond the scattering chamber. A second set of magnets provided a flux density of about 200 gauss just outside the Faraday cup to prevent secondary electrons from emerging.

Design and Characteristics of the Detector

Scattered particles were detected in a telescope of three gas proportional counters. Between the second and third counters was an aluminum foil 5.82 mg/cm^2 thick. By mixing pulses from the first two counters in coincidence and from the last counter in anticoincidence, the telescope counted all particles which stop in the foil. Hence, referring to the thickness of this foil as ΔR, if we place an absorber R in front of the telescope, it detects those particles having a range between R and $R+\Delta R$. Thus by changing R one obtains a differential range spectrum of the scattered particles.

The proportional counters were of the multiple-wire type—two-mil wolfram wires spaced $\frac{1}{4}$ inch apart over an area 2 inches in diameter, suspended with polystyrene spacers between thin aluminum ground planes $\frac{1}{2}$ in. apart. A trio of counters was designed as a unit; i.e., three counters were formed by alternating alumi-

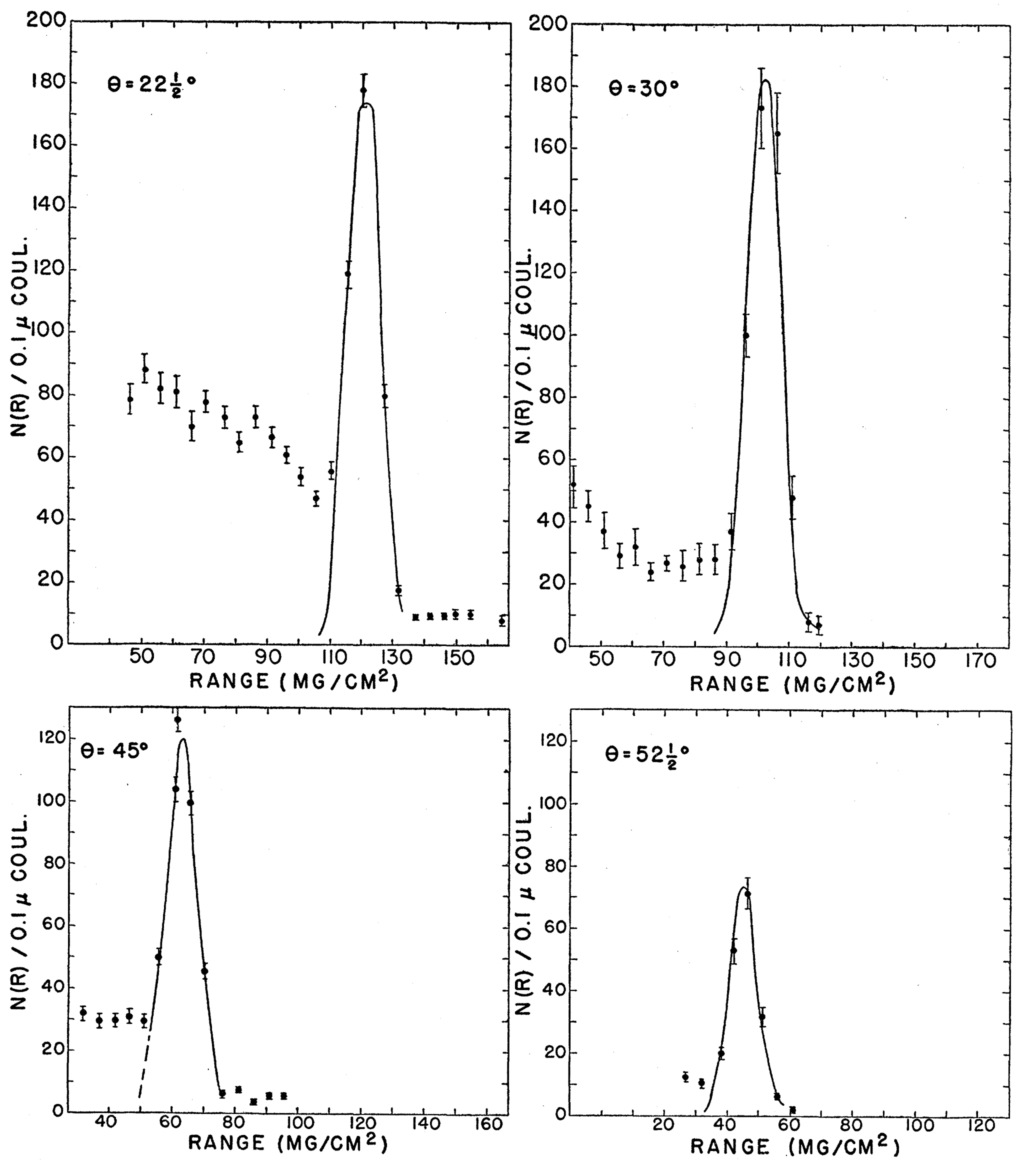

FIG. 3. Differential range spectra of charged particles inelastically scattered when He^4 is bombarded with 32-Mev protons. It is established in the text that the peak is made up of deuterons, and the continuum below the peak, of inelastically scattered protons.

