Ionization Measurement of $B^{10}(n,\alpha)Li^{7}$†

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A thin layer of boron on the wall of a cylindrical ionization chamber exposed to slow neutrons has been used to study the ionization produced by the recoiling particles of the transmutation $B^{10}(n,\alpha)Li^{7}$. Polonium alpha-particles were used for calibration, and the ionization electron pulses were amplified and photographed on an oscilloscope screen. Calculated peak shapes taking into account wall effect and layer thickness were fitted to the experimental number pulse-height curves, and from this information the ionization values were deduced. The ratio of ionization produced by the He^{4} and Li^{7*} recoils was observed to be 1.878 ± 0.014 as compared to the energy ratio 1.7529 determined from conservation of momentum. This difference can be accounted for with the assumption that the average energy per ion pair varies with velocity. The implications of this assumption are discussed.

INTRODUCTION

THE average energy expended by an energetic charged particle in producing an ion pair has been known to be different for different gases. In most gases, it also varies with the type and velocity of the ionizing particle. However, there has been some evidence that the average energy per ion pair is a constant in argon.[1–3] Our previous experiments with (n,p) reactions have seemed to support this view.[4] In extending this work to (n,α) reactions we have found definite evidence that the average energy per ion pair varies with velocity. Similar results have been obtained by Hanna[5] and Cranshaw and Harvey.[6]

EXPERIMENTAL METHOD

The cylindrical ionization chamber used in the experiment is similar in construction to the chamber described in the report of our previous experiments with (n,p) reactions[4] except that the diameters of the cylinder and axial wire were changed to $2\frac{3}{8}$ in. and 0.020 in., respectively. The length of the region of electron collection is 8 in. For calibration a polonium alpha-particle source, deposited on platinum foil by the Erbacher method,[7] was attached to a copper flap so that it could be exposed to the collecting region through a small hole in the wall of the high voltage cylinder or tilted out of position. The amplifier and recording system was the same as that described in our previous experiments in this laboratory.

After outgassing the chamber in a vacuum of 5×10^{-6} mm Hg, two atmospheres of argon and one-tenth atmosphere of CO_2 were admitted. This gas mixture was purified by circulating through calcium hydride[8] maintained at 250°C until saturation was achieved.

The slow neutrons originated from a 200 mC radium-beryllium source surrounded by a cylinder of paraffin and placed below the chamber.

The boron layer was made when BF_3, prepared from calcium fluoborate[9] (enriched to 96 percent in the B^{10} isotope), was admitted to the chamber and circulated through the hot calcium hydride. The formation of a boron layer on the electrode wall was made evident by the disappearance from the pulse distribution of pulses arising from the combined ionization of both the alpha-particle and lithium recoil ion due to disintegrations occurring in the gas and the appearance of pulses due to the separate ionizations of the alpha-particles or the lithium recoil when the transmutations occur on the wall. Petree, Johnson, and Miller[10] also have reported the observation of pulses from the disintegration of boron atoms deposited on the wall of a counter containing BF_3.

Complete saturation was not achieved until the BF_3 had disappeared (by continued purification). Since only one-twentieth atmosphere BF_3 had been admitted to the chamber, it seemed likely that the boron layer might be very thin and useful for a comparison of the ionization of the alpha-particle and lithium recoil. If uniformly deposited over all the interior surfaces of the chamber, this layer was estimated to have a thickness of about 20 μg per square centimeter.

The chamber was disassembled and all parts cleaned except the inside surface of the cylinder in the region of electron collection. The center wire was replaced and the chamber was again assembled, outgassed, and filled with two atmospheres of argon and one-tenth atmosphere of CO_2. After purification of the gas mixture, the maximum pulse height for polonium alpha-

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[9] Allocated by the Isotopes Division of the AEC. The procedure for the preparation and purification of BF^3 is described by J. A. Bistline, Rev. Sci. Instr. **19**, 842 (1948).
[10] Petree, Johnson, and Miller, Phys. Rev. **83**, 1148 (1951).

particles changed less than $\frac{1}{2}$ percent when the chamber voltage was varied from 300 to 1600 volts. This gives us confidence that there was substantially no loss of charge due to electron capture or recombination in this region of voltages. The chamber was operated at 1200 volts, well below the proportional region.

