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THE NATURAL BETA-RAY SPECTRUM OF RaD*

By L. F. Curtiss+

Abstract

The natural β-ray spectrum of RaD has been photographed and the energies of its lines directly compared with those of the RaB β-ray spectrum in the same region. The RaB line $H\rho=660.9$ as measured by Ellis and Skinner[1] has been taken as a standard reference line. The following values for the energies of the lines have been determined:

No.	Intensity	$H\rho$	Energy (volts$\times 10^{-6}$)
1	50	594.3	0.3033
2	3	600.3	0.3092
3	25	709.1	0.4268
4	10	735.2	0.4576
5	1	742.5	0.4663

No. 5 is a line which has previously escaped detection. The analysis of this spectrum for determining the energy of the γ-ray responsible for it shows conclusively that the γ-ray is emitted *after* the disintegration. This is in accord with Meitner's[2] view. The energy of the γ-ray is determined as 0.467×10^{6} volts.

Introduction

The present interpretation of β-ray spectra has led to two views regarding the emission of the γ-ray. Ellis[3] originally contended that the γ-ray was emitted before the disintegration, i.e., before the emission of the nuclear or primary β-ray. On his view of course the secondary β-rays should have energies related to the energy levels of the atom before disintegration. Meitner,[2] on the other hand, concluded that the γ-radiation follows the disintegration and is a direct result of the disturbance of the nuclear levels occasioned by the ejection of the primary β-ray and the transition from one nuclear system to the other.

The settlement of this question is important. However, the experi-

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+ National Research Council Fellow.

[1] C. D. Ellis and H. W. B. Skinner, Proc. Roy. Soc. **105**, 165 (1924).

[2] L. Meitner, Zeits. f. Physik **11**, 35, (1922).

[3] C. D. Ellis, Proc. Cam. Phil. Soc. **21**, 121 (1922).

mental material which throws definite light on the problem is still very meager. In the majority of cases the measurements of β-ray spectra may be interpreted in terms of either hypothesis. This ambiguity arises from the nature of the β-ray spectrum. The accepted interpretation of the β-ray line spectra is that the lines represent groups of electrons which have been ejected from the x-ray levels of the atom by γ-radiation from the nucleus of the atom itself. The velocities of these electron groups are measured by bending them in a uniform magnetic field. The velocity is proportional to the product $H\rho$, where H is the intensity of the magnetic field, and ρ the radius of the circle on which the β-particles travel. For the harder β-radiation of RaB and for all lines in the RaC β-ray spectrum, the observed $H\rho$ should differ by less than one-half percent if the atomic levels for $Z=83$ were used instead of those for $Z=82$. (Since RaB is an isotope of lead and disintegrates into RaC, an isotope of bismuth, these are the two atomic systems with which we are concerned in this case.) This difference is beyond the limit of accuracy of most experiments. However, Ellis and Skinner[1] in their more precise measurements of the RaB β-ray spectrum, find that in the "*C*" region they are forced to use atomic levels for $Z=83$ instead of 82 in order to obtain agreement for the energy of the γ-ray. This is the most decisive previous evidence in favor of Meitner's hypothesis.

It is clear that further evidence on this question can be obtained by studying other radioactive elements which emit comparatively soft γ-radiation. Under such circumstances the secondary electrons of the β-ray spectrum will have lower velocities and therefore energies more nearly comparable with the energies of the atomic levels. The natural β-ray spectrum of RaD is well suited to this purpose. RaD is an isotope of RaB and emits a single γ-ray of energy considerably less than that corresponding to the K limit for this atom. Consequently its β-ray spectrum contains electrons from the L level and levels of lower energy. Numerous measurements have been made of this spectrum but there is still great divergence of opinion concerning it. Various observers disagree even on the number of lines that it contains. For example, Ellis[3] observed five lines whereas Meitner[2] later found but three. For convenience previous results are tabulated below. The quantities given under the name of each author are the values of $H\rho$ for lines which they observed in the RaD β-ray spectrum.