num foils and multiple wire grids contained in a single chamber.

The counters were operated at about 950 volts, and the linear amplifier gains were adjusted so that the largest pulses were not quite overloading. Under these conditions, the coincidence counting rate (i.e., front two counters in coincidence and the last counter in anti-coincidence) as a function of discriminator voltage on the front two counters yielded plateaus which were quite adequate.

A proton transmitted through all three counters gives pulses having a maximum spread (jitter) of $\frac{1}{5}$ microsecond. The subsequent choice of 2-μsec gates for coincidence was made on the basis of the requirement of positive operation of the mixer-scaler.

Electronics

Pulses from proportional counters 1 and 2 generated 2-μsec gates which were fed into the coincidence channels of the mixer-scaler, while pulses from proportional counter 3 generated 3-μsec gates, straddling the 2-μsec gates of channels 1 and 2 in time, which were fed into the anti-coincidence channel of the mixer-scaler. Counting rates were low enough to make pile-up or dead-time corrections completely unnecessary. It was possible to monitor the accidental coincidences by splitting the

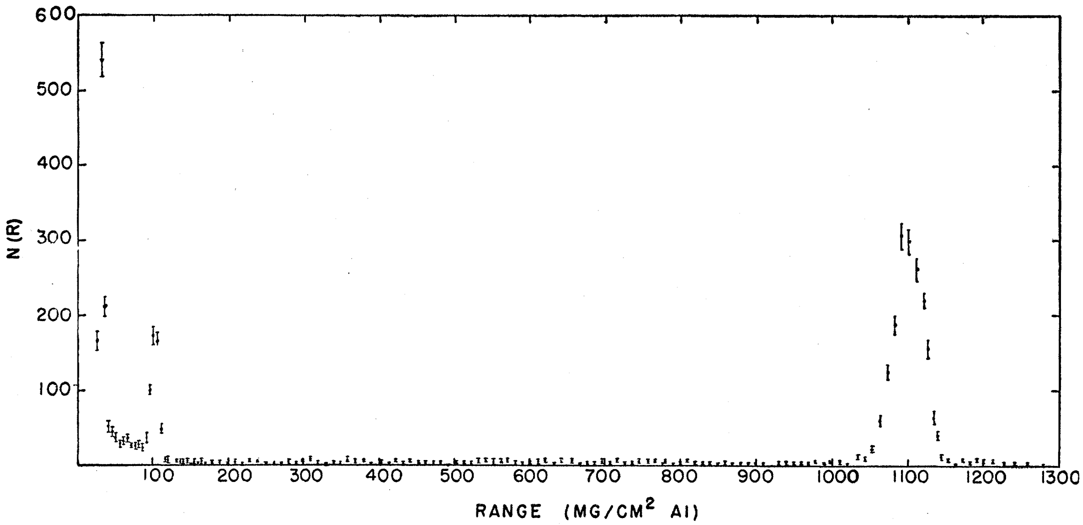

FIG. 4. Range spectrum of charged particles observed at 30°.