A pulse-height distribution was obtained for polonium alpha-particles and for the alphas and lithium recoils from the $B^{10}(n,\alpha)Li^7$ reaction. The amplifier gain used for pulses from the $B^{10}(n,\alpha)Li^7$ reaction was 4.023 times the gain used for the polonium alpha-particle pulses.

RESULTS

Figure 1 displays the pulse-height distribution of 350 polonium alpha-particles. Figures 2(a) and 2(b) show the size distribution of 1050 pulses obtained by exposing the chamber to slow neutrons. The peaks in order of increasing pulse height are ascribed to excited lithium recoil, lithium recoil in the ground state, alpha-particle associated with excited lithium, and alpha-particle associated with lithium in the ground state.

The shape of the pulse-height distribution curve resulting from collection of the ionization electrons for particles emitted from a source located on the wall of an ionization chamber depends upon chamber geometry, particle range, and source thickness. The measurement of the relative ionizations of two particles which have different ranges or which come from sources of different thickness requires the determination of the effect of these factors on the distribution curves. The maximum pulse height depends only upon chamber geometry and the total ionization produced by the particle, except for variations produced by ionization straggling and amplifier noise, and it is the leading edge representing the maximum pulse height of the distribution which must be accurately located.

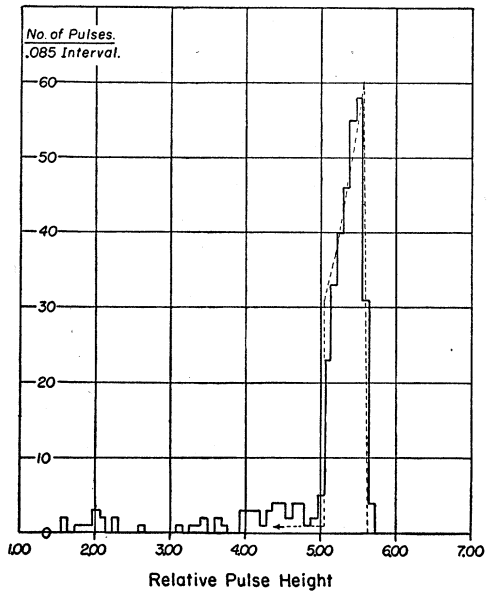

FIG. 1. Pulse-height distribution from a wall source of polonium alpha-particles in a cylindrical ionization chamber. The dotted curve shows the calculated distribution.

The effect of particle range in a cylindrical chamber is twofold. First, the pulse height varies with the angular orientation of the path of ionization, and this variation increases with the path length. Secondly, the number of particles colliding with the curved chamber wall varies with particle range, and the number of such collisions helps to determine the slope of the leading edge of the pulse-height distribution curve. The effect of source thickness is to widen the energy distribution of the particles. For a group of particles of energy E_0 emitted isotropically throughout a layer of thickness T, the width of the peak in their energy distribution as they leave the layer is somewhat larger than T/R where R is the range of the particles measured in the layer. Fitting a calculated distribution curve to the experimental distribution provides an accurate determination of the leading edge of the distribution and also gives an indication of the approximate thickness of the source.

The pulse height P resulting from the collection of ionization electrons which originate along a path extending from r_1 to r_2 (where r_1 and r_2 are measured radially from the cylinder axis) in a cylindrical chamber with inner and outer radii a and b, respectively, and with capacitance C is given by

$$P=\frac{1}{C\log(b/a)}\int_{r_1}^{r_2}\rho(r)\log(r/a)dr, \tag{1}$$

where $\rho(r)$ gives the radial variation in initial ionization electron density along the path. Since $\rho(r)$ varies in a complicated way with the position of the ion path, the calculation of the pulse-height distribution for particles with extended ranges cannot be made explicitly. We define a distance r' such that if all the ionization electrons of total charge Q contained in an extended path were initially formed at this distance from the cylinder axis, the resulting pulse height would be the same as that produced by the actual extended path of ionization. Then,

$$P=\frac{Q}{C}\frac{\log(r'/a)}{\log(b/a)}. \tag{2}$$

For an ion path in the outer portion of the chamber, r' may be approximated by the distance from the cylinder axis to the "center of gravity" of the initial ion distribution. In this experiment b/a is large, the source is on the outer electrode wall, and the range of the particles is considerably less than the chamber radius. With these conditions the approximation introduces negligible distortion into the distribution, particularly in the region of most interest near the leading edge.