The weight of evidence favors a β-ray spectrum of four lines, as originally reported by Danysz. The most recent measurements are by

TABLE I

Hρ values of RaD β-ray spectrum by previous investigators

Danysz[4]	Ellis[3]	Meitner[2]	Black[5]
602	600	602	600
607	605	—	606
—	628	—	—
718	717	718	714
743	742	741	738

Black.[5] These differ from Danysz's by an approximately proportional decrease of the energy of all lines. The failure of certain investigators to detect some of these lines has given rise to considerable discussion regarding their physical significance.[2]

EXPERIMENTAL RESULTS

With the hope of throwing further light on the problem of the emission of the γ-ray, I have taken a number of photographs of the RaD β-ray spectrum. The spectrograph was of the usual type, but with large dispersion ($\rho_{max} = 7.5$ cm). The magnet was one specially designed for the study of β-ray spectra. It has poles 24 cm in diameter and gives a field uniform to one part in 2000 by actual measurement over a circle 15 cm in diameter. This magnet and the spectrograph will be fully described elsewhere.[6] The magnet was excited by an independent set of storage batteries. The current was measured and controlled by a Leeds & Northrup potentiometer and standard resistance in the usual manner. It was found comparatively easy to maintain the current constant to at least one part in 2000 over long periods of time. In order to avoid the trouble of precise absolute measurement of the intensity of the magnetic field, comparison photographs of RaB lines in the same region were made. The line $H\rho = 660.9$, as measured by Ellis and Skinner,[1] was used as a reference line and their value for its energy was accepted as a standard. The lines of the RaD spectrum here reported have thus been measured in terms of this value. It is obvious that the absolute energies of the lines observed here must be as accurate as this value, since a direct comparison of this kind can be made with great precision. Eastman x-ray plates were found at least 50 percent more sensitive than ordinary plates to β-radiation in this region. This test was made by exposing a plate of each kind simultaneously in the spectrograph and developing the two plates

[4] Danysz, Le Radium **10,** 4 (1913).

[5] Black, Proc. Roy. Soc. **109,** 166 (1925).

[6] L. F. Curtiss, J.O.S.A. and R.S.I. in press.

together. Consequently all exposures were made with x-ray plates. The RaD sources were obtained by the anodic deposition on a fine platinum wire in the usual way. Two different preparations of RaD were used, both giving identical results.

The experiments were carried out as follows: The current through the magnet was adjusted to the desired value and after several reversals the switch was thrown to the "direct" position. A source of RaD was inserted into the spectrograph by means of a ground glass joint, the photographic plate having been previously put in position. The spectrograph was then exhausted by a large diffusion pump. A shutter, manipulated by a ground glass joint, protected the plate from β-radiation during the period of exhaustion, which usually was of the order of 20 minutes. After a sufficient exposure the shutter was closed, air admitted, the spectrograph opened, and the plate removed. A fresh plate was then inserted. The RaD source was replaced by a source consisting of the active deposit of radon and a new exposure made. Throughout this time the current through the magnet was maintained constant at its original value with a high degree of accuracy. Thus the second exposure gave the β-ray spectrum of RaB in the same magnetic field. A fiducial mark put on the plate while still in the spectrograph permitted the determination of its position relative to the apparatus. Several experiments were performed in this manner. The measurements on all plates agreed with each other to at least one part in 1000. The results are tabulated below.

TABLE II

$H\rho$ and energy values of RaD β-ray spectrum

No.	$H\rho$	Intensity	Energy (volts$\times 10^{-6}$)
1	594.3	50	0.3033
2	600.3	3	0.3092
3	709.1	25	0.4268
4	735.2	10	0.4576
5	742.5	1	0.4663

The first four lines given above are those originally reported by Danysz in 1913, but the $H\rho$ values and the energies are considerably lower. In addition a faint line of higher energy has been observed which has previously escaped detection. This line appeared on all plates and there is no doubt of its existence, although it is too faint to measure with the same accuracy as the other lines. The value here given was determined from measurements of the plates independently by Mr. R. L. Chenault and by myself. As can be seen from the photographic reproductions, Plate I, there is no trace of the line $H\rho=628$ reported by Ellis. A careful scrutiny of all plates failed to reveal it. The line $H\rho=600.3$ (No. 2 in Table II) is clearly visible even in the photographic reproduction although Meitner failed to detect it. This failure was presumably due to the low dispersion with which she worked. The photographs reproduced in this paper are about one and a half times actual size, and illustrate the dispersion employed. *A* is a reproduction of one of the spectrograms for RaD and *B* shows the comparison spectrum of RaB.

