# Measurement of the Pb *M* Photon Intensity in Po<sup>210</sup> Decay—A Further Gross Inadequacy of the Theory\*

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The probability that a Pb M photon is emitted in Po<sup>210</sup> decay is  $0.91(\pm 15\%) \times 10^{-3}$  photons/alpha, as measured by means of a proportional counter and pulse-height analyzer. This value is in gross disagreement with theoretical estimates. A Pb M spectrum from Po<sup>210</sup> decay shows definite differences from a conventional Pb M spectrum, indicating considerable ejection of higher shell electrons in the Po<sup>210</sup> decay act. In particular, the spectrum from Po<sup>210</sup> contains a 3.7-keV component whose intensity is about 4% of the total spectral intensity and whose energy is consistent with the allowed, but hitherto unreported, transition  $M_1O_{23}$ . The theory of electron ejection in alpha decay is reviewed and previous calculations of ejection probabilities are extended to include ejection to bound states. The estimates of K, L, and M photon yields on this theory are orders of magnitude lower than the measured yields. Possible sources of the discrepancy are discussed.

HE published theories of the ejection of electrons from the electron cortege of a nucleus undergoing alpha decay underestimate by orders of magnitude the measured Pb K and L x-ray intensities that accompany Po<sup>210</sup> decay (references are given at appropriate places below). In Part A of the present paper we report a measurement of the Pb M x-ray intensity from Po<sup>210</sup> decay that deviates from theoretical estimates even more than do the K and L intensities, as one would expect. The measurement is described in considerable detail in order to convince the reader (and the author) that no gross error in the measurement contributes to the astonishingly large discrepancy with the theory, and also to demonstrate that a Pb Mspectrum from Po<sup>210</sup> decay differs significantly from one obtained by conventional means.

The gross inadequacy of the theory for K and Lejection has subsisted unattacked for a long time. Also, the fact that the first of the published theories, Migdal's, accidently gives a reasonable result for K ejection has led some to believe that the theory is adequate for Kelectrons. Consequently we have judged it useful to append, as Part B, a compact review of the theory as it exists at present, together with a tabulation of all the published measurements of K and L, as well as Mejection probabilities. Measured values and theoretical estimates are compared in Sec. V. Possible sources of the inadequacy of the theory are discussed in Sec. VI.

When this paper was completed G. W. Schaefer reported (private correspondence) that some very recent calculations of his, which bypass the use of an asymptotic expansion in the theory, reduce to reasonable proportions the discrepancy between theory and experiment in the case of K electron ejection. With his amiable permission, the results of his as yet unpublished calculations are described in Sec. VI.

#### A. MEASUREMENT OF THE Pb M PHOTON YIELD IN Po<sup>210</sup> DECAY

#### I. Experimental Details

The procedure for measuring the number of Pb Mphotons per Po<sup>210</sup> decay was essentially the same as that described some time ago for a similar measurement of the L photons,<sup>1</sup> with some modifications made necessary by the softness of the Pb M rays.

## A. Photon Detection

Photons from a Po<sup>210</sup> source of known alpha disintegration rate and at known geometry pass through a Be window  $(17.3 \text{ mg/cm}^2)$  into an aluminum-lined proportional counter about 4 in. in diameter and 12 in. long, fitted with a stainless-steel center wire of 0.004-in. diam, and filled with one atmosphere of a 9-to-1 mixture of argon and methane. The center wire is kept at about +2600 V with respect to the grounded counter wall by means of a stable high voltage supply. The pulses from the counter are fed through the conventional preamplifier and nonoverloading linear amplifier and recorded on a commercial 100-channel pulse-height analyzer. The stability of the instrument was entirely adequate; peak positions could be reproduced to better than 1%over a period of several days.

## B. The Po<sup>210</sup> Sources

About 30 mC of Po<sup>210</sup> was obtained from the Monsanto Chemical Co. as an invisible quantity of the dry nitrate. The supplier stated it to assay at 99.9% Po<sup>210</sup>, possible impurities being Bi and Pt. Without further purification, the  $Po^{210}$  was taken up in 3 ml of a 2.4N HNO<sub>3</sub> solution that had been prepared with triply distilled water. This constituted our Po210 stock solution which, assayed by the method of dilution and alpha counting, proved to have a specific activity of 1.956  $\times 10^{10} (\pm 2\%)$  disintegrations per minute per ml (8.81 mC/ml), a value about 1% higher than that computed from the supplier's stated assay.

To prepare a source, a 5 to  $50 \lambda$  aliquot of the stock solution was pipetted into a polystyrene cup 9.86 mm in diameter and 3.2 mm deep, and slowly evaporated, together with the same volume of pipette rinse (2.4N) $HNO_3$ ) under a heat lamp. The resulting  $Po^{210}$  deposit

<sup>\*</sup> Research performed under the auspices of the U. S. Atomic Energy Commission. <sup>1</sup> W. Rubinson and W. Bernstein, Phys. Rev. 86, 545 (1952).

(presumably nitrate or oxide) was distributed over a roughly circular spot 2 to 8 mm in diameter, depending on the aliquot volume, the distribution being fairly uniform except for a considerable concentration of activity at the periphery of the spot.

In order to test for possible effects of source thickness, sources of strengths ranging over a factor of ten were prepared, namely, with aliquot volumes 5, 10, 25, and  $50 \lambda$  (0.044 to 0.44 mC). For each of these strengths, at least two sources were made. The measured x-ray intensities proved to be independent of source strength over this range.

# C. Source Purity

The intensity of the Pb M radiations under study decays with the 138.4-day half-life of Po<sup>210</sup>. This suffices to establish that these radiations are produced in the Po<sup>210</sup> decay act provided that two possible causes of error are eliminated:

(i) The sources may be contaminated with some radionuclide of half-life not greatly different from that of Po<sup>210</sup> and having among its radiations one that could be mistaken for a Pb M ray. But any such known possible contaminant activity would reveal its presence through other radiations; for example, the e-capture nuclide 180-day Au<sup>195</sup>, which emits an  $\sim$ 2-keV Pt M x ray that falls in the energy range of Pb M rays, also emits characteristic K, L, and  $\gamma$  radiations. Such characteristic radiations were looked for a spectra from our sources covering the energy ranges 4-20 keV (proportional counter), 15-125 keV (thin crystal), and 0.1-2 MeV (thick crystal). In each of these ranges there is a known electromagnetic radiation of Po<sup>210</sup>, namely,  $\sim 10.5$ -keV Pb L rays,  $\sim 80$ -keV Pb K rays, and 0.8-MeV  $\gamma$ 's, whose intensities per Po<sup>210</sup> decay are, respectively,  $3 \times 10^{-4}$ ,  $1.5 \times 10^{-6}$ , and  $1.5 \times 10^{-5}$  (see Table IV for references). No contaminant radiation was detectable, though any such would have been obviously present if its intensity were as great as one-tenth that of the Po<sup>210</sup> radiation in the corresponding range. Consequently, contaminant activities could not have contributed more than a few percent to the Pb M photon yield of  $\sim 10^{-3}$  that we report below.