If the center of gravity lies a distance ϵR_0 from the origin of an ionization path of length R_0, the distance

from the center of gravity to the cylinder axis is given by

$$r'=[\epsilon^2R_0^2\sin^2\theta\sin^2\phi+(b-\epsilon R_0\cos\theta)^2]^{\frac{1}{2}}, \quad (3)$$

where we have used a spherical polar coordinate system in which the origin is located at the particle source on the cylindrical wall, the polar axis lies along a radius of the cylinder, and the plane $\phi=0$ contains the cylinder axis. The distribution in space of particles emitted from an isotropic source is given by

$$dN=(N_0/2\pi)\sin\theta d\theta d\phi, \quad (4)$$

where N_0 is the total number of particles emitted into the forward hemisphere. By use of Eqs. (2)–(4) the distribution may be written in terms of σ and ϕ, where σ is the ratio of pulse height P to the maximum pulse height of the distribution. Thus,

$$dN=\frac{N_0a^2\log(b/a)}{2\pi b\epsilon R_0}\left(\frac{b}{a}\right)^{2\sigma}\left[1-\cos^2\phi\left\{1-\left(\frac{b}{a}\right)^{2\sigma-2}+\frac{\epsilon^2R_0^2}{b^2}\sin^2\phi\right\}\right]^{-\frac{1}{2}}d\sigma d\phi. \quad (5)$$

The differential distribution in relative pulse height $dN/d\sigma$ over the acceptable range of σ may then be obtained by numerical integration over ϕ.

The dotted curve in Fig. 1 shows the distribution for polonium alpha-particles calculated from Eq. (5) for the conditions of this experiment in which $b=3.02$ cm, $a=0.0254$ cm, $R_0=2.0$ cm, and ϵ is set equal to 0.6. The slope on the leading edge is accounted for by wall effect. The background of pulses below the peak also results from those particles which collide with the chamber wall so that only a portion of their energy is lost in the chamber gas. The leading edge of the calculated distribution is 1.2 percent (corresponding to 64 kev) higher than the position of the peak. The 10 percent width of the peak is completely accounted for by the variation in position of ionization. (The root-mean-square variation due to noise is just 12 kev.) The agreement between calculated and experimental distributions indicates that source thickness does not contribute significantly to the width of the peak. Hence the alpha-particles are essentially monoenergetic as they leave the source and do not show the spread in energy sometimes observed with polonium sources plated on thin foils.[11]

A similar calculation shows that the spread in pulse heights due to variation in position of the ionization for 1.47-Mev alpha-particles is 1.6 percent of the maximum pulse height. The observed width of the peak for the 1.47-Mev alpha-particles from the $B^{10}(n,\alpha)Li^{7*}$ reaction is about 5 percent, indicating that its shape is determined mostly by the finite thickness of the boron layer and not by the effect of different locations of the ionization discussed above. Similarly, the observed width of the 0.84-Mev Li* peak is about 8 percent while the calculated width due to variation in position of ionization is less than 1 percent.