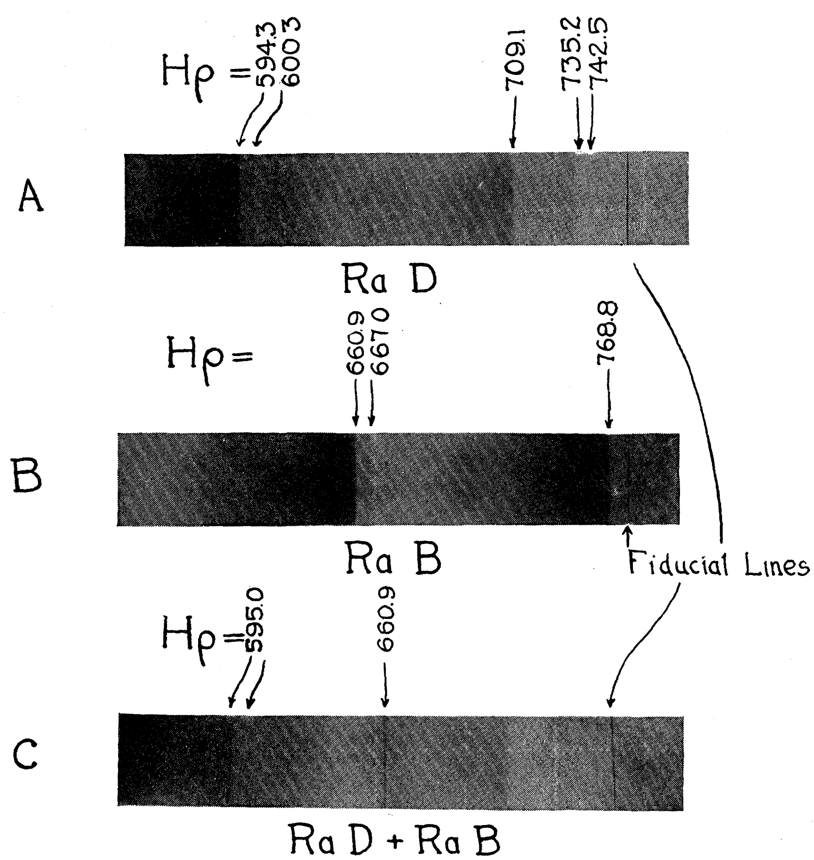

Plate I. Beta-ray spectra of RaD and RaB.

Since the values given in Table II differ consistently by at least one percent from the best previous measurements, a control experiment was considered necessary. The only objectionable feature of the foregoing procedure is the necessity for removing both plate and source between the RaD exposure and the calibration exposure with RaB. It would be much better to obtain the calibration simultaneously with the RaD exposure. Obviously only one line in the one spectrum need be checked against a corresponding line in the other. To this end the fine platinum wire on which the RaD had been deposited was exposed in radon in the usual way and activated with RaB. The wire was then placed in the spectrograph and the two spectra obtained on one plate by the single exposure. The measurements of this plate follow:

	ρ(observed)	$H\rho$	
RaB	6.365 cm	660.9	(Ellis and Skinner)
RaD	5.733 cm	595.0	(calculated)

These lines selected for comparison are the strongest lines in this region of each spectrum. As the above table indicates, the calculated value of $H\rho$ for the RaD lines is 595.0, in good agreement with 594.3 obtained from the first series of experiments. This combination photograph is reproduced in Plate IC.

The importance of the new line $H\rho = 742$ becomes evident when we consider the energy of the γ-ray responsible for this β-ray spectrum. Table III gives the energies corresponding to the observed $H\rho$ values. These energies, increased by the energy of the appropriate x-ray level as indicated, are computed for $Z = 82$ and 83.

TABLE III

Energy of γ-ray from RaD

		$Z=82$		$Z=83$	
$H\rho$	Energy (volts $\times 10^5$)	Level of origin	Energy of γ-ray (volts $\times 10^{-5}$)	Level of origin	Energy of γ-ray (volts $\times 10^{-5}$)
594.3	0.3033	L_{I}	0.4617	L_{I}	0.4669
600.3	0.3092	L_{II}	0.4612	L_{II}	0.4663
709.1	0.4268	M_{II}	0.4623	M_{I}	0.4668
735.2	0.4576	N_{III}	0.4643	N_{I}	0.4672
742.5	0.4663	——	0.4663	——	0.4663

The levels for atomic number 82 have been selected to give the best possible agreement, leaving all other considerations aside. Even so, the agreement is very poor. To secure approximate constancy for the energy of the γ-ray, it has been necessary to omit the M_{I} level and retain the M_{II} level. In the N group only the N_{III} level can be used. Such procedure is contrary to the weight of evidence of all observers in this field. It is now a well established fact that under the conditions here considered the levels will be present in the β-ray spectrum with intensities in the order of their binding energy. If we adhere to this principle, there is no agreement whatever. The newly observed line has an energy of 0.4663 times 10^5 volts. However, the best estimate of the energy of the γ-ray as deduced from the levels of $Z = 82$ in the above table is 0.462 times 10^5 volts. This involves the obvious paradox of a γ-ray causing the emission of β-radiation of still greater energy. It should be noted that the energy of the new β-ray line is more than one percent higher than that deduced for the γ-ray. This difference cannot be attributed to experimental error.

