and auxiliary equipment, was designed and built by Professor R. B. Brode in 1940 and 1941 expressly for the measurement of the mesotron mass. Funds for this equipment were made available by a grant from the Carnegie Institution of Washington. The lower cloud chamber and its camera were built by Professor W. E. Hazen, who suggested the experiment and to whom I am deeply indebted for constant instruction and encouragement. Professor R. T. Birge was very helpful in connection with the statistical analysis of the data.

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# Low Energy Alpha-Particles from Radium

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The large alpha-ray spectrograph and the sensitive track method of detection, as used for studying the polonium alpha-particles, have been used to investigate the alpha-ray spectrum of radium. Thin and uniform radium sources were freshly prepared by depositing radium carbonate on platinum rods, using a modified method of Hahn and Meitner. Microscopic examination has revealed a line at 4.615 Mev, which is identifiable with that found by Rosenblum. From 0.5 to 0.9 Mev below the main line five previously unknown groups have been found. The intensities of the first line and of these last ones are, respectively, about 1800 and 50–20, if the intensity of the main line is set as 100,000. As in the polonium case, if the particle groups come from the nucleus as all experiments have indicated, the ordinary alpha-decay theory is in serious disagreement with the experiments, for the theoretical intensity varies with energy much more rapidly than the observed intensity. Therefore, a mechanism other than a simple penetration through the static potential barrier may be needed. Attempt has been made to explain these results by assuming a strong interaction between the outgoing particle and the rest of the nucleus. This interaction may imply a transfer of kinetic energy from the particle to the residual nucleus, which will then make the decay probability appear much larger. The total probability of decay and the resultant excitation of the nucleus are discussed in terms of this tentative mechanism.

## 1. INTRODUCTION

MORE than a year ago we found a series of weak alpha-particle groups[1] in the energy region below the polonium main alpha-ray line. If these alpha-particle groups are assumed to come from within the polonium nuclei, as all experiments have indicated, leaving the lead nuclei in different excited states, they may be then correlated with the gamma-rays from Po. In general the intensities and energies of these groups are found to be of the similar order of magnitude to those of the gamma-ray lines from Po as found by Bothe. However, the intensities of these groups are quite out of line with the predictions of the ordinary alpha-decay theory which assumes penetration through a static potential barrier. For this reason it is of interest to study similar effects in other radioactive nuclei which have different main decay periods and different $\alpha$-particle energies. The present paper reports the results of an examination of the $\alpha$-particle spectrum of radium.

[1] W. Y. Chang, Phys. Rev. 69, 60 (1946).

## II. EXPERIMENTAL

Because of the longer decay period of radium and the activity of its decay products the work with radium is much more troublesome than with polonium. On the one hand, short exposure times require relatively thick sources entailing $\alpha$-particle straggling. On the other hand, long exposure with thinner sources introduces difficulties caused by background from contamination of the chamber near the source box and from the $\alpha$-particles from the decay products. A compromise between these evils was necessary.

To prevent contamination of the whole apparatus with emanation the old radon must be driven out before the preparation of the source and the radon developed in the source during

preparation and exposure must be prevented from escaping into the spectrograph.

Since radium is a less noble element electrochemically, it cannot be deposited on a wire by a purely electrolytic method. A combination of electrolytic and chemical methods is generally used for this purpose. An electric current transports the ions of the substance to the cathode, where they are chemically precipitated and under suitable conditions form a coherent deposit on the cathode. On this principle, we have adopted and somewhat modified a method of Hahn and Meitner[2] in preparing our line source of radium.

Two mg of radium bromide (about 95 percent pure) were dissolved in 20 cc of distilled water. The solution was heated continuously for three or four hours in a boiling water bath to drive off the radon gas already generated and to let the corresponding active products decay completely. Two platinum wires about 0.8 mm in diameter and 40 mm long were horizontally immersed in the solution about 5 mm below the solution surface. The solution was then saturated with $CO_2$ gas until the milky radium carbonate was redissolved (instead of bubbling $CO_2$ gas through the solution only during deposition). In this way we have sufficient carbonate ions throughout the whole solution for future precipitation and have sufficient acidity (but not too strong) caused by carbonate acid for conducting the electric current. A current of about 20 ma was passed through the solution. Because of the weakness of the acidity there is an extensive alkaline zone around the cathode and in the immediate neighborhood of the cathode the acidity must be smaller than at any other part of the zone because of liberation of hydrogen gas from this part of the solution. The radium carbonate formed in this region would not be dissolved but instead would be deposited on the cathode (presumably partly caused by the remaining velocity toward the cathode of the radium ions). The evolution of the hydrogen gas at the cathode would drive off any radon gas newly formed. In a period of about an hour about $\frac{1}{5}$ to 1/20 mc could be deposited uniformly on the wire. The source was then washed carefully with distilled water and heated gently to red heat in a Pyrex tube. In this way radon was driven off[3] and the deposit made to adhere more firmly to the wire then possibly in non-emanating state. The source prepared in this way was practically free from decay products. Only an extremely small number of the 7-cm alpha-particles were emitted as checked with the alpha-ray counter.

