# The Ionization in Argon and in Air by Single Alpha-Particles as a Function of Their Energy†

WILLIAM P. JESSE, HAROLD FORSTAT, AND JOHN SADAUSKIS
*Argonne National Laboratory, Chicago, Illinois*

(Received October 3, 1949)

An extended series of measurements has been made to determine the possible variation of $W$, the energy to make an ion pair in pure argon, with the energy of the ionizing alpha-particle. The ionization in argon relative to the ionization produced by a comparison polonium alpha has been measured for the alpha-emitters ThX, ThC, ThA, RaC′, ThC′. For alpha-particles in this energy region from 5 to 9 Mev, no deviation of $W$ from the average $W$ for the polonium alpha could be found amounting to as much as $\frac{1}{2}$ percent. This is in marked contrast to the alpha-ionization in air from the work of Stetter. More indirect measurements in the region 1–5 Mev indicate no measurable change in $W$ for alpha-particles in argon, while values derived from the Stetter curve for air show a marked increase of $W$ as zero alpha-energy is approached. Evidence from direct measurements is given to show that the energy to make an ion pair in argon for the $Li^{7*}$ ion emitted from the reaction $B^{10}(n,\alpha)Li^{7*}$ is apparently the same as the average for the polonium alpha-particle.

## INTRODUCTION

FOR many years the ionization by alpha-particles in various gases has been studied both experimentally and theoretically, especially in regard to the relation between the total ionization produced by a single alpha-particle and its energy. The literature is too voluminous to allow here even a bibliography of individual papers pertaining to this subject. A good summary of the experimental results up to 1944 has been given by L. H. Gray,[1] and some of the theoretical aspects have been discussed in an older paper by Livingston and Bethe[2] and in a more recent paper by Bohr.[3] Certain phases of the subject have also been treated by Fano.[4] The general theory would indicate a somewhat greater energy to produce an ion pair by the alpha-particle as it approaches zero energy, but no satisfactory quantitative relations have as yet been derived. The theory seems even more inadequate to deal with the energy to make an ion pair in the similar case of recoil ions such as the $Li^7$ ion in the $(n,\alpha)$ reaction for $B^{10}$. On the experimental side the situation is somewhat more favorable, but even a casual reading of the reference cited brings out many fundamental contradictions between the work of various experimenters. Thus it seems probable that $W$, the average energy in electron volts to make an ion pair, is not constant in air for alpha-particles between 0 and 5 Mev, since relative ionization experiments show a marked difference in the behavior of air[1] as contrasted with a group comprising hydrogen and the noble gases. However, no very reliable work is available showing whether $W$ is really constant with varying energy in the latter group of gases nor how $W$ varies with the nature of the ionizing particle. Most measurements of this sort have been confined to air, and even in the older work with the noble gases, one is never sure that the extreme precautions now known to be necessary to insure the purity of the gas have been taken. Thus it seemed desirable to carry out a series of experiments to determine the variation of $W$ with energy in the noble gases and the variation of $W$ from particle to particle.

## APPARATUS AND METHOD

The best region in which to work with alpha-particles would seem to be the range from 5 to 9 Mev, where there are a large number of natural alpha-emitters for which the alpha-energies have been determined with great precision by Briggs[5] and others. Our first measurements were therefore made in argon with these elements.

A brief description has already been given of the apparatus[6] and general method used together with a table of data showing the consistency obtainable. In general, an ion chamber, of which various types have been used, is connected to a vibrating reed electrometer,[7] which in turn feeds into a Brown recorder. An alpha-particle, usually from an active deposit on one of the electrodes, produces a number of ions in the very pure argon of the chamber. The sudden rise of potential of the floating insulated electrode from the collection of such a charge is amplified in the reed and causes a sharp jump of the pen of the recorder. The length of this jump is proportional to the change of potential and hence to the charge collected. These jumps are very reproducible and are quite large. At full instrumental sensitivity the length of such a jump for a polonium alpha in argon is well over 10 cm.

Perhaps the point which should be most emphasized is that this instrument is, in its operation, akin to an electrostatic electrometer, measuring the accumulated

† A summary of the preliminary work in this paper was given at the June, 1948, meeting of the American Physical Society. W. P. Jesse and H. Forstat, Phys. Rev. **74**, 1259(A) (1948).

[1] L. H. Gray, Proc. Camb. Phil. Soc. **40**, 95 (1944).