The last column of the table shows the excellent agreement obtained for the energy of the γ-ray when the levels for $Z=83$ are used. The energy is constant to one part in 500. Furthermore the intensities from the various levels follow in the order to be anticipated. Now the M_{I} level and the N_{I} level fit perfectly. No level is assigned to the new line since it is already approximately equal to the energy deduced for the γ-ray. The simplest explanation of this apparent equality is that this line represents particles ejected from the outer levels of the atom where the binding energy is relatively insignificant. The intensity favors this explanation very strongly. The other alternative is to accept Meitner's hypothesis that some at least of the primary β-radiation from the nucleus should escape with energy exactly equal to that of the γ-ray. However, the reason for such an assumption is not altogether clear. Furthermore, the intensity of this line does not agree with such an interpretation. This is discussed below.

The excellent agreement obtained for the various levels for atomic number 83 is in itself a strong argument for assuming that the γ-ray ejected the electrons in the β-ray spectrum from the levels of this atom. This means that the γ-ray is emitted after the emission of the primary β-particle when the electrons have adjusted themselves to their new energies. The fact that no agreement whatever can be obtained with the line $H\rho=742$ on the basis of the levels of atomic number 82 reinforces this argument most decidedly. Consequently it is safe to conclude that the γ-ray is emitted after disintegration in accord with Meitner's view.

The interesting question arises as to what becomes of the primary β-radiation. Ellis in his interpretation accounted for this radiation in the continuous spectrum. But he supposed the primary radiation to come after the emission of the γ-ray. Under these circumstances it is conceivable that the primary particles might come out of the nucleus with energies varying over a considerable range and thus constitute a continuous spectrum. Since it now appears fairly certain that the disintegration takes place *first*, we expect the primary particles to be emitted with sharply quantised energy. They should then of course appear as a line in the β-ray spectrum. In the case of RaD, conditions are particularly simple. It emits only one γ-ray. Therefore we should anticipate that the number of primary β-particles would be equal to the number of γ-rays emitted. If this were true the primary line would be the strongest line, since the energy of the γ-ray has been shared among the various atomic levels. This argument would rule out the line $H\rho = 742$ as a primary β-ray line.

There are at least two ways to account for the absence of the primary β-ray line. One possibility is a suggestion due to Rosseland.[7] He proposed the explanation that the particles originally leave the nucleus with a definite energy. In passing through the strong fields in the neighborhood of the nucleus they might be expected to radiate energy. He calculated this possible loss of energy by radiation and found that it was of the right order of magnitude to distribute the particles into a continuous spectrum. This explanation requires that variable portions of the energy be radiated and it is hard to see why this should happen. Much more information is needed concerning the intensity of the continuous spectrum before this question can be answered. Another possibility, though a somewhat doubtful one, enables us to eliminate both the continuous spectrum and the primary line from the β-ray spectrum. If we adopt Meitner's view and assume that the primary particle has energy exactly equal to that of the γ-ray, we may suppose that this primary particle experiences an inelastic collision with the first electron in the atomic levels which has less binding energy than that of the primary particle. The result would be a binding of the β-particle in the atomic level and the ejection of the electron originally in the level. However, this ejected electron should have exactly the same energy as those expelled by the γ-radiation. Therefore they would form part of the same line on the photographic plate. Presumably this could take place at other levels of lower energy so that most or all of the primary particles were absorbed. Therefore we should have little or no primary β-radiation and no continuous spectrum. The great difficulty with this hypothesis is that such collisions from any ordinary point of view must be relatively improbable. However, to explain the intensity of the lines, considering all γ-radiation to be converted in the atomic levels of the atom from which it originates, we are forced to assume an abnormally high coefficient of absorption for the γ-ray. It may well be that the same is true for a β-particle travelling from the nucleus out through the various electron levels. Since there is considerable radiation from neighboring atoms present, some support for this view can also be obtained from Milne's[8] statistical analysis of the liberation of photo-electrons. He finds that he is compelled to assume that external radiation can stimulate the capture of an electron. This might make it possible to account for the high probability of capture necessary on this view.

In conclusion, I should like to thank Dr. S. C. Lind of the Fixed Nitrogen Laboratory, for providing me with the old radon tubes from

[7] Rosseland, Zeits. f. Physik **14,** 173 (1923).

which the RaD sources were obtained, and Dr. Bardwell for much valuable advice concerning the manipulation of this material.