When the solution had no other acidity than that caused by carbonate acid, about 45 volts were required to send the above current through the solution (the two wires were about 2 cm apart). In this case a white uniform coating was formed on the wire. When the solution had traces of hydrochloric acid, the required voltage dropped to two or three volts and under these circumstances a black coating having practically no radioactivity was formed on the wire. Therefore the solution before being saturated with $CO_2$ must be practically neutral. The source after use could be dissolved in the original solution with very dilute HCl acid. After this the solution had to be gently heated to dryness two or three times before the above procedure was repeated, in order to be sure that the solution was free from acid. The platinum electrodes could not be left in the solution when not in use; otherwise, under the attack of the radiations, platinum would pass into the solution,[4] and this might interfere with the deposition.

The experimental arrangements, i.e., the spectrograph and the disposition of the Lucite chamber slits, etc., were exactly the same as in the experiments on the polonium alpha-particles. The technique of employing the track method to record the alpha-particles was also the same. In each of the experiments the time of exposure was four hours. It may be mentioned again that, in this case as well as in the polonium case, the number of irregular tracks on each plate is, under favorable conditions, negligibly small in the region near the main line and is only a small percentage of the corresponding number of regular tracks in the region further away. From blank

[2] O. Hahn and L. Meitner, Zeits. f. Physik **26**, 161 (1924); F. Tödt, Zeits. f. physik Chemie **113**, 329 (1924).

[3] Only a very small percentage of the radon gas can escape from even an untreated solid radium source, if it is not heated strongly. In our spectrograph the source-box is quite near to the opening connected to the pumping system; hence any traces of radon gas given off can be drawn into the pump almost immediately.

[4] Mme Curie, le Radium **4**, 349 (1907); E. Rutherford, *Radioactive Substances and their Radiations* (Cambridge University Press, England, 1913), pp. 314–315.