(ii) The sources may contain chemical impurities that are excited to x-ray emission by the alpha bombardment to which they are subjected. Possible chemical contaminants whose K, L, or M x rays have energies lying in the range of Pb M energies are P, S, Cl, the elements  $_{38}$ Sr to  $_{48}$ Cd, and the elements  $_{77}$ In to  $_{84}$ Po. A theoretical estimate of the corresponding K and Lintensities was made with the help of cross sections computed from curves given by Merzbacher and Lewis.<sup>2</sup> Per  $\mu$ g of impurity the impurity K photon intensity would be about 1%, and the impurity Lphoton intensity would be about 3%, of the observed Pb *M* photon intensity from our sources. The estimate of *K* photon intensity is verified roughly by our observation that the intensity of Ar *K* rays from the 16.5  $\mu$ g of Ar contained in 1 cc of air between a source and the counter window was ~15% of the Pb *M* intensity from the source, a result that is also consistent with reported measurements<sup>3</sup> on *K* photon production in S and Ca under Po<sup>210</sup> alpha bombardment.

On the basis of these estimates, the sources certainly contained no impurities sufficient to account for any but a small fraction of the M photon intensity that we observed. By direct chemical analysis of stock solution aliquots equal to those used for source preparation, there were no detectable quantities of PO43-, SO42or Cl<sup>-</sup>, the limits of detection being 1  $\mu$ g of S or Cl (by nephelometric analysis) and 0.1  $\mu$ g of P (by spectrophotometric analysis). The elements Sr to Cd, whose L radiations are of concern, were not analyzed for, but they would have been detected under the microscope or by way of their K radiations if they had been present in quantities ( $\sim 30 \ \mu g$ ) sufficient to double the true Pb M photon intensity. In any case, it is highly unlikely that the Po<sup>210</sup> stock solution contained these comparatively rare elements in view of the absence of significant amounts of such ubiquitous contaminants as P, S, and Cl. There remains the possibility that Mradiations from impurity elements Ir to Po made a significant contribution to the observed M intensity. But such quantities of impurities would have been strikingly apparent under the microscope. For example, a Po<sup>210</sup> source containing 30  $\mu$ g of added Pb, in which numerous crystallites of Pb(NO<sub>3</sub>)<sub>2</sub> were visible under 100 $\times$  magnification, showed a Pb M intensity only  $\sim 50\%$  greater than that of the clean sources which contained no visible impurities under 100× magnification.

As to the possibility that impurities giving rise to x rays were contained in the material of the plastic source mount, this was eliminated by the observation that the Pb M intensity from a source in a plastic cup was essentially the same as that from a source in a cup made of pure Cr.

In view of all these tests, it is fairly certain that the  $Po^{210}$  sources contained no significant quantities of radioactive or chemical impurities, at least at the time of preparation. Any contamination subsequent to the time of preparation would manifest itself in anomalous half-lives and lack of reproducibility of results from replicate sources.

#### D. X-Ray Counting Efficiency

The counting arrangement is shown in Fig. 1. A source cup fits snugly in a well in a Lucite block which, in turn, is fastened to a Lucite disk  $\sim 6$  mm thick, pierced with an aperture 9.63 mm in diameter, the upper surface of which lies ordinarily 11.76 mm above

<sup>&</sup>lt;sup>2</sup> E. Merzbacher and H. W. Lewis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 34, p. 166.

<sup>&</sup>lt;sup>3</sup> W. Bothe and H. Fränz, Z. Physik 52, 466 (1929).

the plane of the source. The disk can be positioned reproducibly 2 mm below the Be counter window. Aperture and counter window are accurately centered on the axis of the well. The well depth can be varied by means of spacers, and provision is made for flowing gases through the well to replace the air. In this counting arrangement, with the source at the standard distance, 11.76 mm below the aperture, the probability that the counter detects a photon emitted by the source is  $0.0148\pm0.0010$  (the indicated error being an extreme), determined empirically as the product of solid angle and window transmission. Effectively all photons that enter the counter gas are recorded. The details of the solid angle and the window transmission measurements were the following.

With the source 11.76 mm below the aperture, the solid angle for acceptance of photons by the counter is  $0.0364\pm2\%$ , determined by taking the ratio of the counting rates of a source at the standard distance (11.76 mm) and at 51.12 mm below the aperture, and multiplying this ratio by the computed solid angle for the 51.12 mm distance, for which the computation can be made with negligible error. The counting rates were taken with the space between source and window filled with He, which has negligible absorption for these x rays. The value adopted for the solid angle, 0.0364, includes 4% correction on the counting-rate ratio that takes account of the different average window thicknesses encountered by photons from the two source positions.

The fraction of the incident Pb M spectrum that escapes absorption in the 17.3  $mg/cm^2$  Be counter window is  $0.407 \pm 5\%$ , as determined directly from the decrease in photon intensity when additional Be absorbers were placed over the counter window. In general, this method of measuring the transmission factor of a spectrum results in a value that is too high because the spectrum grows harder with increasing absorber thickness. But in the present case the method is quite accurate, as shown by a comparison of the measured value, 0.407, with computed values 0.40-0.426 obtained on the basis of known Be absorption coefficients,<sup>4</sup> the known Pb M line energies,<sup>5</sup> and various reasonable guesses concerning the relative intensities of the Pb M lines<sup>6</sup> (the individual M line intensities cannot be obtained from our own data because of strong overlapping).



FIG. 1. Source mount on counter.

#### II. Discussion of the Observed Spectra

The identification of the observed radiations as Pb Mlines was established by comparing a spectrum from one of our standard sources with a spectrum of known Pb M lines produced by the alpha bombardment of Pb. A source of the latter, the "Pb source," was made by evaporating an aliquot of the Po<sup>210</sup> stock solution in a Pb metal cup of the same dimensions as the standard source cups. In the course of evaporation, the nitric acid in the aliquot attacked the Pb metal, leaving a residue of several mg of PbO in which the Po<sup>210</sup> atoms were imbedded. They were deeply enough imbedded to cause extensive absorption of the soft photons emitted in Po<sup>210</sup> decay, so that the radiations from this source consisted almost exclusively of x rays excited by alpha bombardment of Pb atoms close to the surface of the PbO, as we can infer from the following considerations.

When a spectrum from a standard source was taken with air between source and counter window, about 15% of the observed total intensity consisted of Ar K rays excited by alpha bombardment of the ~15  $\mu$ g of Ar in the air. No Ar rays could be detected in a corresponding spectrum from the Pb source. It follows that the alphas in the Pb source suffered considerable energy loss before escaping into the air. But a PbO thickness sufficient to stop the alphas from the decay of a given Po<sup>210</sup> atom is sufficient to stop the photons from the same decay.<sup>7</sup> We mention that the total intensity from the Pb source was about three times that from a standard source. The shape of the spectrum from the Pb source was probably somewhat distorted by selfabsorption.

The identification of the radiations from a standard source with those from the Pb source was made on spectra taken with  $N_2$  between source and counter window to avoid excitation of Ar x rays. The spectra, shown in Fig. 2, were each normalized by equating to

<sup>&</sup>lt;sup>4</sup> Values obtained by interpolating data in the compilation by C. E. Ehrenfried and D. E. Dodds, AFSWC-TN-59-33 (January 1960).

<sup>&</sup>lt;sup>6</sup> Ý. Cauchois and H. Hulubei, Longueurs d'Onde des Émissions X et des Discontinuités d'Absorption X (Hermann & Cie, Paris 1947). They list 16 diagram and 8 nondiagram lines.

<sup>&</sup>lt;sup>6</sup> Visually estimated intensities are reported by E. Hjalmar, Z. Physik **15**, 65 (1923), and the order of relative intensities is described by E. Lindberg, Nova Acta Reg. Soc. Sci. Upsala 7, No. 7 (1931). According to A. E. Sandström, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 30, p. 236, the old intensity measurements are unreliable. No newer measurements appear to be available.