output of linear amplifier 2, introducing an arbitrary delay into another variable gate unit, and feeding this delayed gate into a separate mixer-scaler with the undelayed gates of the other channels. The mixer-scalers were effectively gated to count only during the beam by feeding a "beam gate" into the third coincidence channel.

Solid Angle Calculations

The solid angle calculations have been made.[7] The correction due to a finite beam width has been calculated and found negligible, and corrections due to the rapid variation of cross section with angle have been made.

Errors Affecting the Cross Section Measurement

The differential cross section for an observed process is proportional to

$$d\sigma/d\Omega \approx TN/[PCV(\Omega S)], \tag{1}$$

where T is the temperature in degrees absolute, P is the pressure of the target gas, C is the capacitance of the condenser which is charged by the collected beam to a voltage V, ΩS is the solid angle times scattering length determined by the slit geometry, and N is the number of particles detected.

In the course of a run one measured the number of particles which stopped in an aluminum foil of thickness ΔR, i.e., each point of the spectrum represents $(dN/dR)\Delta R$. Hence, to determine the number of particles scattered into the detector as a result of a given reaction, one measured the area under the peak and divided this by ΔR:

$$N = \frac{1}{\Delta R}\int \frac{dN}{dR}\Delta R dR. \tag{2}$$

[7] J. Benveniste, University of California Radiation Laboratory Report UCRL-1689, (thesis) (unpublished).

Area measurements were made by fitting theoretically derived line shapes to the experimental points as in Fig. 3. It is noted that Eq. (1) becomes

$$d\sigma/d\Omega_a \approx T/[PCV(\Omega S)\Delta R]. \tag{3}$$

Combining the several contributing errors quadratically, the estimated error in the measurement of the absolute differential cross section as a result of contributions from all sources except statistics was ± 2.6 percent at 30°. Further, the estimated error of the relative cross section measurement as a result of contributions from all sources of error except statistics was ± 1.5 percent at 30°.

RESULTS AND CONCLUSIONS

Search for Excited He^4 Level

A typical spectrum is shown in Fig. 4. The peak at 1100 mg/cm^2 represents elastically scattered protons; that at 30 mg/cm^2, elastically scattered alphas. The interesting group of particles is that at 100 mg/cm^2. As will be shown, they are deuterons from the reaction

$$p + \mathrm{He}^4 \rightarrow \mathrm{He}^3 + d. \tag{4}$$

If they had proved to be protons, they would have indicated the existence of an excited level of the helium nucleus.

Identification of Low-Energy Peak

From the kinematics of the reaction (4) the energy of the deuterons is

$$E_d = \frac{2}{25}E\cos 2\phi + \tfrac{3}{5}(\tfrac{4}{5}E - 18.3) + \frac{4}{25}E\left[6\left(1-\frac{22.9}{E}\right)\right]^{\frac{1}{2}} \times\cos\phi\left[1-\frac{\sin^2\phi}{6(1-22.9/E)}\right]^{\frac{1}{2}}, \tag{5}$$

TABLE I. Deuteron energies in the reaction $p+He^4 \to He^3+d$. The column headed E_d lists the deuteron energies expected from the kinematics of the reaction. The column headed E_d(obs) contains the energies of the particles having the observed ranges, assuming the particles to be deuterons. ϕ is the angle which the scattered deuteron makes with the beam direction.