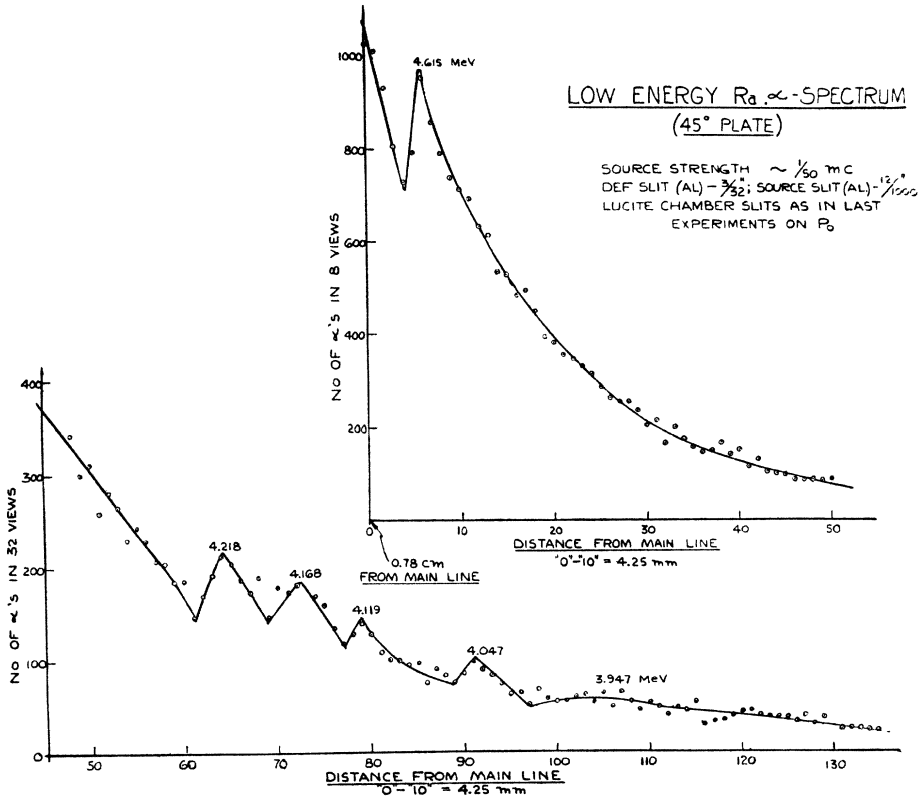


FIG. 1. Low energy-number distribution of the radium alpha-particles. Lower curve is an energy continuation of the upper one. The first group is identifiable with that found by Rosenblum. In the still lower energy region some less prominent groups seem to appear.

experiments the background due to contamination of the chamber was found also negligibly small in comparison with the respective small group intensity.

### III. RESULTS

Figure 1[5] shows the low energy distribution of the radium alpha-particles. It was obtained by exposing in the spectrograph plates inclined at 45° to the alpha-particle beam from a radium source of about 1/50 mc. The lower curve is a continuation (in energy) of the upper one. The intensity axes have however different scales; each figure on the upper vertical axis represents track population in 8 views, while that on the lower one is for 32 views. It is seen that near the main group (not shown) is a prominent line, the energy of which is 4.615 Mev and may therefore be identified with the line, 4.612 Mev, found by Rosenblum.[6] Below this group no distinct groups seem to exist until one reaches the lower curve, where four or five groups seem to appear. However, since the peak intensities of these lines relative to the background are not much larger than their respective statistical fluctuations, it cannot be certain that they are real. To be sure about their existence we took another exposure with a stronger source and examined the plates only in this portion of the curve.

Figure 2 represents the energy-intensity distribution so obtained with a source which was about five times stronger than that used for Fig. 1. The discrete groups, as can be seen from the curve, appear here much more distinctly than in Fig. 1;

[5] To be sure that the existence of the discrete groups is not caused by "psychological prejudice," the two halves along the length of the plate were independently examined in two separate periods, i.e., one being examined in the first week while the other in the fourth week. The two curves so obtained are very similar to each other. Curve 1 is the result of these two.

[6] S. Rosenblum and J. Perrin, Comptes rendus **195**, 317 (1932).