[2] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 245 (1937).

[3] N. Bohr, Kgl. Danske Vid. Sels. Math.-Fys. Medd. **18**, 8 (1948).

[4] U. Fano, Phys. Rev. **70**, 44 (1946).

[5] G. H. Briggs, Proc. Roy. Soc. **157**, 183 (1936).

[6] W. P. Jesse and H. Forstat, Phys. Rev. **73**, 926 (1948).

[7] Palevsky, Swank, and Grenchik, Rev. Sci. Inst. **18**, 298 (1947).

charge by the potential rise induced rather than the height of a transient pulse. Since the time of response of the circuit is of the order of one second, both positive and negative ions are collected. The usual counting rate is of the order of 3 to 10 counts/min.

Most of the comparisons between the ionization produced by alpha-particles were carried out in a cylindrical chamber of 10-cm internal diameter and height 7 cm. The collecting electrode, a circular plate of 8 cm diameter, was $4\frac{1}{2}$ cm from the base plate of the chamber. Thus the field was that between two parallel plates, with a certain distortion at the edges due to the presence of the side walls. No apparent difficulty was experienced because of such inhomogeniety. In fact, the exact geometry of the chamber seemed relatively unimportant.

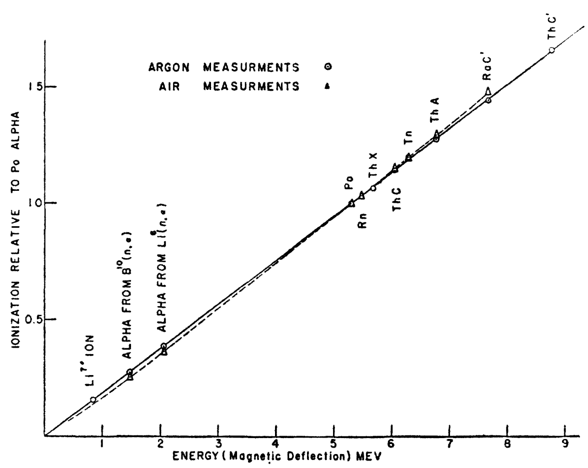


FIG. 1. The ionization of various alpha-particles in argon and in air, relative to that for the polonium alpha, plotted against alpha-particle energy.

The alpha-emitting elements were put on a highly polished button of nickel or stainless steel, inserted flush with the surface at the center of the circular electrode. The polonium was deposited from solution on the button by the usual method, and the other elements were deposited by the standard method of collecting recoil atoms.

The quantity of radioactive material to give five counts per minute is, of course, so small for any of the elements used that the normal sample absorption is completely negligible. However, one can never be sure that some sort of surface interaction between the deposited layer and the metal may not give a distorting effect such as has been recently reported.[8] In our work we could discover no evidence of any distortion of our plotted curves which could be attributed to such a phenomenon. Moreover, measurements with disks of nickel, stainless steel, and platinum all gave consistent results.

The argon used was of "spectroscopic purity" and was taken directly from glass breaker flasks. A calcium cell was used as a monitor, and a large number of runs were made with argon repeatedly circulated through the cell. Except in two doubtful cases, we could determine no marked change in the alpha-ratios after circulating the argon over the calcium. This is contrary to the general experience of those using pulse chambers with linear amplifiers. It must be remembered, however, that our instrument is probably much less sensitive to minute impurities than a linear amplifier system, where the time of flight of a collected ion or electron is an extremely important factor.

Great care was taken to insure voltage saturation in the chamber. Saturation curves of single alpha-jumps against voltage showed a rapid rise to a constant value. With further large increase of voltage no change in the length of the jumps could be observed within the limit of experimental statistical error. In a cylindrical chamber, with a polonium alpha-beam collimated parallel to the cylindrical electrode, we have obtained saturation with fields of approximately 15 volts/cm at an argon pressure of 60 cm. For uncollimated beams at the higher pressures used (up to 160 cm of mercury) one obtains correspondingly higher fields for saturation. In all cases the chamber voltage used was considerably higher than the voltage judged to give saturation.