<sup>&</sup>lt;sup>7</sup> The range of Po<sup>210</sup> alphas in Pb is ~15 mg/cm<sup>2</sup> as computed from Fig. 2 of "1960 Nuclear Data Tables, Part 3" by J. B. Marion (USAEC, Washington, 1960), and the Pb mass absorption coefficient for the photons of interest is ~1200 cm<sup>2</sup>/gm. Note that a thickness of 15 mg/cm<sup>2</sup> is provided by PbO crystallites of ~15- $\mu$  diam.



FIG. 2. Pb M spectra from Po<sup>210</sup> decay and from Pb bombarded with Po<sup>210</sup> alpha particles. Data taken with N<sub>2</sub> between source and counter window.

100 the counts in the channel at peak maximum, which were  $\sim 3000$  and  $\sim 1500$ , respectively, for the standard source and the Pb source.<sup>8</sup> Although the peak shapes differ in a nontrivial way, the positions of the two peak maxima, which can be fixed within an extreme experimental uncertainty of  $\sim \pm 1/2$  channel, or  $\sim \pm 0.03$ keV, are effectively identical, their common value, 2.40 keV, being consistent with the known energies of the Pb *M* lines (see Table I). We consider that in view

TABLE I. Pb M diagram lines.ª

Transition	Other designation	Energy (keV)	Qualitative intensity <sup>b</sup>
$\begin{array}{c} \hline & M_5N_7 \\ M_4N_6 \\ M_3N_5 \\ M_2N_3 \\ M_2N_4 \\ M_1N_3 \\ M_3O_5 \\ M_3O_1 \\ M_2N_1 \\ M_3N_4 \\ M_4N_3 \end{array}$	$lpha_1\ eta\ \gamma\  au\  au\  au$	$\begin{array}{c} 2.346\\ 2.443\\ 2.652\\ 1.839\\ 3.121\\ 3.202\\ 3.045\\ 2.921\\ 2.663\\ 2.630\\ 1.942\end{array}$	strongest 2nd strongest 3rd strongest 4th strongest rather strong very weak

a See reference 5. The factor used for conversion from x units to keV was 12372. b E. Lindberg, reference 6.

of the purity tests described in Sec. IC, the differences in peak shapes are not due to contaminant radiations, but, rather, must be ascribed to the two different processes of spectral excitation: The component lines in the spectrum emitted by the excited Pb daughter of  $Po^{210}$  decay have relative intensities different from those in the spectrum of Pb excited by alpha bombardment. However, even considerable variations in the relative intensities of the component lines have only a small effect on the position of the peak maximum, as shown in the Appendix, so the effective identity in the positions of the peak maxima in Fig. 2 suffices to establish that the radiations from  $Po^{210}$  are Pb *M* rays. By comparison, the radiations closest in energy to the Pb *M* lines, namely, the *M* lines of Tl and Bi, would peak at ~2.35 and ~2.51 keV, respectively, as shown by computations based on Eq. (A4) of the Appendix.

Some inferences can be drawn concerning the relative intensities of the component lines in the spectra of Fig. 2 by comparing these spectra with synthetic spectra computed from Eq. (A2) of the Appendix with various trial values of the relative intensities. In Fig. 2. the experimental half-width (full-width at half-maximum) of the spectrum from the Pb source is  $9.7 \pm 0.2$ channels (the error being an extreme), a value that is effectively that of monochromatic Pb  $M\alpha$  or  $M\beta$  radiation. Such a half-width is possible only if the  $\alpha$  and  $\beta$ intensities together constitute at least 90% of the total spectral intensity; as shown by the construction of synthetic spectra, a superposition of the  $\alpha$  and  $\beta$  lines alone would have a half-width of  $10.0\pm0.25$  channels for all relative intensities  $\alpha:\beta$  between about 2:3 and 5:2, while the addition of  $\gamma$  line intensity equal to 10% of the total intensity would increase the half-width to  $10.4 \pm 0.25$  channels, i.e., outside the error in the halfwidth of the Pb source spectrum. Thus, if the latter spectrum is assumed to consist of an overlap of exclusively the Pb  $M\alpha$ ,  $\beta$ , and  $\gamma$  lines, the contribution of the  $\gamma$  intensity to the total intensity is less than 10%

<sup>&</sup>lt;sup>8</sup> The correspondence between channel number and energy in Fig. 2 was established by means of an auxiliary V<sup>49</sup> source which provides the two known energies necessary to fix a linear energy scale, namely, the 4.512-keV Ti K line, and the 1.53-keV escape peak that accompanies it when the counter gas is Ar.



FIG. 3. Pb M spectrum from Po<sup>210</sup> decay. Data taken with He between source and counter window. The triangled points designate a synthetic spectrum computed as described in the text.

in the spectrum as observed, and less than 7.5% in the spectrum corrected for absorption in  $\rm N_2$  and the Be window.

The spectrum from the standard source in Fig. 2, on the other hand, has a half-width of  $10.8\pm0.2$ channels, which indicates that significant intensities of other radiations are present in addition to the  $\alpha$  and  $\beta$ . Subtraction of the Pb source spectrum from the standard spectrum reveals that the latter contains radiations of energies 1.9, 2.9, and 3.7 keV, whose respective intensities are about 5%, 7%, and 11.5% of the total observed intensity, or, after correction for absorption in N<sub>2</sub> and Be, 24%, 3%, and 4%. The 1.9and 2.9-keV peaks are consistent with known Pb M line energies listed in Table I. The 3.7-keV peak corresponds to no known Pb M diagram line, and its energy is suspiciously close to that of the 3.694 keV Ca K line. However, chemical analysis (nephelometry) of the Po<sup>210</sup> stock solution showed that the amount of Ca in the source was less than 2  $\mu$ g, so that, by a calculation of the kind described in Sec. IC, Ca K lines could account, at most, for only a twentieth of the observed 3.7-keV intensity. We are, therefore, forced to conclude that the 3.7-keV line is characteristic of  $Po^{210}$  decay. A transition giving rise to such a Pb M line would be  $M_1O_{2,3}$  (3.765 keV), a line that has not been reported, though it is allowed by the selection rules. Its possible existence in the spectrum from Po<sup>210</sup> supports an argument that we will make in the theoretical discussion, namely, the Pb daughter of Po<sup>210</sup> decay must be stripped of its N and O electrons to an extent that x radiations otherwise suppressed by the Auger process can be emitted.

In addition to the 3.7-keV line (which we will designate " $M_1O_3$ "), one other difference between the Pb M spectrum from Po<sup>210</sup> and a conventional Pb M

spectrum can be demonstrated with some certainty. In Fig. 3 the circled points show a (normalized) spectrum from a  $Po^{210}$  source, taken this time with He between source and window to emphasize the low-energy components. The triangled points show a synthetic spectrum computed from Eq. (A2) of the Appendix, with the energies given in Table I and with a selected set of relative intensities roughly consistent with the qualitative intensity estimates made on conventional spectra.<sup>6</sup> The intensities selected were

$$\alpha:\beta:\gamma:\tau:M_2N_4:"M_1O_3"=100:66.7:33.3:55:10:9.5$$

(corrected to 100:75.7:44.6:18.2:18.9:19 to take account of absorption in the Be window of the counter). The synthetic spectrum, the computation of which, it should be emphasized, depends in no way on data obtained with Po<sup>210</sup> sources except for the " $M_1O_3$ " data, reproduces the observed spectrum closely except for the difference peak at 1.58 keV which contributes about 4% of the total intensity and is probably the 1.489-keV Al K line excited in the Al liner of the counter. In order to achieve this agreement we were forced to assume that the intensity of the 1.839-keV  $\tau(M_5N_3)$ line (or, perhaps, the sum of the intensities of  $M_5N_3$ ,  $M_4N_3$ , and  $M_4N_2$ ) is greater than the sum of the intensities of the 2.652-keV  $\gamma(M_3N_5)$  line and the 3.121-keV  $M_2N_4$  line, in contrast to the order of line intensities observed in spectra taken by conventional means (see Table I). Such an intensity reversal is consistent with a theoretically expected preferential ejection of  $M_5$  electrons and of  $N_4$  and  $N_5$  electrons in the Po<sup>210</sup> decay act.