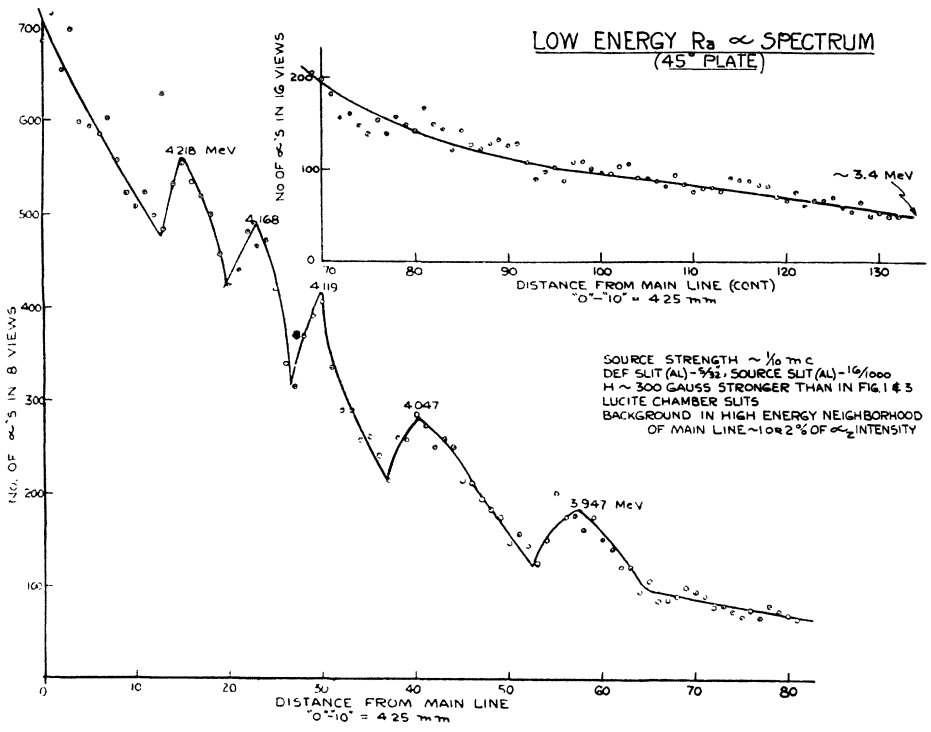


FIG. 2. The last part of the curve in Fig. 1 (and its still lower extension) repeated with a stronger radium source (about five times stronger). The less prominent groups appear more distinctly in this curve. No distinct groups seem to appear in the still lower region.

their peak intensities are several times larger than the respective statistical fluctuations. The positions of the corresponding groups in these two different curves may agree with one another to within about 0.02 Mev. Allowing for the difference in number of the microscopic views counted and in the width of the source and defining slits used, the intensities of any one group in these two curves are about in the same ratio as the relative strengths of the two sources. The microscopic examination has been extended down to a region about 1.5 Mev below the main line. There is no indication of any prominent alpha-particle groups in this region. This is shown in the upper curve of Fig. 2, a continuation of the lower one.

It may be interesting to see if the form of the distribution curve will change after a radium source is left in air for a period of time. Figure 3 was obtained after the same source as used for Fig. 2 was left in air for 30 hours. It is seen that the groups have become broader and less prominent, particularly the lines 4.168 Mev and 3.947 Mev. This must be because of (1) the change in surface condition of the source and (2) the increase in background caused by the growth of the decay products from radium. However, the second effect in this low energy region of the spectrum must be very small, for the energies of the alpha-particles from all the decay products are greater than that of the alpha-particles from radium and experiment shows that the background even in the high energy neighborhood of the radium main line is only about five percent of the $\alpha_2$ intensity. Therefore, the change in the distribution form may be chiefly caused by the change in surface condition of the source.

As in the polonium case,[1] the energies of these groups have been calculated by comparison with that of the main group, the latter being taken as 4.793 Mev. The relativistic correction has been neglected as before. The results and probable errors are summarized in Table I. The energy value of each group in the table (as well as on the curves) represents the average of two corresponding values from Figs. 1 and 2, which were obtained with the two radium sources. The rela-

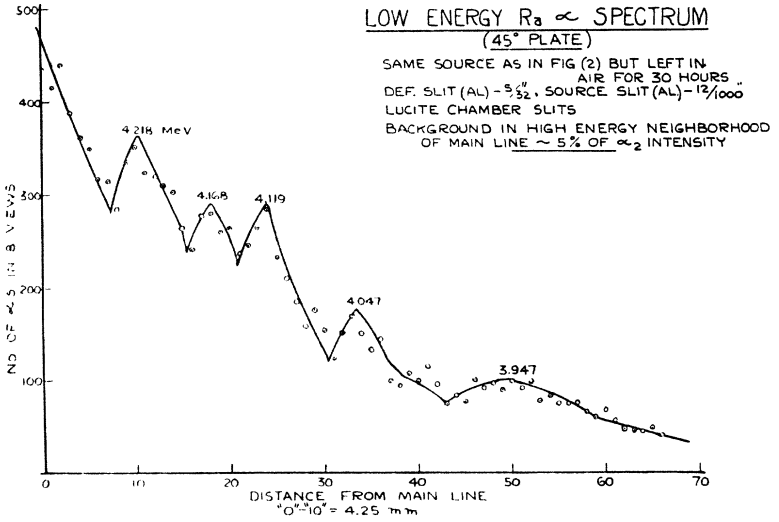


FIG. 3. Same portion of the curve showing the weak low energy groups, obtained with the same radium source after it was left in air for about 30 hours. The weak groups appear broader and less prominent.

tive integral intensities were estimated after the general backgrounds were subtracted from the peak intensities, as in the case of polonium alpha-particles, according to the natural trend of the background of the first group in each curve. These are in the last column of Table I, each figure also representing the average of two corresponding values from Figs. 1 and 2. It must be pointed out that in this case the general backgrounds are much larger and the lines are broader than in the polonium case, presumably caused by the straggling of the alpha-particles through a much greater thickness of the source. Therefore, it is more difficult to find the energy positions and particularly to estimate the relative integral intensities. In the latter case the values can be easily off by a factor of two or more, and the listed intensities may perhaps be on the larger side.