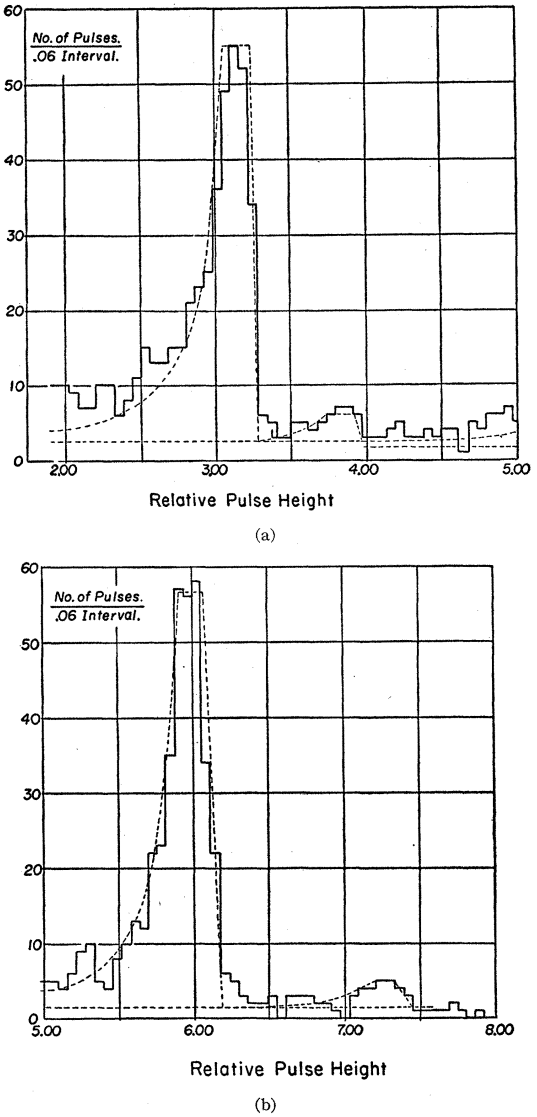

FIG. 2. (a) Pulse-height distribution of Li recoils from a thin boron layer on the ionization chamber wall from the reaction $B^{10}(n,\alpha)Li^7$; (b) pulse-height distribution of alpha-particles from a thin boron layer on the ionization chamber wall from the reaction $B^{10}(n,\alpha)Li^7$.

In order to calculate the effect of source thickness on the pulse distribution, we must assume an analytic relation between energy and residual range of the particle. Assuming the residual range to be proportional to E^n, a particle with initial energy E_0 at depth x in the layer leaves the surface with energy E given by

$$E=E_0[1-x/(R\cos\theta)]^{1/n}, \quad (6)$$

where R is the range of the particle measured in the layer material and θ is the angle between the path of

[11] Coche, Faraggi, Avignon, and Haissinsky, J. phys. et radium **10**, 312 (1949); Phys. Rev. **75**, 1963 (1949).

the particle and the normal to the layer surface. For an isotropic source, the distribution in space of the particles emitted from an elemental layer of thickness dx at depth x is given by

$$dN=(N_0/T)\sin\theta d\theta dx, \tag{7}$$

where N_0 is the number of particles emitted into the forward hemisphere by the entire layer of thickness T. If we write E' for E/E_0, the distribution in energy of these particles is given by

$$dN=(N_0nx/TR)E'^{n-1}(1-E'^n)^{-2}dE'dx. \tag{8}$$

Integration over x gives

$$dN/dE'=(N_0nR/2T)E'^{n-1} \quad \text{for} \quad E'^n\geq 1-T/R, \tag{9}$$

$$dN/dE'=(N_0nT/2R)E'^{n-1}(1-E'^n)^{-2} \quad \text{for} \quad E'^n\leq 1-T/R. \tag{10}$$

For alpha-particles of energy 1 to 2 Mev, the exponent n in the range-energy relation is approximately unity, for which Eqs. (9) and (10) become

$$dN/dE'=N_0R/2T \quad \text{for} \quad E'\geq 1-T/R, \tag{11}$$

$$dN/dE'=(N_0T/2R)(1-E')^{-2} \quad \text{for} \quad E'\leq 1-T/R. \tag{12}$$

The superposition of the spread in pulse height for monoenergetic particles due to variation in position of ionization as determined by Eq. (5) upon the distribution in energy (and hence in total ionization) of particles emitted from a layer of finite thickness is complicated. However, we note that for particles whose range is small compared to the chamber radius the total spread in pulse height due to variation in position of ionization is small, and the differential distribution of pulses over this small interval may be considered constant. With this approximation the superposition of the two factors contributing to peak width can be readily accomplished graphically.