Throughout the experiment only the ratio of the ionization of the alpha-particle in question to that of the comparison polonium alpha was determined. In finding this ratio, either of two methods could be used. Because of statistical fluctuations, the length $L$ of successive jumps recorded for a given alpha-particle varied. To determine the most probable value of $L$, in the first method the conventional procedure was followed. From the recorded data the number of alphas was found lying between the limits $L$ and $L+\Delta L$, as the length $L$ was increased by successive increments $\Delta L$. $\Delta L$ was an arbitrarily chosen small increment of length amounting to one or two tenths of a scale division. A plot, number of events within the interval against length of jump, gave an essentially Gaussian distribution, both for the alpha-particle in question, and for the reference polonium alpha. The ratio of the lengths corresponding

TABLE I. Relative ionization and energy for alpha-particles in argon.

| Element | Ionization relative to Po$\alpha$ | Calculated energy for Po$=5.298_4$ Mev | Energy by magnetic deflection Mev | Percent difference | Average slope $I/E$ from origin |
|---|---|---|---|---|---|
| Po | 1.0 | | $5.298_4$ | | 0.1887 |
| ThX | 1.067 | 5.653 | $5.681_3$ | $-0.5$ | 0.1878 |
| ThC(mean) | 1.142 | 6.050 | $6.053_7$ | $-0.1$ | 0.1886 |
| Tn | 1.198 | 6.347 | $6.281_8$ | $+1.0$ | 0.1907 |
| ThA | 1.276 | 6.761 | $6.774_4$ | $-0.2$ | 0.1884 |
| RaC′ | 1.444 | 7.653 | $7.680_2$ | $-0.4$ | 0.1881 |
| ThC′ | 1.663 | 8.812 | $8.775_9$ | $+0.4$ | 0.1895 |

[8] Haissensky, Faraggi, Coche, and Avignon, Phys. Rev. **75**, 1963 (1949).

to the peak values for these two plots was taken as the value for the relative ionization for the two particles.

If the plots for the two alpha-particles have the same shape, as they do here, both being essentially Gaussian in character, and if they are well resolved so there is no overlap, then a second method is permissable. Here one takes the *average* length of the jumps included under each plot and determines the ratio. Where the above conditions are fulfilled, the two methods give very consistent results. The second method seems to have some advantage in runs of several hours, if the ratio is determined from groups of data chosen periodically, as within every half-hour. In this case, we are determining the ratio from data taken much more simultaneously; hence, any possible slow drift in the overall sensitivity of the apparatus is minimized.

## RESULTS AND DISCUSSION

The results of our comparison of various alpha-emitters with polonium are shown in Fig. 1. Here the ordinate represents the ionization of each single alpha used relative to the ionization of a comparison polonium alpha in argon. On the horizontal axis are plotted the values of alpha-energies determined by magnetic deflection[9] methods. These energies in the region 5–9 Mev are supposed to be correct to within at least 1 part in 5000. On the scale as plotted, the experimental points seem to lie very well on a straight line through the origin. No accurate conclusion can be drawn as to minor deviations from such a line. Such can best be seen in Table I.

In the third vertical column of this table are shown the alpha-energies calculated from the ionization ratios of the second column on the assumption that the energy of the polonium alpha is $5.298_4$ Mev. In column 4 are shown the true alpha-energies as determined by the magnetic deflection method. If a strict proportionality in the interval between 5 and 9 Mev exists between the energy of the alpha-particle and the ionization in argon produced by it, then the values in columns 3 and 4 should be the same. The difference in percent between these values is given in column 5. In column 6 we have indicated the slope $I/E$ of an imaginary line drawn from the origin to the experimental point in question. The strict proportionality indicated above would be shown in a constant value for this slope.

The result for thoron is at once seen to be in disagreement with the other values. A plausible reason for such a discrepancy would seem to be as follows. Thoron, a gas derived from the decay of ThX on our plate, might well diffuse into the argon and decay there, following its 54-sec. half-life. If this occurs, the ionization due to the recoil atom would be included in the alpha-measurement, increasing it by the order of 2 percent. If the thoron on the average decayed about half the time in the gas, our value, high by about 1 percent, would seem to be explained.

If one accepts this explanation and omits the thoron point from further consideration, then the agreement, within the limits of experimental error, between columns 3 and 4 and the apparent constancy in column 6 would indicate a direct proportionality between ionization and alpha-energy in argon for alphas of energy 5 to 9 Mev.