ϕ_{lab}	E_d	E_d(obs)
15.0	12.41±0.15	12.40±0.05
22.5	11.47±0.15	11.25±0.05
30.0	10.54±0.15	10.35±0.05
37.5	9.29±0.1	9.32±0.07
45.0	7.94±0.1	7.85±0.07
52.5	6.56±0.1	6.27±0.1
60.0	5.25±0.1	5.40±0.1

where E is the energy in the laboratory system of the incident protons at the center of the scattering chamber, and ϕ is the angle which the scattered deuteron makes with the beam direction.

E may be quite accurately determined in the following way: The range of the elastically scattered protons includes, in addition to the aluminum absorber, a distance (equal to the radius of the scattering chamber) in the target gas, the entrance foil to the counter telescope, and the counter filling gas.

The nonaluminum portion of the range was equivalent to about 14 mg/cm² aluminum, and was the least precisely known contribution to the total range. However, the energy of the elastic protons was not sensitive to small uncertainties in the total range and was considered to be practically as precisely known as the range-energy data from which it is derived.[8] Then from the relation expressing the energy of protons elastically scattered from helium,

$$E_p=(E/25)[16+\cos 2\phi+8\cos\phi\{1-\tfrac{1}{16}\sin^2\}^{\frac{1}{2}}\phi], \quad (6)$$

one can find E. From range measurements of the elastic peak at seven different angles, $E=31.5\pm0.1$ Mev.

On the other hand, the precision with which the energy of the low-range particles was known depended strongly on how well known the nonaluminum portion of the range was known. It turned out that in addition to the calculations based on distances, gas pressures, and rates of energy loss, one had an excellent, independent, experimental check of this contribution. The energy of the elastically scattered alpha-particle is

$$E_{He^4}=(16/25)E\cos\phi, \quad (7)$$

and was as well known as E. The experimentally observed range of the alpha-particle must be that corresponding to E_{He^4} from the range-energy curves. Thus, the difference between this range and the aluminum absorbers plus counter foils was the nonaluminum contribution to the range.

In Table I, the column headed E_d lists the expected deuteron energies from reaction (4) as given by Eq. (5). The column headed E_d(obs) contains the energies of the particles having the observed ranges, assuming the particles to be deuterons. The agreement is seen to be quite good. However, it still remained to be seen whether the scattering data were explainable on the basis of inelastically scattered protons.

If there were an excited level of the helium nucleus,

$$p+He^4 \to Li^{5*} \to He^{4*}+p', \quad (8)$$

the inelastic proton energy would have the angular distribution

$$E_{p'}=\frac{E}{25}\cos 2\phi+\frac{16}{25}E\left(1-\frac{5}{4}\frac{\epsilon}{E}\right)+\frac{8}{25}E\left[1-\frac{5}{4}\frac{\epsilon}{E}\right]^{\frac{1}{2}}\cos\phi\left[1-\frac{\sin^2\phi}{16(1-5\epsilon/4E)}\right]^{\frac{1}{2}}, \quad (9)$$

where ϵ is the energy above the ground state of the presumed excited state. To determine a consistent value for ϵ, one measures the range of the low energy group of particles at some angle, say 45°. From the range-energy relations one can determine the energy of a proton having the range of the particles observed; then inserting this $E_{p'}$ and the previously determined E into Eq. (9), one finds that $\epsilon=21.2$ Mev. The column headed $E_{p'}$ in Table II lists the results of Eq. (9), using this value of ϵ. $E_{p'}$(obs) is the energy of a proton having the range experimentally measured. The discrepancy is seen to be quite significant.

Another identification of the low energy peak may be made by comparing the dE/dx of these particles with that of the elastic protons. It will be recalled from the description of the operation of the proportional counter telescope that all counted particles entering the counters have the same residual range. The dE/dx of deuterons and protons having the same residual range differs by about 30 percent; hence, one expects a difference of about 30 percent in the pulse height observed in the front counters. Inserting enough absorber before the counter telescope to see just the elastic protons and plotting counting rate *vs* discriminator voltage on the front two counters yields the curve of Fig. 5(a). Figure 5(b) is a similar curve obtained with the absorber required to see just the particles of the low energy peak.