Note added to proof: Since the above paper was submitted for publication the library has received the issue of the Proceedings of the Cambridge Philosophical Society for November, 1925. A series of remarkable papers in this number present evidence indicating that the gamma-ray follows the disintegration for RaB and RaC. Rutherford and Wooster (Proc. Camb. Phil. Soc. **22**, 832, 1925) show that RaB emits the natural L spectrum of atomic number 83. Of course this is not a conclusive proof of the emission of the gamma-ray after disintegration. There is a possibility that the gamma-ray is emitted before disintegration but that the *x-ray transitions* are delayed and occur after the disintegration. The significant point of their result is that it does not contradict the supposition that the disintegration occurs first. If, on the other hand, the x-ray spectrum of atomic number 82 had been obtained there could be no doubt that the disintegration followed the gamma-ray emission. In confirmation of the results obtained by Rutherford and Wooster, Black has measured some of the lines of the natural L beta-ray spectrum of RaB. He also finds that these correspond to atomic number 83. Since this phenomenon is one step further removed from the emission of the gamma-ray than the one investigated by Rutherford and Wooster, it likewise does not constitute a *proof* that the gamma-ray follows disintegration. However, Ellis and Wooster (Proc. Camb. Phil. Soc. **22**, 844, 1925) have devised an experiment which seems to give definite proof. They wrapped a radon tube in platinum foil and then exposed the platinum surface in radon and obtained the active deposit on the outside of the platinum wrapping. Using this as a source in a beta-ray spectrograph they obtain the natural beta-ray spectra of RaB and RaC from the active deposit and the similar spectra excited in the platinum foil by the gamma radiation from the radon. They thus obtain a $\delta H\rho$ for each of the observed lines corresponding to the difference between the K, L, etc., energy for platinum and for RaB and RaC. This enables them to determine whether these levels are those of 82, 83 or 84. The measurements given in their paper very decidedly favor atomic number 83 for the lines of the RaB spectrum and atomic number 84 for the lines of the RaC spectrum. These results are thus a confirmation of the hypothesis proposed by Meitner that the disintegration *precedes* the emission of the gamma-ray, and there now seem to be no exceptions to this rule.

Bureau of Standards,
Washington, D. C.,
December 15, 1925.

[8] E. A. Milne, Phil. Mag. **47**, 222 (1924).

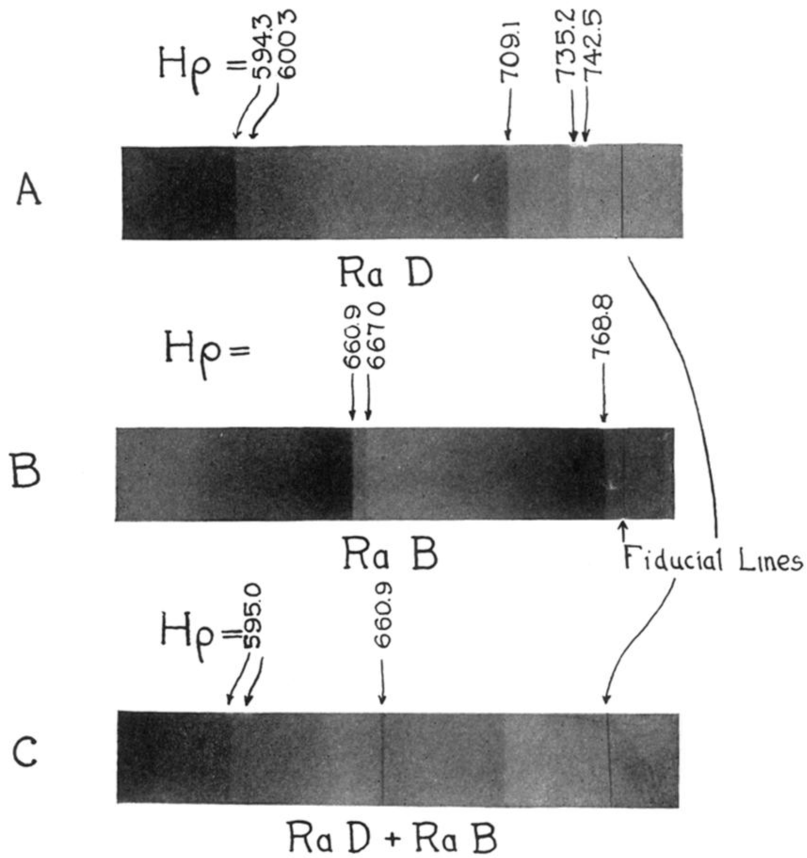

Plate I. Beta-ray spectra of RaD and RaB.