One has only to compare these results with the results for alpha-particles in air to see the difference between the two gases. We have not carried out the same experiment for air, but apparently very careful measurements have been made in air by G. Stetter.[10] Instead of using the natural alpha-particles, as has been done here, he has obtained different ranges and energies by slowing down a collimated beam of ThC′ alpha-particles by passage through an absorbing column of air. A complete curve has been thus plotted relating the alpha-ionization with the extrapolated range of the alpha-particle. We have from this curve determined the relative air ionization corresponding to the same alpha-emitters used above. Here, except for ThC, we have used the tabulated extrapolated ranges given by Holloway and Livingston[9] for natural alpha-particles. It should be pointed out that these are not rigorously the ranges appropriate to Stetter's curve, since his experimental method gives a somewhat greater straggling than is characteristic of the alphas from the natural elements. It is not believed that any serious error is involved, especially for the alpha-particles of energy higher than polonium.

In Table II are given the results for air so obtained. The experimental points are also shown in the plot for air in Fig. 1. In the table a marked trend is seen in vertical columns 6 and 7 which was not apparent in the argon readings. This variation indicates a much steeper slope for energies larger than polonium for the ionization-energy curve for air compared with the *average* slope from the origin to the polonium point. This trend is shown clearly in the plot in Fig. 1.

Although we have evidence above of the constancy of $W$ in argon within our experimental error for alpha-energies from 5 to 9 Mev, and that this value is the same as the *average* $W$ from 0 to 5.3 Mev, we still have no knowledge as to possible minor variations of $W$ below 5 Mev, especially in the region of the origin. Theory would predict a rise of $W$ near the origin with a decreased slope of the ionization-energy curve. Any least-squares extrapolation from our data to determine an exact intercept on the energy axis with any real significance we must regard, after some trials, as futile, since the extrapolated value is extremely sensitive to even one or two slightly erratic experimental points.

Since there are no natural alpha-particles below 5 Mev with energies as well determined as the ones used, we have had to rely in this region on more indirect

[9] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 (1938).

[10] G. Stetter, Zeits. f. Physik **120**, 639 (1943).

TABLE II. Relative ionization and energy for alpha-particles in air, computed from the work of Stetter.

| Element | Extrapolated range (cm) at 15° and 76 cm | Ionization relative to Po $\alpha$ | Energy calculated for Po =5.298$_4$ Mev | Energy by magnetic deflection Mev | Percent difference | Average slope $I/E$ from origin |
|---|---|---|---|---|---|---|
| Po | 3.870 | 1.0 | | 5.298$_4$ | | 0.1887 |
| Rn | 4.076 | 1.036 | 5.490 | 5.486$_0$ | +0.1 | 0.1889 |
| ThC | 4.760 | 1.150 | 6.093 | 6.053$_7$ | +0.6 | 0.1900 |
| Tn | 5.035 | 1.194 | 6.325 | 6.281$_8$ | +0.7 | 0.1900 |
| ThA | 5.672 | 1.293 | 6.849 | 6.774$_4$ | +1.1 | 0.1908 |
| RaC′ | 6.953 | 1.478 | 7.833 | 7.680$_2$ | +2.0 | 0.1925 |

methods. Fortunately some fairly accurate values of alpha-energies from nuclear reactions have recently been determined, which have been of great help.

For these indirect methods, the relations existing between ionization and alpha-energy derived from such available data are shown in Table III. Here, just as in Table II, the ionization in air relative to the polonium alpha was determined from Stetter's values from the extrapolated ranges. The latter were determined from the mean ranges by adding 0.028 cm, the difference between the mean and extrapolated range for the Po alpha, which value has been determined quite accurately by Holloway and Livingston. The earlier mentioned objection to this procedure applies also here, perhaps to an even greater degree. As has already been pointed out, the Holloway-Livingston range-energy curve,[9] derived between 0 and 5 Mev in essentially the same way as Stetter's, is likewise a range-ionization in air curve. Hence, from the mean ranges one can determine the ionization in air for the chosen particle relative to the Po alpha by simply taking the ratio of the "energies" as read from the $H$ and $L$ curve. The agreement between the Stetter and the $H$ and $L$ determination is seen in vertical columns 4 and 5 to be very good.

For the $Li^6(n,\alpha)H^3$ reaction the mean range is taken from the cloud-chamber result of Bøggild and Minnhagen;[11] for $B^{10}(n,\alpha)Li^{7*}$ a mean of the results of Bøggild,[12] 0.709 cm, O'Ceallaigh and Davies,[13] 0.715 cm and Bower, Bretscher and Gilbert,[14] 0.735 cm has been used. The last result has been corrected for the variation of stopping power with alpha-energy for the gases of the cloud chamber but not by as large an amount as has recently been suggested.[15] The other values already include such a correction.