To summarize the rather involved argument of this section: A source prepared by evaporating an aliquot of the  $Po^{210}$  stock solution in a Pb metal cup emits a Pb M spectrum with little admixture of the radiations

from the polonium. A spectrum from this source and one from a standard source have effectively identical positions in peak maxima, though the peak shapes differ. The identity in peak position suffices to establish that the soft radiations from  $Po^{210}$  are Pb M lines. The differences in peak shapes are ascribed to the different relative intensities of the component lines resulting from the different modes of spectral excitation. The spectrum from Po<sup>210</sup> can be closely reproduced by a synthetic spectrum, in the computation of which the relative intensities of the component lines are assumed to be  $\alpha:\beta:\gamma:\tau:M_2N_4:::M_1O_3:=100:66.7:33.3:55:10:$ 9.5. The presence of the " $M_1O_3$ " line and the high intensity of the  $\tau$  line, which are in definite contrast to spectra obtained by conventional means, are taken as indications that in the process of Po<sup>210</sup> decay there is enough ejection of electrons from shells higher than the *M* shell to affect significantly the relative intensities of the M lines.

It is worth repeating that the purity tests described in Sec. IC insure that only a minor fraction of the observed x-ray intensity can be attributed to the presence of P, S, Cl, and Ca chemical impurities in the sources.

#### **III.** The Measured Photon Yield

The measured Pb M photon counting rates of six different sources are shown in Table II. Each rate is a

	(counts	$/min)/\lambda$	Ratio
Source	Fresh <sup>b</sup>	Aged <sup>e</sup>	fresh/ageo
$50\lambda - 1$	286.9	279.5	1.026
$50\lambda - 4$	264.3	279.7	0.945
$10\lambda - 1$	270.5	•••	•••
$10\lambda - 2$	267.7	268.6	0.997
$5\lambda - 1$	278.6		
$5\lambda - 3$	281.8	279.6	1.008
Mea	in of fresh sourc	es = 275.0	
Mea	an deviation $= 7$	5 = 2.7% of m	ean

TABLE II. Pb M photon counting rates.<sup>a</sup>

<sup>a</sup> The tabulated counting rates are those with He between source and counter window. See text. -<sup>b</sup> Counted within 24 days after time zero, and corrected back. <sup>c</sup> Counted about two half-lives later, and corrected back.

sum of the counts accumulated in the  $\sim 60$  channels over which the spectrum was spread, corrected for a background 2-10% of the total count, and divided by the aliquot volume and the counting time. In the source designation, the aliquot volume of Po<sup>210</sup> stock solution used to prepare the source is given as, e.g.,  $50 \lambda$ .  $1 \lambda$  of stock solution had a Po<sup>210</sup> disintegration rate of 1.956  $\times 10^{7} (\pm 2\%)$  dis/min at time zero (see Sec. IB). The tabulated photon rates, which have been corrected back to time zero, are those for sources in the standard counting arrangement described in Sec. ID, with He between the source and window. The actual counting was done with an air atmosphere, and correction to a He atmosphere was made by multiplying the observed

rate in air by  $1.60(\pm 2\%)$ , the number obtained in an auxiliary measurement of the ratio of the counting rates in He and air. For each source, about 10<sup>5</sup> counts were accumulated.

We have rejected the results obtained with five other sources, one of which was 23% lower, and the others 14 to 40% higher, than the mean rate reported here, on the ground that in four of these cases the half-life of the x-ray intensity differed by 10% or more from that of Po<sup>210</sup>, and that in the fifth case the source cup was swelled from overheating during source preparation. The cause of these discrepancies was not investigated.

As the observed counting rate we take the mean of the "fresh" rates in Table II, namely, 275.0 counts/  $\min/\lambda$ . Then the Pb *M* photon yield is this number multiplied by 0.96 to correct for the "Al radiation" (mentioned in Sec. II), divided by the counting efficiency 0.0148 (Sec. ID), and finally divided by the Po<sup>210</sup> disintegration rate  $1.956 \times 10^7$  dis/min/ $\lambda$  (Sec. IB). The result is

M photon yield = M photon rate/Po<sup>210</sup> decay rate  $=0.91 \times 10^{-3} (\pm 15\%),$ 

the indicated error being an extreme. An old measurement on the yield of soft photons from Po<sup>210</sup> by Curie and Joliot gave the value  $1.5 \times 10^{-3}$ , i.e.,  $\sim 65\%$  more than the value reported here (see Table V).

We mention that the L photon yield measured on the same sources proved to be  $2.79 \times 10^{-4}$ , a result equal to 0.95 the result previously reported<sup>1</sup> as 2.93  $\times 10^{-4} (\pm 15\%).$ 

# APPENDIX TO PART A

# Insensitivity of Peak Position to Variations in Component Intensities

Auxiliary experiments show that an experimental peak observed with monochromatic x rays can be fitted to a Gaussian over an energy range that includes at least 90% of the peak area. Empirically the half-width (full width at half-maximum) of an experimental monochromatic peak is proportional to the square root of the energy, so that, in view of a half-width of 0.857 keV obtained with the 4.512-keV Ti K radiation from a V<sup>49</sup> source, monochromatic radiation of energy E has

half-width = 
$$0.403E^{1/2}$$
 keV. (A1)

The standard deviation of a Gaussian is 0.425 the half-width. Then, given the energies and intensities of any set of x-ray lines, the corresponding experimental composite spectrum can be represented with considerable accuracy by

$$y(x) = \operatorname{const} \sum_{i} \frac{a_i}{\sigma_i} \exp\left[-(x - E_i)^2 / 2\sigma_i^2\right], \quad (A2)$$

where y(x) is the intensity (per unit energy range) of the composite spectrum at energy x; and  $E_i$ ,  $a_i$ ,  $\sigma_i$  are, respectively, the energy, intensity, and computed standard deviation of the *i*th component line.

We apply Eq. (A2) to the case of the Pb M spectrum, with the assumption that the latter consists of an overlap of only the three most intense lines, namely,  $\alpha, \beta, \gamma$  in Table I. These lines are so closely spaced in energy that their Gaussians can be assumed to have effectively the same half-width, which we take to be the mean of the individual half-widths computed from (A1), namely, 0.625 keV. Under these assumptions, the position of the composite peak maximum is the solution of

$$x = \frac{\sum_{i} a_{i}E_{i} \exp\left[-(x-E_{i})^{2}/2\sigma^{2}\right]}{\sum_{i} a_{i} \exp\left[-(x-E_{i})^{2}/2\sigma^{2}\right]} \quad (i=\alpha, \beta, \gamma) \quad (A3)$$

where  $\sigma$ , the common standard deviation of the Gaussians, has the value 0.265 keV.