The dotted curves of Figs. 2(a) and 2(b) show the calculated pulse distributions in which variation in position of ionization and source thickness have been taken into account as described above. The rms variation due to noise and ionization straggling is 13 kev, and recoil from the gamma-ray contributes an additional 15-kev spread for the excited lithium nuclei. While these contributions to the shape of the pulse distributions are not included in the dotted curves, they have been taken into account in locating the most probable position of the leading edge of each peak. The best fit between experimental and calculated distributions occurs for an assumed equivalent layer thickness of 55 kev for alpha-particles and 64 kev for lithium recoils. Taking 0.8 as the stopping power of boron for alpha-particles, this corresponds to a layer of 30 $\mu g/cm^2$.

The ionizations relative to polonium alpha-particles, deduced from the position of the leading edge of the pulse distributions as determined by fitting experimental and calculated curves, are: $I_\alpha=0.3286\pm0.0032$; $I_{\mathrm{Li}}=0.1756\pm0.0024$; $I_\alpha^*=0.2726\pm0.0018$; $I_{\mathrm{Li}}^*=0.1451\pm0.0012$. The ratios of alpha ionization to lithium recoil ionization are $I_\alpha/I_{\mathrm{Li}}=1.872\pm0.028$; $I_\alpha^*/I_{\mathrm{Li}}^*=1.878\pm0.014$.

The ratio of ground-state transitions to excited state transitions is 0.063 ± 0.009. This is in good agreement with the ratio 0.067 found by Bøggild[12] from about 400 cloud-chamber tracks and with the value 0.062 ± 0.001 found by Hanna,[5] but is appreciably higher than the value 0.0427 ± 0.0015 obtained by Cuer and Lonchamp[13] from the observation of more than thirty thousand tracks in nuclear plates.

DISCUSSION

The energy ratio E_α/E_{Li} can be precisely calculated from the atomic masses by the conservation of momentum and is 1.7529. This is considerably different from the observed ionization ratio 1.878 ± 0.014. Consequently, it is necessary to assume that the average energy per ion pair depends upon velocity or charge of the ionizing particle. The ionization ratio observed in this experiment is in good agreement with the ratio 1.89 ± 0.02 reported by Stebler, Huber, and Bichsel[14] and the ratio 1.841 ± 0.025 calculated from measurements reported by Hanna.[5] On the other hand, Jesse[3] measured the total ionization produced in argon by the $B^{10}(n,\alpha)Li^{7*}$ reaction and obtained the correct reaction energy within 0.4 percent probable error by assuming a constant energy per ion pair for both the Li^7 nuclei and the alpha-particles.

The reaction energy for $B^{10}(n,\alpha)Li^7$ is known to be 2.793 ± 0.003 Mev[15] from the precisely measured Q values for $B^{10}(p,\alpha)Be^7$ and $Li^7(p,n)Be^7$. When the lithium nucleus is left in the excited state, the energy released is less than the value just quoted by 479 ± 1 kev, this being the energy of the gamma-ray emitted when the excited nucleus makes the transition to the ground state.[16] Thus, the energy of each of the particles is known, and from our ionization measurements the average energy per ion pair relative to a polonium alpha-particle (5.2984 Mev) may be calculated.

Table I shows the energy of each particle and its ionization and average energy per ion pair relative to polonium alpha-particles. Also shown are Hanna's measurements of the average energy per ion pair for the 1.473-Mev alpha-particle and the 0.841-Mev Li ion relative to Pu^{239} 5.159-Mev alpha-particles. Hanna also measured the combined ionizations of the alpha-particle and Li ion both for the ground state and the excited state of Li and found that his measurements were

[12] J. K. Bøggild, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **23**, No. 4 (1945).
[13] P. Cuer and J. P. Lonchamp, Compt. rend. **232**, 1824 (1951).
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[16] Brown, Snyder, Fowler, and Lauritsen, Phys. Rev. **82**, 159 (1951).

TABLE I. Relative ionization and average energy per ion pair of the α-particles and Li^7 ions from the reaction $B^{10}(n,\alpha)Li^7$.