TABLE II. Proton energies in the reaction $p+He^4 \to He^{4*}+p'$. The column headed $E_{p'}$ lists the expected energies of the scattered protons if there exists an excited state of the He^4 nucleus at 21.2 Mev. The column headed $E_{p'}$(obs) contains the energies of protons having the ranges experimentally measured.

ϕ	$E_{p'}$	$E_{p'}$(obs)
15	8.15±0.15	9.20±0.05
23.5	7.72±0.15	8.30±0.05
30	7.16±0.15	7.70±0.05
37.5	6.51±0.10	6.93±0.05
45	5.77±0.10	5.80±0.07
52.5	5.02±0.10	4.65±0.10
60	4.28±0.10	4.03±0.10

[8] J. H. Smith, Phys. Rev. **71**, 32 (1947).

The dE/dx of the particles of the low energy peak is seen to be about 25 percent larger than that of protons, and hence these particles are certainly deuterons.

Consideration of the Continuum

We now turn our attention to the continuum on the low range side of the deuteron peak.

The most likely sources for a continuum of protons of the observed range are the reactions

$$p+\mathrm{He}^4\rightarrow\mathrm{He}^3+n+p', \tag{10}$$

$$p+\mathrm{He}^4\rightarrow\mathrm{H}^3+p''+p', \tag{11}$$

where p' and p'' represent inelastically scattered protons. It is clear that observation of p' will yield information on the states of the $(n+\mathrm{He}^3)$ and $(p''+\mathrm{H}^3)$ systems which may also be referred to as the excited states of the He^4 nucleus (He^{4*}). For example, if the wave function describing the $(n+\mathrm{He}^3)$ system became unduly large for a given relative momentum, the probability that the proton (p') would come off with a related momentum (given by the conservation laws) would be correspondingly large. Reactions (10) and (11) may be rewritten

$$p+\mathrm{He}^4\rightarrow\mathrm{He}^{4*}+p', \tag{12}$$

$$\mathrm{He}^{4*}\rightarrow\mathrm{He}^3+n, \tag{13}$$

$$\mathrm{He}^{4*}\rightarrow\mathrm{H}^3+p'', \tag{14}$$

with somewhat less probability for the mode

$$\mathrm{He}^{4*}\rightarrow\mathrm{He}^4+\gamma. \tag{15}$$

The spectrum of p' should be a peak with an energy spread commensurate with the breadth of the excited He^4 level, while that of p'' would appear rather spread out due to the motion of its parent He^{4*}, and its area would be somewhat less than half that of the p' peak. If there were an excited level, therefore, one could expect to be able to detect a bump on the continuum. Actually, no bump (exhibiting the proper range-angle dependence) was discernible; therefore, all one can do is calculate a maximum cross section for the formation of an excited state by estimating the maximum area of a peak which might have been masked by the counting statistics. A calculation of this sort of the spectrum obtained at 22.5° (see Fig. 3) yields

$$\frac{(d\sigma)}{(d\Omega)_{\mathrm{lab}}}\leqslant 0.34 \text{ millibarn/steradian.}$$

Unfortunately, the intrusion of elastically scattered alpha-particles masks some of the low range portion of the small angle spectra where it would be desirable to hunt for a low energy group of protons. For example, at 22.5° it would not be possible to detect a small group of protons having a range less than 50 mg/cm^2 (5 Mev). Reference to the kinematical equation shows