The first iteration solution of (A3),

$$x = \sum_{i} a_{i} E_{i} / \sum_{i} a_{i}, \quad (i = \alpha, \beta, \gamma)$$
 (A4)

suffices for our purposes, since for all relative intensities roughly consistent with published rough intensity estimates,<sup>6</sup> the second iteration solution corrects the first by only  $\sim 0.01$  keV, which is less than the experimental uncertainty in the peak position.

With the  $\alpha$ ,  $\beta$ ,  $\gamma$  energies listed in Table I and assumed relative intensities  $\alpha:\beta:\gamma(\alpha \geq \beta \geq \gamma)$  lying between 1:1:1 and 6:4:1, the peak positions computed from (A4) lie between 2.480 and 2.409 keV.

### B. COMPARISON OF MEASURED AND THEORETICAL VALUES

#### **IV.** Review of Theory

The model and the approximations are those of the first of the theories, Migdal's,<sup>9</sup> with the very important modification, introduced by Levinger,<sup>10</sup> that the effect of nuclear recoil is taken into account. We introduce nuclear recoil in a way alternative to Levinger's by centering the coordinate system on the recoiling daughter nucleus, as suggested by Schwartz<sup>11</sup> and correctly worked out by Grard.12 We extend the previous calculations to include the probabilities that the electrons are ejected to bound states as well as to the continuum. In the following exposition atomic units are used exclusively, and the formalism is that of Dirac.13

We choose a coordinate system in which at times t < 0 the Po<sup>210</sup> nucleus bearing its complement of electrons moves along the negative z axis in the positive z direction with velocity **u**. When it reaches the origin,

at t=0, it decays, the resulting Pb<sup>206</sup> daughter being brought to a stop at the origin while the alpha moves off along the positive z axis with velocity **v**. The initial magnitude of **u** and the terminal magnitude of **v** are, respectively, 0.14 and 7.32 a.u., in view of the 5.3-MeV decay energy of Po<sup>210</sup>.

If  $\psi$  designates an eigenstate of an electron in a fixed Po<sup>210</sup> atom of nuclear charge Z+2, the corresponding state in the atom moving with velocity **u** (at times t < 0) is  $e^{iuz} \psi(x, y, z-ut)$ ), so that the electronic state at t=0, the instant of decay, is

$$e^{iuz}\psi(x,y,z)\rangle.$$
 (1)

This is the initial state. A final state is  $|E'\rangle$ , an eigenstate belonging to the energy eigenvalue E' of the Hamiltonian E of an electron in the field of the Pb nuclear charge Z. Let  $|E^0\rangle$  be the eigenstate of E that corresponds to the eigenstate  $\psi$  of an electron in a Po atom fixed at the origin. We write

$$\psi \rangle = |E^0\rangle + |1\rangle. \tag{2}$$

For  $t \ge 0$ , the Hamiltonian is that of an electron moving in the combined fields of a Pb nucleus fixed at the origin and an alpha particle moving along the z axis with velocity v, i.e.,

$$H(t) = E + V(t), \tag{3}$$

where V(t) is the electrostatic potential between the electron and the alpha particle,

$$V(t) = -2[x^{2} + y^{2} + (z - vt)^{2}]^{-1/2}.$$
(4)

Note that by the adoption of this form of V(t) we are assuming that the alpha particle is emitted with zero angular momentum with respect to the nucleus and instantaneously assumes its full terminal velocity.

At t=0 Eq. (4) gives the potential due to an alpha particle located at the origin, so that H(0) in Eq. (3) is the Hamiltonian of which  $\psi$  is an eigenstate, belonging to eigenvalue  $H^0$ , say. Then on taking the  $|E'\rangle$  component of  $H(0)|\psi\rangle = H^0|\psi\rangle$  with  $\psi\rangle$  given by Eq. (2), we obtain

$$\langle E' | \mathbf{1} \rangle = - \frac{\langle E' | V(0) | E^0 \rangle}{E' - E^0} + O(Z^{-2}) \text{ terms } (E' \neq E^0),$$
 (5)

an expression that will be needed later.

The probability that an electron in a state of form (1) at t=0 will be found in state  $|E'\rangle$  at  $t=\infty$  is  $|w(E')|^2$ , where<sup>13</sup>

$$w(E') = \langle E' | T^*(\infty) | e^{iuz} \psi \rangle, \qquad (6)$$

the operator  $T^*(t)$  being the solution of

$$idT^*/dt = e^{iEt}V(t)e^{-iEt}T^* = V^*T^*.$$
(7)

We limit ourselves to first-order time-dependent perturbation theory, setting

$$T^* = 1 + T_1^*, \tag{8}$$

<sup>&</sup>lt;sup>9</sup> A. Migdal, J. Phys. (U.S.S.R.) 4, 449 (1941).
<sup>10</sup> J. S. Levinger, Phys. Rev. 90, 11 (1953). Some numerical errors in this paper are corrected in J. S. Levinger, J. Phys. Radium 16, 556 (1955).
<sup>11</sup> H. M. Schwartz, Phys. Rev. 100, 135 (1955).
<sup>12</sup> F. Grard, Compt. Rend. 243, 777 (1956).
<sup>13</sup> P. A. M. Dirac, *The Principles of Quantum Mechanics* (Oxford University Parces London 1059).

University Press, London, 1958), 4th ed., p. 172.

where, from (7) and the fact that  $T_1^*(0) = 0$ ,

$$T_1^*(\infty) = -i \int_0^\infty V^*(t) dt.$$
(9)

With Eqs. (8), (2), and the further approximation

$$e^{iuz}\psi\rangle = (1+iuz)\psi\rangle \tag{10}$$

(which is valid, since u=0.14 a.u. and the amplitude of  $\psi$  is negligible at distances a few times  $Z^{-1}$  atomic units), Eq. (6) becomes, for  $E' \neq E^0$ ,

$$w(E') = \langle E' | (1+T_1^*(\infty))(1+iuz) | (|E^0\rangle + |1\rangle) = \langle E' | 1\rangle + \langle E' | T_1^*(\infty) | E^0\rangle + iu\langle E' | z | E^0\rangle + O(Z^{-2}) \text{ terms.}$$
(11)

The second term on the right is, from Eq. (9),

$$\begin{split} \langle E' | T_1^*(\infty) | E^0 \rangle &= -i \int_0^\infty e^{i(E' - E^0)t} \langle E' | V(t) | E^0 \rangle dt \\ &= \frac{\langle E' | V(0) | E^0 \rangle}{E' - E^0} + \int_0^\infty \frac{e^{i(E' - E^0)t}}{E' - E^0} \langle E' | \dot{V}(t) | E^0 \rangle dt, \end{split}$$

so that in view of Eq. (5), Eq. (11) reduces to

$$w(E') = iu\langle E' | z | E^0 \rangle + \int_0^\infty \frac{e^{i(E'-E^0)t}}{E'-E^0} \langle E' | \dot{V}(t) | E^0 \rangle dt + O(Z^{-2}) \text{ terms.} \quad (12)$$

If, following Migdal, we expand the integral in Eq. (12) by successive partial integrations, the result can be put into the form

$$w(E') = iu\langle E' | z | E^0 \rangle$$
  
+  $i \frac{2v}{(E' - E^0)^2} \langle E' | (-z/r^3) | E^0 \rangle + R, \quad (13)$ 

with

$$R = -2 \sum_{k=2}^{N} i^{k} k! \frac{v^{k}}{(E' - E^{0})^{k+1}} \left\langle E' \left| \frac{P_{k}(z/r)}{r^{k+1}} \right| E^{0} \right\rangle$$

+remainder, (14)

where r is the radial variable and  $P_k(z/r)$  is the normalized kth Legendre polynomial.