Particle	Energy, Mev calculated from independent data[a]	Ionization relative to Po-α (this exp.)	Av. energy per ion pair relative to Po-α This exp.	by C−H formula	Av. energy per ion pair relative to Pu^{239}-α Hanna's exp.[b]	by C−H formula
α	1.778±0.002	0.3286±0.0032	1.021±0.010	1.021		1.021
α*	1.473±0.002	0.2726±0.0018	1.020±0.007	1.026	1.027±0.009	1.026
Li	1.015±0.002	0.1756±0.0024	1.091±0.015	1.059		1.059
Li*	0.841±0.002	0.1451±0.0012	1.094±0.009	1.068	1.080±0.011	1.067

[a] See references 15 and 16.
[b] See reference 5.

consistent with the empirical formula for alpha-particles in argon proposed by Cranshaw and Harvey,[6]

$$W=27.5+1.9E^{-\frac{1}{2}},$$

where W is the average energy in electron volts per ion pair and E is the initial alpha-particle energy in Mev. For Li ions the value of W is taken to be the same as for alpha-particles with the same initial velocity. For comparison with the measured values Table I includes the average energy per ion pair for each particle relative to Po^{210} and Pu^{239} alpha-particles as calculated by the Cranshaw-Harvey formula.

While the alpha-particle ionizations are consistent with the values of W calculated from the Cranshaw-Harvey formula, the Li ionizations measured in this experiment indicate a slightly greater variation in W with particle velocity than is given by this formula when applied to Li ions.

In addition to Jesse, Stebler[14] and Franz and Westmeyer[17] have made determinations of the $B^{10}(n,\alpha)Li^7$ reaction energy using total ion collection in argon and assuming a direct proportionality between energy and ionization. The energy values deduced by Stebler are about 3 percent low (4–5 percent in O_2 and N_2) while those of Franz and Westmeyer are about 2 percent high but with a stated probable error of 4 percent. Stebler used U^{238} alpha-particles (4.180 Mev) for calibration; Franz and Westmeyer used slowed-down alpha-particles (~3 Mev) from polonium.

Facchini, Gatti, and Germagnoli,[18] using electron collection in argon, measured the separate ionizations of the 2.05-Mev alpha-particles and 2.73-Mev tritons from $Li(n,\alpha)H^3$ reaction relative to the ionizations produced by U^{234} and U^{238} alpha-particles. Their results are consistent with the assumption of constant energy per ion pair for the three alphas and the triton within 1 percent probable error. By the Cranshaw-Harvey formula the average energy per ion pair for a 2.05-Mev alpha-particle relative to a U^{238} alpha-particle is 1.014.

Our previous results[4] on the ionization measurements of $He^3(n,p)H^3$ and $N^{14}(n,p)C^{14}$ seemed to indicate the same value of W for the protons, tritons, and C^{14} nuclei as for the polonium alpha-particles used for calibration. However, a detailed analysis of the pulse-height distribution curve for the polonium alpha-particles emitted from a wall source in terms of wall effect (similar to the analysis carried out here) was not carried out in that case. Indications are that such an analysis would decrease slightly the value deduced for the ionization produced by the disintegration products of the (n,p) reactions relative to the ionization produced by polonium alpha-particles. Such a decrease would indicate a variation in W with particle type or velocity as found in the present experiment. However, we are unable to make a reliable estimate of the size of this correction because the exact position of the alpha-particle source in the wall of the outer cylinder is not known.

Fano's[2] considerations of the ionization process make reasonable the approximate constancy of W with particle type and velocity. However, it should not be surprising that small variations in W should exist in view of the loss of energy by nuclear collisions which constitutes a changing fraction of energy loss with velocity, particularly at low velocities.[19] In this respect the experiment performed here is in reasonable agreement with Hanna's results. The disagreement with other experiments[3,14,17] previously referred to may be caused by factors whose influence on the relationship between energy and ionization is only imperfectly understood. Among these factors is the possible presence of small quantities of molecular gases which are mixed with the argon used in the chamber. (The work of Colli and Facchini[20] suggests that the usual purification procedure for argon[21] does not remove traces of nitrogen, for example.) The presence of these gases, while it is not responsible for any loss of ionization charge, may affect the division of particle energy between excitation and ionization.

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[19] J. K. Knipp and R. C. Ling, Phys. Rev. **82**, 30 (1951).
[20] L. Colli and U. Facchini, Rev. Sci. Instr. **23**, 39 (1952).
[21] E. D. Klema and J. S. Allen, Phys. Rev. **77**, 661 (1950).