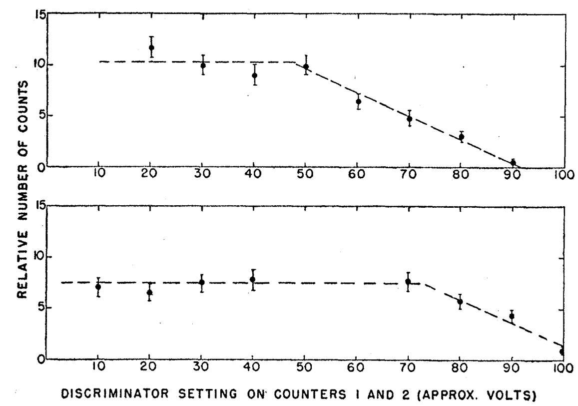

FIG. 5(a). dE/dx measurement of elastically scattered protons. The abscissa is the discriminator setting in volts. The pulse-height distribution in the front two counters of the proportional counter telescope yields a measurement of dE/dx which, with the known residual range, serves to identify the particle. (b) dE/dx measurement of the particles of the low energy peak.

that a group of protons of this energy would result if there were a level in He^4 at 23.3 Mev.

Finally, making the transformation to the c.m. system, we may conclude that the differential cross section for detecting an excited level in He^4 as high as 23.3 Mev above the ground state by means of inelastically scattered protons is less than 0.1 mb/sterad at 45° (c.m.).

If there exists a single excited level of the He^4 nucleus in the energy region and with the cross section indicated by Argo *et al.*,[5] a low energy proton group would have been seen. The absence of such a low energy proton group confirms and extends the findings of some other workers in this field.

The most natural extension to the work of the Los Alamos group was performed by Falk and Phillips,[9] who extended the excitation curve for the reaction $\mathrm{H}^3(p,\gamma)\mathrm{He}^4$ to incident proton energies of 3.4 Mev. No resonance is observed; rather, the γ-ray yield continues to increase in the way predicted by the calculations of Flowers and Mandl.[10] However, the existence of a weak (up to 30 percent), broad resonance superposed on the rising yield curve is not ruled out.

Allred,[11] in studying the reaction $\mathrm{He}^3(d,p)\mathrm{He}^4$, sought a group of low energy protons as evidence for the existence of an excited He^4 level. He reports an upper limit to the cross section for observing these protons at a c.m. angle of 102.7° of 0.2×10^{-27} cm^2, implying the absence of an excited He^4 level below 20.9 Mev.

Finally, the continued high level of the inelastically scattered protons, as far as we have been able to see, requires the existence of at least two levels, each having a half-width of about a million volts and a separation

[9] C. E. Falk and F. C. Phillips, Phys. Rev. **83**, 468 (1951).
[10] B. H. Flowers and F. Mandl, Proc. Roy. Soc. (London) **A206**, 131 (1951).
[11] J. C. Allred, Phys. Rev. **84**, 695 (1951).

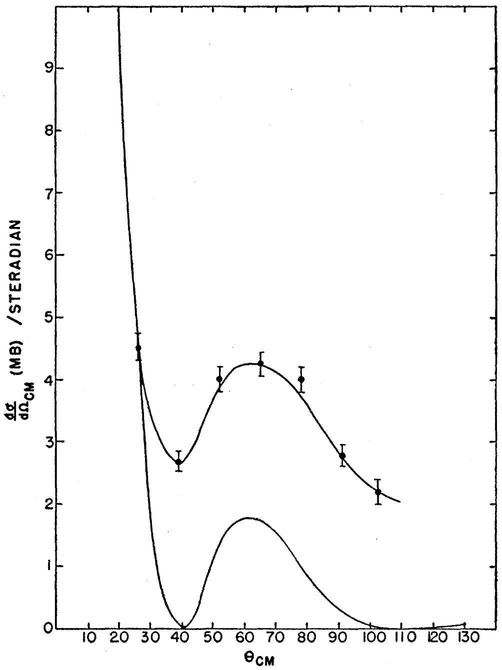

FIG. 6. Comparison of experimental yield of deuterons from the reaction $p+He^4 \rightarrow He^3+d$ with that predicted by the Butler theory. The curves have been normalized at the smallest angle observed, since the assumptions of the Butler theory are expected to be most valid for interactions involving small momentum transfers.

of about a million volts. However, whether there are just two levels or several closely spaced levels contributing to this region cannot be decided from this work.