The form of Eq. (14) shows, as particularly emphasized by Levinger,<sup>10</sup> that the last two terms of Eq. (13) constitute an asymptotic, rather than a convergent, expansion of the integral in Eq. (12). But reasonable results are frequently obtained by use of only the first term in asymptotic expansions, so, with Migdal, we discard the R term in Eq. (13) (and in so doing, probably incur a very large error; see the account of Levinger's and Schaefer's calculations in Sec. VI).

The matrix element of  $-z/r^3$  in Eq. (13) can be neatly transformed into one of z by the observation<sup>9</sup> that in view of the classical equation of motion  $\ddot{z}$   $=-Zz/r^3$  we have in quantum mechanics

$$-\frac{z}{r^3} = \frac{\ddot{z}}{Z} = \frac{1}{Z} [\dot{z}, E] = \frac{1}{Z} [[z, E], E],$$

where i[a,b]=ab-ba. On substituting this into Eq. (13) and neglecting R, we obtain as the probability that the electron will be found in state  $|E'\rangle$  at  $t=\infty$ ,

$$|w(E')|^{2} = (u - 2v/Z)^{2} |\langle E' | z | E^{0} \rangle|^{2}.$$
(15)

The total probability of electron ejection in the alpha decay act, i.e., the probability that after decay the electron is not in the state  $|E^0\rangle$  of the Pb atom that corresponds to the state  $\psi\rangle$  of the Po atom, is the sum of  $|w(E')|^2$  over all permitted final states  $|E'\rangle$ . This sum is readily evaluated by standard methods from available tabulations of the squares of hydrogen dipole-moment matrix elements.<sup>14</sup> We write it in the form

$$P_{k}(n,l) = \frac{(u-2v/Z)^{2}}{Z^{2}}C_{k}(n,l), \qquad (16)$$

where  $P_k(n,l)$  is the probability, summed over the 2(2l+1) electrons in a given (n,l) configuration, that an electron in this configuration will be ejected;  $C_k(n,l)$  is the appropriate sum from the table of squares of hydrogen dipole-moment matrix elements; and the factor  $Z^2$  in the denominator corrects from squared hydrogen dipole-moments (which are tabulated in atomic units of area) to squared dipole moments of a hydrogenic atom of nuclear charge Z.

Except for a trivial numerical difference, Eq. (16) is the same as the one that Levinger<sup>10</sup> derived [his Eq. (45)] by taking as the perturbation the moving center of charge of the alpha particle and the recoiling daughter nucleus. It differs from the corresponding expression of Migdal<sup>9</sup> by the presence of the nuclear recoil velocity u. For Po<sup>210</sup> Migdal's probabilities are about 10 times Levinger's, in view of the values u=0.14, v=7.32, and  $Z=\sim65$  to  $\sim80$ .

The subscript k in Eq. (16) has been appended to distinguish among ejection probabilities computed with different assumptions as to what final states are permitted. There is some uncertainty about the latter because higher electron shells are depleted to an unknown extent in the decay act. Before decay the Po<sup>210</sup>, which is presumably in the form of PoO<sub>2</sub>, has the electronic structure of Po<sup>4+</sup>, i.e., two 6s electrons and all lower shells filled. For the outermost electrons the decay act constitutes a sudden, rather than adiabatic, perturbation, and consequently there is a high probability that they will be ejected,<sup>15</sup> making their

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<sup>&</sup>lt;sup>14</sup> H. A. Bethe and E. E. Salpeter, Quantum Mechanics of Oneand Two-Electron Atoms (Academic Press Inc., New York, 1957), Eq. (60.7), p. 253, and Table 13, p. 264. <sup>15</sup> The sudden perturbation ejection probability in  $\alpha$  decay is

<sup>&</sup>lt;sup>16</sup> The sudden perturbation ejection probability in  $\alpha$  decay is theoretically four times that in  $\beta$  decay (reference 9), and in the  $\beta$  decay of high Z atoms the measured probability that at least one electron is ejected is ~0.2 according to A. H. Snell and F. Pleasonton, Phys. Rev. 111, 1338 (1958).

states available to electrons from the deeper shells. There is probably a quite significant ejection of electrons from the O and N shells, as we inferred (Sec. II) from a consideration of the relative intensities of some of the M x-ray lines, and as we might suspect by extrapolating from the measured probabilities of K, L, and M ejections, the respective values of which are, roughly, 10<sup>-6</sup>, 10<sup>-3</sup>, and 10<sup>-2</sup>.

In view of this uncertainty concerning the permitted final states, we have computed the  $C_k(n,l)$ 's of Eq. (16) for two different, rather extreme, cases: k=5designates the case in which the P shell is assumed to be stripped, while the O and N shells remain intact, i.e., all and only those states with n > 5 are available; k = 4designates the case in which the O shell, as well as the P shell, is completely stripped, so that all and only those states with n > 4 are available. In addition, we have computed the case  $k = \infty$ , i.e., ejection to the continuum only, as a check of Migdal's arithmetic and for the purposes of comparison. The  $C_k(n,l)$  values for these different cases<sup>16</sup> are entered in Table III.

$C_{\infty}(n,l)^{\mathrm{a}}$	n	1	2	3
(only continuum	0	0.575	1.8	2.95
states available)	1	• • •	3.3	7.83
	2	•••	•••	6.48
·	$\backslash n$			
$C_5(n,l)$	<i>l</i>	1	2	3
(states of $n > 5$	0	0.625	2.4	6.08
available)	1	• • •	4.98	18.15
	2	•••	•••	21.33
	\n			
$C_4(n,l)$	Ì	1	2	3
(states of $n > 4$	0	0.65	2.8	9.48
available)	1	•••	6.28	30.5
	2	•••	•••	43.65

TABLE III. Theoretical  $C_k(n,l)$  values.

<sup>a</sup> See footnote 16.

A rough idea of how the theoretical ejection probabilities are affected by the different assumptions concerning what final states are available is provided by comparing the numbers  $C_k(n) = \sum_l C_k(n,l)$  computed from Table III. For K, L, and M ejection, respectively, the ratios  $C_{\infty}(n): C_5(n): C_4(n)$  are 0.92:1:1.04; 0.69:1: 1.23; 0.38: 1:1.83. It is seen that almost all K ejection is to the continuum, while a preponderance of Mejections is to bound states.

## V. Comparison of Theoretical and Measured Photon Yields

Since the theory estimates the probability that an electron is ejected from a given shell in the decay act, while experiment measures the photon yield, i.e., the probability that a corresponding x-ray photon is

<sup>16</sup> Except for trivial differences, the  $C_{\infty}(n,l)$ 's are one-fourth Migdal's values, since the latter contain the square of the alpha particle charge as a factor.