## IV. DISCUSSION

In the case of polonium we performed several experiments under different experimental conditions, to see if the weak alpha-particle groups did actually come from within the polonium nucleus. From these tests, we are inclined to believe that the polonium alpha-particle groups do have a nuclear or atomic origin. For such an origin further evidence is furnished by a comparison of the radium alpha-ray spectrum with that of polonium. If the weak particle groups were caused by some external effects inherent in the instruments, one would expect the radium alpha-particles to have the same spectrum as (or at least similar spectrum to) the polonium alpha-particles, for the experiments on radium alpha-particles were done under the same conditions as the last experiments on polonium alpha-particles. However, the two spectra of radium and polonium are entirely different from each other as can be seen in Fig. 4. In this figure the horizontal axis represents the group energies relative to that of the main group, and the figures above the vertical lines are the corresponding relative integral intensities. The upper spectrum is for polonium while the lower one is for radium. It is seen that the radium groups so far observed occur only within one-Mev energy difference, whereas several polonium groups occur beyond one-Mev difference. Besides, no coincident groups (i.e. having the same energy positions) have been found.

As in the polonium case, if the particle groups do actually come from the nucleus, the emission of an alpha-particle must leave the residual

TABLE I. Group energy of radium alpha-particles.

| Group | Group energy in Mev | $E_n=\alpha_0-\alpha_n$ in Mev | Relative integral intensity |
|---|---|---|---|
| $a_0$ | 4.793 | 0 | $10^5$ |
| $a_1$ | 4.615±0.015 | 0.178 | 1800 |
| $a_2$ | 4.218±0.015 | 0.575 | 45 |
| $a_3$ | 4.168±0.015 | 0.625 | 33 |
| $a_4$ | 4.119±0.015 | 0.674 | 35 |
| $a_5$ | 4.047±0.015 | 0.746 | 25 |
| $a_6$ | 3.947±0.015 | 0.846 | 18 |

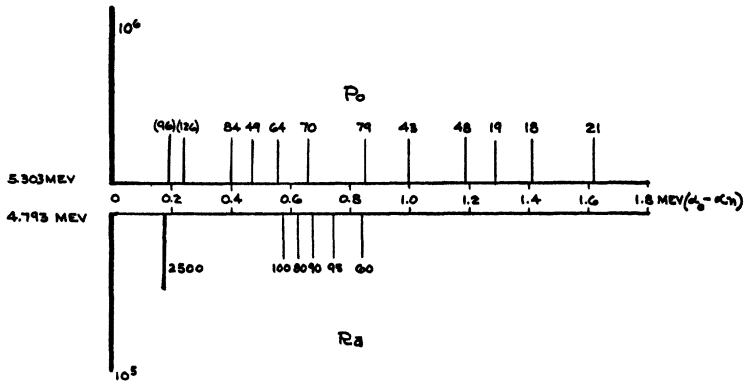


FIG. 4. A comparison of the spectra, respectively, from Po and Ra. They were obtained under the same experimental conditions but are thus shown to be quite different.

nucleus, i.e., $_{86}Rn^{222}$ in the present case, in an excited state. The differences between the main group energy and the individual group energies give the corresponding energy states thus excited of the product nucleus, neglecting the effect of the recoil energy on the excitation energy. These are in the third column of Table I. The gamma-ray line, 0.178 Mev (to be expected from the transition between the first excited state and the ground state), agrees within the experimental error with the line, 0.189 Mev, as observed by Hahn and Meitner.[7] Other gamma-ray lines of much smaller intensities must be expected as a result of transitions from the higher levels to the lower ones. Actual measurements of these lines would certainly be complicated by the presence of the very much stronger lines emitted from the decay products of radium. Moreover, a method of detection for these weak gamma-ray lines must have a sensitivity comparable with that of the track method for the alpha-particles. However, it is fortunate for the possibilities of future experimentation in this direction that a very much stronger (and hence thicker) source can be used for the gamma-ray measurements than for the alpha-particle measurements, on account of the very much higher penetrating power of the gamma-rays.