Pick-Up Deuterons

It is interesting to compare the angular dependence of the differential cross section for the production of deuterons from the reaction $He^4(p, d)He^3$ with that predicted by the Butler theory.[12] Butler considers an incident deuteron to be stripped, with the neutron, say, deposited in the target nucleus. He finds that the angular distribution of the remaining proton depends in a distinctive way on the angular momenta and parities of the states of the initial and final nuclei. To date, the theory has been applied successfully to the assignment of angular momentum and parity to one nucleus when that of the other was known.[13–18]

Here, we are concerned with the inverse reaction, so we invoke the principle of detailed balancing which tells us that in the c.m. system, the differential cross section for the inverse reaction is directly proportional to that for the direct reaction; further, the proportionality factor includes only statistical weights, so that the angular dependences of the two differential cross sections are the same. Hence, applying the Butler theory to the reaction inverse to ours, $He^3(d, p)He^4$ will yield to us directly the angular dependence of the differential cross section for observation of our deuterons. Since the spins and parities of the initial and final nuclei are known, comparison of the theoretical and experimental data might be considered a check of the theory; or if the theory is considered to be a reasonable approximation to the truth, and if one believes the principle of detailed balancing, the theory may serve to fix the "size" of the He^3 nucleus.

If the captured neutron carries with it zero angular momentum, Butler gives

$$\sigma(\theta) \approx \left\{ \frac{1}{K^2+a^2} - \frac{1}{K^2+(a+b)^2} \right\}^2 \times \left| \left[\left(\kappa_s + \frac{\partial}{\partial r} \right) \left\{ \left(\frac{r}{2} \right)^{\frac{1}{2}} J_{\frac{1}{2}}(Zr) \right\} \right]_{r_0} \right|^2 \quad (17)$$

$$\approx \left\{ \frac{1}{K^2+a^2} - \frac{1}{K^2+(a+b)^2} \right\}^2 \left| \frac{\kappa_s}{Z} \sin Zr_0 + \cos Zr_0 \right|^2,$$

where

$$K^2 = \tfrac{1}{4} K_d^2 + k_{ps}^2 - K_d k_{ps} \cos\theta,$$

$$Z^2 = K_d^2 + k_{ps}^2 - 2K_d k_{ps} \cos\theta,$$

$$K_d^2 = (2M_d/\hbar^2) E_d, \quad k_{ps}^2 = (2m_p/\hbar^2) E_p,$$

$$i\kappa_s = k_{ns}, \quad k_{ns}^2 = (2m/\hbar^2) E_n,$$

$$E_n = E_d - \epsilon - E_p,$$

and ϵ is the deuteron binding energy; $a = 0.23 \times 10^{13}$ cm^{-1} and $b = 1.4 \times 10^{13}$ cm^{-1} are parameters of the internal wave function of the deuteron; r_0 is the interaction radius between the neutron and the target nucleus (He^3). Figure 6 is a comparison of the theoretical curve for $r_0 = 4.2 \times 10^{-13}$ cm, with a curve drawn through the experimental points. In spite of the somewhat meager experimental data, the fit, as determined by the coincidence of maxima and minima, is considered rather good. The increasing divergence of the two curves at large angles is not unexpected in view of the meaning of r_0. By definition, the proton does not penetrate the region of radius r_0 about the target nucleus; however, this assumption is expected to be violated in collisions involving high momentum transfers, i.e., large scattering angles.