TABLE IV. M	Ieasured Pb	photon	yields	in Po <sup>210</sup>	decay.
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Shell	Measured yield	Authors' stated error	Source strength, mC of Po <sup>210</sup>	Source thickness, mC/cm <sup>2</sup>	Refer- ence
K	$1.5 \times 10^{-6}$ 2.00 1.6 1.5	$\pm 33\%$ 16 31 27	213 1 8.1–11.1 7.5	$213 \\ 4 \\ \sim 10(?) \\ ?$	a b c d
L	$2.2 \times 10^{-4}$ 2.93 4	$\pm 23\% \\ 15 \\ \cdots$	$\begin{array}{c} 8.1 - 11.1 \\ 0.5 - 4 \\ 27 - 38 \end{array}$	$\sim 10(?)$ 0.3-2.3 $\sim 135-190$	c e f
Μ	1.5×10 <sup>-3</sup> 0.91	 ±15%	27–38 0.044–0.44	$\sim$ 135–190 0.05–0.5	f g

<sup>a</sup> M. A. Grace, R. A. Allen, D. West, and H. Halban, Proc. Phys. Soc. (London) A64, 493 (1951),
 <sup>b</sup> W. C. Barber and R. H. Helm, Phys. Rev. 86, 275 (1952).
 <sup>c</sup> M. Riou, J. Phys. Radium 13, 487 (1952).
 <sup>d</sup> V. V. Ovechkin and E. M. Tsenter, Soviet J. At. Energy, English Transl. 2, 344 (1957).
 <sup>e</sup> W. Rubinson and W. Bernstein, Phys. Rev. 86, 545 (1952).
 <sup>f</sup> I. Curie and F. Joliot, J. Phys. Radium 2, 20 (1931).
 <sup>g</sup> W. Rubinson, this paper.

emitted, a correction for fluorescence yield is necessary. It will be convenient to correct the theoretical values.

In Table IV are listed all the measured photon vields in Po<sup>210</sup> decay known to us. For each of the shells there are at least two independent measurements made with different techniques on sources of widely different strengths. In no case is the discrepancy between the different measurements as great as a factor of 2.

The theoretical photon yields were computed from Eq. (16) for the case k=5 [i.e., with use of the  $C_5(n,l)$ coefficients from Table III, which was felt to be a more reasonable choice than the case k=4 in view of the fact that although there are real differences between a Pb M spectrum from Po<sup>210</sup> decay and a conventional spectrum (see Sec. II), the differences are relatively minor. The Z's used in the computation were effective Z's obtained by correcting the  ${}_{82}$ Pb nuclear charge by means of Hartree's<sup>17</sup> screening constants for <sub>80</sub>Hg<sup>2+</sup>. The fluorescence yields were taken to be 0.96 for the K shell,<sup>18</sup> 0.39 for the L shell,<sup>19</sup> and 0.04 for the M shell.20

Theory and measurement are compared in Table V. The orders-of-magnitude discrepancies there displayed cannot be reduced significantly by computing the theoretical values with the  $C_4(n,l)$ , instead of the

<sup>17</sup> D. R. Hartree, *The Calculation of Atomic Structures* (John Wiley & Sons, Inc., New York, 1957), p. 117. <sup>18</sup> A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (Interscience Publishers, Inc., New York,

Spectroscopy 1 acres (intersective 1 assumed, 1..., 1959), p. 81. <sup>19</sup> See reference 18, p. 85, Eq. (7.53). In applying this equation we assumed that the number of  $L_{\rm I}$  holes is given by  $C_5(2,0)$  in our Table III, and the numbers of  $L_{\rm II}$  and  $L_{\rm III}$  holes are, respec-tively, 1/3 and 2/3 of  $C_5(2,1)$ . The mean L fluorescence yield betwiesd in this way 0.30 is identical with the measured mean tively, 1/3 and 2/3 of  $C_5(2,1)$ . The mean L fluorescence yield obtained in this way, 0.39, is identical with the measured mean Pb L fluorescence yield reported by E. T. Petronus, Jr., C. H. Braden, and L. D. Wyly, Phys. Rev. 105, 681 (1957), who excited the Pb L rays by means of 22 keV x rays. <sup>20</sup> A. A. Jaffe, Phys. Abstr. 58, 360 (1955), Abstract 2782. The value, which derives from measurements on the Bi M lines in

RaD decay, is given as  $0.037 \pm 0.007$ .

Shell	$\frac{\text{Effective}}{Z}$	Mean fluo- rescence yield	Theoretical ejection probability, $P_5$	Theoretical photon yield	Measured photon yield	Measured Theory
K	81.7	0.96	1.42×10-7	1.36×10-7	2.0 ×10 <sup>-6</sup>	15
L	${78.4 (2s)}$ ${77.8 (2p)}$	0.39	$2.76  imes 10^{-6}$	1.07×10-6	2.9 ×10 <sup>-4</sup>	270
М	$ \begin{cases} 71.7 & (3s) \\ 70.1 & (3p) \\ 65.8 & (3d) \end{cases} $	0.04	5.56×10 <sup>-5</sup>	2.22×10 <sup>-6</sup>	0.91×10-3	410

TABLE V. Comparison of theoretical and measured photon yields in Po<sup>210</sup> decay.

 $C_5(n,l)$ , coefficients (see the last paragraph of Sec. IV). It will be noticed that if the  $C_4(n,l)$  coefficients are the more appropriate ones (i.e., if the O shell is very extensively denuded in the decay act), then the M fluorescence yield used above is probably much too low.

If in Eq. (16) we take u=0, i.e., if we neglect recoil, as Migdal does, then the discrepancies between theory and measurement are reduced by roughly a factor of 10.

### VI. Discussion

It is highly unlikely that any essential part of the gross discrepancy between theory and experiment displayed in Table V can be attributed to a misinterpretation of the experimental data, rather than to the theory.<sup>21</sup> For example, the absence of any detectable electromagnetic radiations other than Pb x rays and the 0.8-MeV gamma (see Sec. ICi) effectively eliminates the possibility that the decay act gives rise to a variety of gamma rays that are converted in the different shells; such conversion would have to be effectively complete, a concatenation of circumstances too intolerable to contemplate.

What is disconcerting about the gross inadequacy of the theory is that the model and the mathematical approximations are of a conventional kind which seem to have proved adequate in the much-studied analogous problem of electron ejection by bombardment with heavy particles from an external source.<sup>22</sup> The latter problem differs from ours in that the projectile comes from  $-\infty$  instead of from the origin, and impinges on the target atom with all impact parameters between zero and  $\infty$  with respect to the nucleus instead of being limited to zero impact parameter, the probability of electron ejection being obtained by averaging over all impact parameters. Perhaps the passable agreement with experiment found by this method is simply due to the fact that it weights the more distant collisions much more heavily than the close ones, where the difficulties arise.

The assumptions that enter the model for electron ejection in alpha decay, such as the point-charge approximation for the alpha and alpha emission with zero angular momentum, have been justified by the original authors.<sup>9,10</sup> We point out further that with the potential function Eq. (4), which entails that the alpha particle instantaneously assumes its full terminal velocity and the recoil nucleus is instantaneously halted (classically a Po<sup>210</sup> alpha attains 95% of its terminal velocity in traversing two-thirds the radius of the K shell), we are assuming a perturbation that is less adiabatic than it is in fact, which makes for a theoretical *overestimate* of the electron ejection probability.

As concerns the mathematical approximations in the theory, we have mentioned Levinger's observation that the last two terms on the right in Eq. (13) constitute an asymptotic, rather than a convergent, expansion. He found that retention of the k=2 term of R in the calculations has only a negligible effect on the estimate of K ejection, but increases that of L, and presumably<sup>28</sup>

<sup>23</sup> J. S. Levinger (private communication).