Jesse and Sadauskis,[16] from a comparison of the relative ionization of samarium and polonium alpha-particles in argon and in air, have shown that the ratio of ionization $(\mathrm{Sm/Po})_{\mathrm{argon}}$ is about 5 percent higher than the ionization ratio $(\mathrm{Sm/Po})_{\mathrm{air}}$. An extension of this work with retarded alpha-particles has resulted in a fairly accurate curve relating the two ratios in argon and in air. Details of this curve will soon be published in another paper relating to other phases of this work. In vertical column 7 are given the multiplying factors to convert the ratio (Ionization for given alpha-particle/Ionization of Po alpha) in air to the corresponding ratio in argon. The latter ratio is given in column 8.

The energy release for each of these nuclear reactions‡ has been independently measured by a method involving the magnetic deflection of the emitted alpha-particle from closely associated reactions. The $Q$-values are 4.788±0.023 Mev for the $Li^6$ reaction[17] and 2.316 ±0.006 Mev for the $B^{10}$ reaction[18] resulting in the excited state for $Li^7$. The corresponding energies for the alpha-particles are given in column 9, and the average slopes $I/E$ are given for air and argon in the last two columns. Here again this slope for argon is seen to be in excellent agreement with the values in the last column

TABLE III. Ionization-energy relations for particles below 5 Mev.

| Particle | Mean range cm | Extrapolated range cm | Corresponding relative ionization in air | | | Ratio rel. ion argon / rel. ion air | Ion. in argon relative to Po $\alpha$ | Direct meas. of particle energy Mev | Slope $I/E$ from origin | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | Stetter | $H$ and $L$ | mean | | | | Air | Argon |
| Alpha from $Li^6(n,\alpha)H^3$ | 1.04 | 1.068 | 0.363$_6$ | 0.369$_9$ | 0.366$_8$ | 1.05$_7$ | 0.387$_7$ | 2.060 | 0.1780 | 0.1882 |
| Alpha from $B^{10}(n,\alpha)Li^{7*}$ | 0.720 | 0.748 | 0.255$_7$ | 0.256$_7$ | 0.256$_2$ | 1.08$_3$ | 0.277$_5$ | 1.474 | 0.1738 | 0.1883 |
| Direct ionization measurements in argon of $B^{10}(n,\alpha)Li^{7*}$ reaction | | | | | | | | | | |
| Alpha from $B^{10}(n,\alpha)Li^{7*}$ | | | | | 0.256$_3$ | 1.08$_3$ | 0.277$_6$ | 1.474 | 0.1739 | 0.1883 |
| $Li^{7*}$ ion from $B^{10}(n,\alpha)Li^{7*}$ | | | | | | | 0.158$_3$ | 0.841 | | 0.1882 |

[11] J. K. Bøggild and L. Minnhagen, Phys. Rev. **75**, 782 (1949).
[12] J. K. Bøggild, Kgl. Danske Vid. Sels Math.-Fys. Medd. **23**, No. 4 (1945).
[13] C. O'Ceallaigh and W. T. Davies, Proc. Roy. Soc. (A) **167**, 81 (1938).
[14] Bower, Bretscher, and Gilbert, Proc. Camb. Phil. Soc. **34**, 290 (1938).
[15] C. W. Gilbert, Proc. Camb. Phil. Soc. **44**, 447 (1948).
[16] W. P. Jesse and J. Sadauskis, Phys. Rev. **75**, 1110 (1949); Phys. Rev. **76**, 163 (1949).
‡ We are indebted to Professor W. A. Fowler for a private communication giving us these values before their formal publication.
[17] Tollestrup, Fowler, and Lauritsen, Phys. Rev. **76**, 428 (1949).
[18] Chao, Lauritsen, and Tollestrup, Phys. Rev. **76**, 586 (1949).

of Table I, while for air a comparison of the slopes in Table III with those of Table II shows a falling off of the order of 10 percent.