Butler and Symonds[19] compared the theoretical angular distributions for the reactions $T(d, n)He^4$ and

[12] S. T. Butler, Proc. Roy. Soc. (London) **A208**, 559 (1951).
[13] L. Schecter, Phys. Rev. **83**, 695 (1951).
[14] Burrows, Powell, and Rotblatt, Proc. Roy. Soc. (London) **A209**, 461 (1951).
[15] Burrows, Powell, and Rotblatt, Proc. Roy. Soc. (London), **A209**, 478 (1951).
[16] Burrows, Gibson, and Rotblatt, Proc. Roy. Soc. (London), **A209**, 489 (1951).
[17] Burge, Burrows, Gibson, and Rotblatt, Proc. Roy. Soc. (London) **A210**, 534 (1952).
[18] W. M. Gibson and E. E. Thomas, Proc. Roy. Soc. (London) **A210**, 543 (1952).

[19] S. T. Butler and J. L. Symonds, Phys. Rev. **83**, 858 (1951).

$He^3(d, p)He^4$ for 10.5 deuterons with the experimental curves of Brolley *et al.*[20] and Allred[11] and found $r_0 = 4.3\times10^{-13}$ cm. The difference is not considered significant.

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States of Solid Methane as Inferred from Nuclear Magnetic Resonance*

KAZUHISA TOMITA
Department of Physics, Faculty of Science, University of Kyoto, Kyoto, Japan
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As there are different points of view concerning the molecular dynamical states in solid methane, especially concerning the λ-transition at 20.42°K, we have investigated the manner in which the new method of nuclear magnetic resonance throws light on the problem. Both the one-parameter theory and the method of perturbation were applied to the analysis of the proton resonance data given by Thomas, Alpert, and Torrey.

The characteristic time τ_c for molecular reorientation was estimated from the spin-lattice relaxation data, and it was found that above 20°K molecules may be considered to reorient against some fixed hindrances due to neighboring molecules. At about 65°K a marked alteration is present in the dominant mechanism of the above reorientation, which is considered to be either correlated or independent molecular rotation. However, below 20°K most of the molecules seem to be in the ground state of rotational oscillation and occasionally (about 10^7 times per second) tunnel or flip to neighboring equivalent orientations. It was proposed that we should discriminate between two pictures of the local magnetic field, possibly in relation to the frequency of resonance. This idea was confirmed by reproducing the line width data.

A perturbation calculation, assuming the $F\bar{4}3m$ arrangement of the molecules and taking account of the above situation, gave the entire shape of the absorption line, which was in close agreement with experimental data observed in the lowest temperature range.

I. INTRODUCTION

ABOUT ten years ago Clusius, Popp, and Frank[1–4] carried out a series of thermal measurements on solid methane and found a λ-change in the specific heat at 20.42°K. Although many authors have considered the mechanism of the λ-transition since that time, no decisive conclusions seem to have been obtained.

According to the results of x-ray analysis[5] the carbon atoms form a face-centered cubic lattice (see Fig. 1), and scarcely any difference was observed[6] between the lattice structures above and below the λ-point. The only remaining degrees of freedom being those of molecular reorientation, Clusius suggested that the transition should be a rotational one as suggested by Pauling.[7]

Because x-ray analysis could not fully locate the hydrogen atoms in the molecule, it seemed hopeless to verify the molecular rotation directly, and some indirect verifications have been attempted. These are connected with the thermal behavior of solid methane both above and below the λ-point.

Eucken examined[8] the rotational part of the specific heat and obtained a value of about 3 cal/mole deg between the λ-point and the fusion point, which seems to suggest that the molecules are rotating freely in the classical sense. Theoretical calculations showed,[9] however, that the free rotation of a single molecule may become classical only above 50°K.

At the lowest temperature the value of the zero-point entropy has been used as a starting point. Free rotation is excluded because it would require $S=S_0+(2/16)R\ln5$, which is definitely greater than the observed value $S_0=R\ln16$. Since $2^4=16$, Clusius proposed a random distribution of proton spin over the whole crystal, which implies that a spin is correlated equally with spins in the same molecule and with those in the other molecules. Were this the case, the resultant spin of a molecule could easily be changed, and sublimation from such a state should give a single species having

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