TABLE II. Theoretical spin changes in different Ra alpha-transformations.

| Type of alpha-transformation | $\lambda_k \times 10^{15}$ sec.$^{-1}$ | $v_k(1+4/222) \times 10^{-9}$ cm/sec. | $r_{eff} \times 10^{12}$ cm | $j_k$ |
|---|---|---|---|---|
| $a_0$ | 14000 | 1.547 | 0.720 | 9 |
| $a_1$ | 252 | 1.519 | 0.702 | 10 |
| $a_2$ | 6.3 | 1.452 | 0.764 | 8 |
| $a_3$ | 4.6 | 1.443 | 0.792 | 6 |
| $a_4$ | 4.7 | 1.435 | 0.812 | 5 |
| $a_5$ | 3.5 | 1.422 | 0.844 | 4 |
| $a_6$ | 2.5 | 1.405 | 0.876 | 0 |

[7] O. Hahn and L. Meitner, Zeits. f. Physik **26**, 161 (1924).

The same difficulty as in the case of polonium also exists in the present case, when one tries to apply the current alpha-decay theory to the observed energy-intensity relation. In the first place, the unacceptably large spin changes as calculated from the theory in the usual way for the different alpha-transformations here occur again as can be seen from Table II. The values vary from 10 to 0 as the alpha-transformation is changed from the 'normal-normal' one to the 'normal-highest' one. Since both radium and radon have even mass numbers as well as even charge numbers, both of them are expected to have zero spins in their respective ground states; but the spin change in the corresponding transformation as calculated from the theory is 9. Secondly, this disagreement between theory and experiment can be revealed more clearly from the Geiger-Nutall curves as is shown in Fig. 5. The line joining the crosses is for the members of the radium family and can generally be described by the theory. For the convenience of discussion we may take it as a theoretical curve. It is seen that the departure of the observed intensities of the radium groups becomes larger as the energy separation from the main group energy gets larger just as in the case of the polonium groups, and that the two curves except the initial portions for the polonium and radium groups are practically parallel.

One way out of the difficulty mentioned above

may be suggested from the Geiger-Nutall curves in Fig. 5, if one has to ascribe these alpha-particle groups to the nuclear origin. We have seen that at a given kinetic energy of the emitted alpha-particle the observed intensity is much larger than the theoretical value. One can of course speak of this same fact in another way round by saying that at a given probability of emission the observed kinetic energy of the emerging alpha-particle is smaller than would be expected from the theory. Therefore, according to this latter version one may say that a certain amount of the particle's kinetic energy is lost during emission by some process which has a mechanism other than the simple penetration through the static potential barrier. As mentioned once before,[8] it may be reasonable to assume tentatively that the outgoing particle, even when it is almost completely free of the potential barrier, may interact strongly with the residual nucleus. And as a result of this interaction, it may transfer to the latter a certain portion of its kinetic energy beside the recoil energy. The energy so imparted may then become the energy of some internal mode of motion of the residual nucleus, the product nucleus consequently being excited to different quantum states. Hence, at a given probability of the alpha-emission, the observed kinetic energy of the particle should be smaller than would be expected from the alpha-decay theory. In other words, the observed intensity would then appear greater for the observed kinetic energy, as can be seen from the Geiger-Nutall curves in Fig. 5. The experimental results may thus be explained in general terms.

On the basis of the above interpretation, one may be able to say something about the relative order of magnitude of the effects for two different types of nuclei, which emit two different main groups of alpha-particles. Alpha-particles from a decay process of low energy will have to traverse a thicker wall of the potential barrier than those from a decay process of higher energy. Consequently a larger interaction and hence a greater portion of the kinetic energy to be given back to the residual nucleus may be expected. As a result of this the intensity of a weak group (relative to

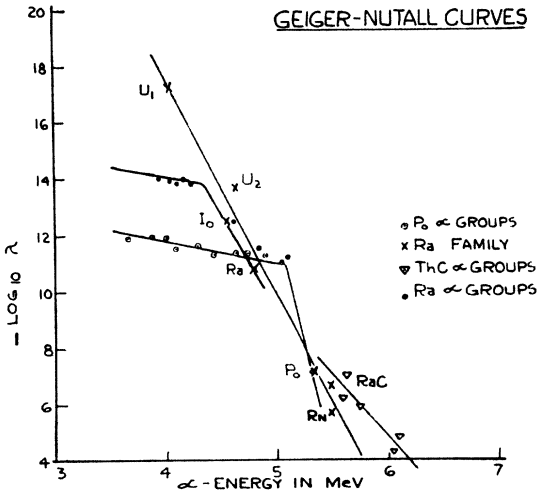


FIG. 5. The Geiger-Nutall curves of the Ra and Po groups as compared with those for the members of the Ra-family and for the ThC-groups.

the main intensity) at a given energy separation from the main line would appear larger in this case than in the case of higher main group energy. A comparison of the relative intensities of the weak groups of polonium and radium, respectively, at about the same energy separation from the main lines seems to bear out this interpretation.