In the last two horizontal rows in Table III are listed the results of direct measurements of the $Q$ for the $B^{10}(n,\alpha)Li^{7*}$ reaction by an ionization chamber method. These measurements were made in this laboratory in a chamber similar to that described above. The runs were in argon to which had been added about 0.1 percent of enriched $BF_3$. The method was essentially that described above. A mean of six determinations at various pressures up to 160 cm of mercury gave for a total $Q$ of the reaction $2.310\pm0.010$ Mev. The error here given was arbitrarily set from our past experience with such measurements. In this case it is about three times the standard deviation for the series of measurements and does not include any possible systematic errors. No corrections of any sort have been made to the data, such as a correction for the finite size of the chamber. This is believed to be very small but would tend to raise the values obtained. A comparison of our value with that of Chao, Lauritsen, and Tollestrup shows an excellent agreement, well within the combined experimental errors of the two determinations.

Unless we assume that the agreement here is the result of pure chance, i.e., a combination of experimental errors in both determinations forcing them into agreement, which of course may possibly be the case, we are forced to the conclusion that the average $W$ for the combined ionization from $Li^{7*}$ and the accompanying alpha is the same as for the average $W$ for the polonium alpha. From this it would seem reasonable to assume the average $W$ for $Li^{7*}$ the same in argon as the average $W$ for the polonium alpha. For comparison with the previous values from ranges, the appropriate values from these direct measurements have been calculated in Table III. The measured ionization has been partitioned in the same ratio as for the energies, and ionization values computed for air by now dividing by the ratio in vertical column 7. The applicability of this ratio, derived for alpha-particles, to the $Li^7$ ion is, of course, questionable. Hence, no values for the $Li^{7*}$ ion in air were computed.

In the complete curves in Fig. 1 and from the tables there seems no evidence of a departure in argon, within the error of experiment, from a strict proportionality between ionization and particle energy. In air the plotted points between 2 and 6 Mev seem to fall fairly well on a straight line drawn from about +0.175 Mev on the energy axis through the polonium alpha-point. Numerically the values $I/E'$, where $E'=E-0.175$ Mev, for the experimental values agree within $\frac{1}{3}$ percent inside this region. In the region 0–2 Mev and from 6–7.5 Mev the plotted points lie somewhat above this line, i.e., the curve is slightly concave upward. The values for RaC′ and for the $B^{10}$ alpha are about 1 percent high in such a calculation.

In support of our present findings as to the essential constancy of $W$ for alpha-particles in argon, it would seem worth while to mention two further pieces of evidence which, while not conclusive in themselves, at least support the above results.

1. It has been shown by Jesse and Sadauskis[16] that the alpha-range-energy curve of Holloway and Livingston on the basis of ionization measurements in air can be converted to a corresponding curve involving measurements in argon. The latter curve gives values in very good agreement with ranges and alpha-energies independently determined for a number of nuclear reactions. The agreement in a few cases has already been shown here indirectly in Table III. Furthermore, when the revised alpha-range-energy curve is used to determine the proton range-energy curve by Blackett's[2] transformation equation, the latter curve gives values in much better agreement with recently determined nuclear data. These revised range-energy curves are now consistent with other data, where the former curves were markedly inconsistent. This fact would seem a plausible argument for the fundamental assumption that in argon the value of $W$ is at least approximately constant.

2. In recent experiments involving the measurement of the relative ionization of $\beta$-rays in argon, Curran, Angus, and Cockroft[19] have found that the energy to produce an ion pair in argon is nearly constant within the energy range 200 ev to 50 kev. This result is consistent with our findings above for alpha-ionization, since the latter ionization is made up in part of ionization from $\delta$-rays with energies of a few kev. The fact, that in air for $\beta$-particles this direct proportion is no longer valid[20] but the energy to make an ion pair increases at low $\beta$-energies, is also consistent with the similar behavior for alpha-particles in air. The constancy of $W$ for $\beta$-particles in argon would not seem, however, an entirely conclusive proof of the constancy for alpha-particles, since undoubtedly some ions are formed in the alpha-track by processes other than ionization by $\beta$-particles within the energy range investigated by Curran, Angus, and Cockroft.

## ACKNOWLEDGMENTS

It is a pleasure to express our thanks to Professor H. A. Bethe, Professor Edward Teller, and Dr. Francis R. Shonka for many stimulating discussions of this general subject. Our thanks are due also to Mr. L. C. Ellsworth, whose painstaking skill in the construction of our ion chambers has added greatly to the success of this work.

[19] Curran, Angus, and Cockroft, Phil. Mag. **40**, 36 (1949).
[20] Reference 1, p. 91.