<sup>&</sup>lt;sup>21</sup> Levinger (reference 10) conjectured that the high measured value of the K photon yield, namely,  $\sim 2 \times 10^{-6}$ , was due to a 10% internal conversion of the 0.8-MeV Po<sup>210</sup>  $\gamma$ , the intensity of the latter being  $1.22(\pm0.06) \times 10^{-5}$  [R. W. Hayward, D. D. Hoppes, and W. B. Mann, J. Res. Natl. Bur. Std. 54, 47 (1955)]. However, it now appears to be definitely established that the internal conversion of these gammas is only  $\sim 1\%$ , so that this process can account for only about a tenth of the observed K x rays. For, the 0.8 MeV Po<sup>210</sup> gamma is E2 [S. De Benedetti and G. H. Minton, Phys. Rev. 85, 944 (1952)], and therefore, has a K conversion coefficient of 0.9% [M. E. Rose, G. H. Goertzel, B. I. Spinrad, J. Harr, and P. Strong, Phys. Rev. 83, 79 (1951)]. In agreement with this, it is found experimentally [V. V. Ovechkin and E. M. Tsenter, Soviet J. At. Energy, English Transl. 2, 344 (1957)] that the internal conversion K rays constitute  $10(\pm 3\%)$ of the total K ray intensity from Po<sup>210</sup>.

<sup>&</sup>lt;sup>22</sup> The first wave-mechanical treatment of the latter problem on this model was that of J. A. Gaunt, Proc. Cambridge Phil. Soc. **23**, 732 (1927). The equivalence of Gaunt's method and a Born approximation method was shown by N. F. Mott, Proc. Cambridge Phil. Soc. **27**, 553 (1930–31). The whole subject is reviewed by E. Merzbacher and H. W. Lewis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 34, p.

<sup>166,</sup> who point out that the apparent good agreement between measured cross sections and those computed in Born approximation with nonrelativistic wave functions may be accidental in view of the fact that when relativistic wave functions are used, the theory is high by more than a factor of 2. J. Bang and J. M. Hansteen [Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **31**, No. 13 (1959)] have reworked the theory of K ejection under heavy particle bombardment with Gaunt's model modified to include the repulsion between the projectile and target nuclei. This modification causes a considerable reduction in the cross sections. Bang and Hansteen's comparison of their theoretical results with more recent measured cross sections for the ejection of K electrons by relatively slow protons shows agreement within roughly a factor of 2. The agreement between different recent measurements is no better than about a factor of 2. It appears that this old, basic problem has hardly been settled definitively. However, there do not seem to arise in it the order-of-magnitude discrepancies that we find in the case of alpha decay.

M, ejection by an order of magnitude. Note (Table V) that this reduces the deficiency in the theoretical estimates of L and M ejections to the same order as Kejections.

If recourse to the asymptotic expansion is avoided altogether, there is apparently a very considerable improvement in the theory as shown in some very recent calculations of the K ejection probability by Schaefer.<sup>24</sup> Working with essentially the model described in Sec. IV, and evaluating the relevant matrix elements exactly, he finds a K ejection probability about half the experimental value. He finds further that with a standard kind of correction for external screening, the theory is brought to give about 80%of the experimental value. However, when he uses a more realistic model, in which the assumption of constant alpha velocity is replaced by that of a classically accelerated motion of the alpha particle and the recoil nucleus, the theoretical value for K ejection falls again to about half the measured value. Schaefer hopes that the use of relativistic state functions may restore agreement with experiment. In any case his work seems to have brought the discrepancy between theory and experiment within a range acceptable for calculations made with hydrogenic wave functions.

A few comments on the use of hydrogenic wave functions in problems involving high Z atoms. Their relative success in treating a variety of electron ejection phenomena presents something of a theoretical problem, since theoretically hydrogenic continuum wave functions normalized in the usual way should be adequate only in cases where the continuum energy is very much greater than the binding energy of the electron in its initial state.<sup>25</sup> This condition is certainly not met in the present theory; the most probable energy of an electron ejected to the continuum is zero, and with increasing energy the probability drops precipitously to a limiting energy dependence of the form  $energy^{-9/2}$ , with a great preponderance of the ejected electrons having energies less than the binding energy of the initial state.9

In the analogous problem of electron ejection in beta decay, where the perturbation is sudden, instead of adiabatic, and nuclear recoil is unimportant, the theoretical ejection probabilities are roughly correct, being generally about one-half the measured values.<sup>26</sup> However, in practice these ejection probabilities are computed by way of the completeness property, i.e., by subtracting from 1 the probability that no transition occurs together with the sum of the probabilities of otherwise allowed transitions that are forbidden by the Pauli principle. In this way of calculation, which avoids the problem of the normalization of the continuum states altogether, the use of hydrogenic functions can be expected to result in reasonably good rough answers.<sup>27</sup> (In an attempt to apply this method to the case of electron ejection in alpha decay we obtained an absurd result.) When the calculations cannot be made by appeal to the completeness property, a theory that uses hydrogenic wave functions seems inadequate to account for the experimental results in beta decay. Charpak and Suzor<sup>28</sup> have used proportional counters in coincidence to study both the x-ray intensities and the energy distributions of ejected electrons in the beta decays of P<sup>32</sup> and S<sup>35</sup>. The observed x-ray intensities agree roughly with the theoretical estimates made by use of the completeness property; the observed intensities in the energy distributions are grossly in excess of the theoretical estimates, which, of course, are computed without use of the completeness property.

<sup>27</sup> How good is indicated by the following comparison for the case of He<sup>6</sup>  $\beta$  decay, where states adjacent to the ground state are not forbidden by the Pauli principle, so that no significant changes in screening constants are involved. A. Winther [Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 27, No. 2 (1952)] has used Hylleras' very accurate wave functions for He (7 parameter) and Li (10 parameter) to compute the probability that in the  $\beta$  decay of He<sup>6</sup> the two He<sup>6</sup> electrons remain as ground-state electrons of the Li<sup>+</sup> daughter. He finds this probability to be 0.670, so that the probability of exciting at least one electron is 0.33. We have calculated the same case with hydrogenic 1s functions, where the problem involves only one parameter, namely, the effective charge Z of the He nucleus. The probability of The interference of the two 1s electrons is  $2(1-p^2)$ , where, as shown by a simple integration,  $p = \{[(Z+0.5)^2-0.25]/(Z+0.5)^2\}^{3/2}$ . With Z taken to have its variational value 27/16, we find  $2(1-p^2)$ =0.297, i.e., 90% of the value obtained in Winther's very elaborate calculation.

<sup>28</sup> G. Charpak and F. Suzor, J. Phys. Radium 20, 25, 31 (1959).

<sup>&</sup>lt;sup>24</sup> G. W. Schaefer (private communication).
<sup>25</sup> H. S. W. Massey, Appendix to a paper by E. H. S. Burhop, Proc. Cambridge Phil. Soc. 36, 43 (1940).

<sup>&</sup>lt;sup>28</sup> For the theory see Migdal (reference 9); Levinger (reference 10); E. L. Feinberg, J. Phys. (U.S.S.R.) 4, 424 (1941); and H. M. Schwartz, J. Chem. Phys. 21, 45 (1953). There exist about M. In Schwartz, J. Chem. First. 21, 43 (1933). There exist about ten measurements, most of them with errors of  $\sim 50\%$ . In the case of the  $\beta$  decay of RaE, the measured K, L, and M ejection probabilities are, respectively, 1.2, 2.2, and 1.7 times the theo-retical values computed by us from Schwartz's tables. [K and L ejection probabilities measured by F. Boehm and C. S. Wu, Phys. Dec 02, 519 (1054) and L and M eigeting meabelificity encoder Rev. 93, 518 (1954); and L and M ejection probabilities measured by G. A. Renard. J. Phys. Radium 16, 576 (1955).] The fact that the measured values are higher than the theoretical ones is consistent with an observation by Winther (reference 27) that computed ejection probabilities increase with the accuracy of the wave functions used to compute them.