The probability of the alpha-decay may then be a result of the probabilities of the following three processes, namely (1) the probability of formation of the alpha-particles within the nucleus (according to Bethe),[9] (2) the probability of penetration through the potential barrier (according to Gamow),[10] and (3) the probability apparently contributed by the interaction in the manner as described in general terms above. It is obvious that, on account of the interaction between the emerging alpha-particle and the residual nucleus, the effective form of the potential barrier may be much more complicated than has been usually assumed in the current alpha-decay theory, and it may not be at all a static one. Instead, it may be a complicated dynamical one. Furthermore, for the emission of alpha-particle groups of much lower energy than the main group energy from the same nucleus, the third process has to play a much more important role than the

[8] W. Y. Chang, Phys. Rev. 69, 254 (1946).

[9] H. A. Bethe, Rev. Mod. Phys. 9, 163 (1937).

[10] G. Gamow, *Structure of Atomic Nuclei* etc. (Oxford University Press, New York, 1937).

first two processes in order to give agreement with the experimental results.

As to the excitation of the residual nucleus, we may have two fairly distinct steps: (1) "Internal Excitation"—the alpha-particle may leave the interior of the nucleus with different possible energies, leaving the nucleus in the corresponding "partial excited states," and (2) "External Excitation"—the emerging alpha-particle may interact with the nucleus while it is traversing the potential barrier and consequently it may excite the residual nucleus to different possible modes of motion as already mentioned above. The final likelihood of excitation of the product nucleus may then be the resultant of these two "partial excitations"; indeed, we will expect from the general principles of quantum mechanics that it will be, not the probabilities themselves, but the probability amplitudes for these two processes which will be additive. There seems no reason to believe that alpha-particles will leave the nuclear interior all with one energy, putting the responsibility of exciting the nucleus entirely to the second step. Besides, it may be reasonable to say that step (1) takes place independently whether alpha-particles inside the nucleus already exist as sub-units or whether they are created at the instant of emission. Generally, one may assume in terms of Bohr's idea of many-body model that of the maximum amount of energy available within the nucleus the alpha-particle may leave the nucleus with the whole amount or with a certain definite portion. However, higher states of the nucleus will be excited more easily by the interaction process, if the above explanation ("external excitation") is reasonably correct.

A few exposures have been taken for the alpha-ray spectra of RaC, RaC′, RdTh, ThC, ThC′ etc., and microscopic analysis of the plates has been in progress. We hope that the present difficult problem can be revealed more clearly and definitely, when we get more results for these elements.*

I should like to express my sincere thanks to Professor R. Ladenburg, Professor E. P. Wigner, and Professor J. A. Wheeler for their kind interest and useful discussion in this work, and to Mr. Thomas Coor for his assistance in some of the experiments.

* *Note added to the proof on the integral intensities of the weak alpha-particle groups:* In a previous paper (Phys. Rev. **69**, 72 (1946)) we mentioned that the integral intensities of the weak groups were obtained by subtracting the general background which was determined by the natural trend of the first weak group's background in each curve, and that this subtraction of the background is very arbitrary. Recently we have found in the Po case that the values of the intensities (except $\alpha_1$ and $\alpha_2$) relative to $\alpha_0$ will become about $\frac{1}{2}$ to $\frac{1}{3}$ of the corresponding previous values listed in the above mentioned paper, if the area of each small group is measured down only to the minima of the curve. This is because of the fact that the base and hence the area of each group becomes considerably larger as we go further down below the minima. Similarly in the present case of radium the values (except $\alpha_1$) relative to $\alpha_0$ will become about $\frac{1}{2}$ of the corresponding values in Table I. Therefore, one may not be surprised if the integral intensities of any small group relative to the main group intensity, as given previously, is off by a factor of three. For comparison with gamma-ray intensities in the case of polonium, reference should be made to a letter by Professor N. Feather in Phys. Rev. **70**, 88 (1946).