Internal Bremsstrahlung and Ionization Accompanying Beta Decay*

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Internal bremsstrahlung spectra from S^{35} and Pm^{147} have been investigated with the NaI scintillation spectrometer. Both the absolute cross section of production and the energy distribution agree with theoretical predictions. The theoretical internal bremsstrahlung spectrum of RaE was calculated by using both the observed forbidden β spectrum and an allowed β distribution. No significant differences in shape were found in these two cases except in the absolute cross section of production.

The ionization accompanying beta decay was studied in the case of RaE and also in Pm^{147} by measuring the characteristic x-rays in an NaI scintillation counter as well as in a proportional counter spectrometer. Both K and L radiations were observed, and their absolute cross sections were compared with the theoretical calculations of Migdal, Feinberg, and Levinger. Because of the approximate nature of the theoretical calculations, the agreement between the theoretical and experimental results is considered satisfactory.

I. INTRODUCTION

THE process of β decay is accompanied by various secondary effects which have been subjects of investigations for the last two decades. One of these secondary effects is the interaction of the nuclear beta particle with the radiation field of the nucleus causing the emission of internal bremsstrahlung (continuous x-rays). Another secondary effect is the perturbation of the atomic electrons resulting from either the sudden change of nuclear charge or simply the direct collision between the beta particle and the inner electrons. This perturbation induces electronic excitation to an unoccupied discrete level or ionization to the continuum. The hole in the inner electron shell thus created will be immediately filled up by an electron from the outer shell or from the continuum. Characteristic x-rays or Auger electrons will be emitted as a result. Additional secondary effects which are expected as the result of beta decay are the annihilation of the electron-positron on the atomic core and pair creation during β decay. However, we shall concern ourselves here only with the the production of inner bremsstrahlung and the ionization of the atom.

II. INTERNAL BREMSSTRAHLUNG

1. Introduction

The probability of emission of the internal bremsstrahlung and the shape of the bremsstrahlung spectrum have been investigated both experimentally and theoretically in various laboratories. The more recent experimental investigations are by Wu;[1] by Stahel and Guillessen[2] with the ionization chamber method; by Novey;[3] and by Bolgiano, Madansky, and Rasetti[4] with a NaI

scintillation spectrometer. The β-radioactive substances used in these investigations were P^{32}, Y^{91}, and RaE^{210}. Investigations on electron-capture processes were made by Maeder and Preiswerk,[5] by Bell et al.,[6] and also by Anderson and his co-workers.[7] Calculations on the internal bremsstrahlung spectrum were first made by Knipp and Uhlenbeck[8] and by Bloch[9] for allowed β transitions with a polar vector interaction. Chang and Falkoff[10] and Madansky[11] extended these calculations to first and second forbidden β transitions and different types of interactions. (Allowed: scalar or tensor, first forbidden; scalar or tensor ($\Delta I = 2$, yes), second forbidden; scalar.) Their results indicate that both the ratio of γ intensity to β intensity and the shape of the bremsstrahlung spectrum are almost the same for forbidden transitions as for allowed transitions, irrespective of the types of interactions. The production of internal bremsstrahlung due to the electron-capture process was first theoretically investigated by Morrison and Schiff.[12]

Furthermore, in evaluating the production of internal bremsstrahlung, one usually treats the over-all process in two steps:

$$N_0 \xrightarrow{(1)} P+\nu+e' \xrightarrow{(2)} P+\nu+e+\gamma(k),$$

where (1) represents the transition from initial to intermediate state with the transformation of the nucleon and the creation of a pair of light particles and (2) represents the transition of the electron from the intermediate state to the final state with the simultaneous emission of a light quantum of energy k.

The joint probability of the over-all process of emission of a quantum of energy k can be calculated with the

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[1] C. S. Wu, Phys. Rev. 59, 481 (1941); references to earlier papers given here.

[2] E. Stahel and J. Guillessen, J. phys. radium 1, 12 (1940).

[3] T. B. Novey, Phys. Rev. 89, 672 (1953).

[4] Bolgiano, Madansky, and Rasetti, Phys. Rev. 89, 679 (1953); L. Madansky and F. Rasetti, Phys. Rev. 83, 187 (1951).

[5] D. Maeder and P. Preiswerk, Phys. Rev. 84, 595 (1951).

[6] Bell, Tauch, and Cassidy, Science 115, 12 (1952).

[7] Anderson, Wheeler, and Watson, Phys. Rev. 87, 668 (1952).

[8] J. K. Knipp and G. E. Uhlenbeck, Physica 3, 425 (1946).

[9] F. Bloch, Phys. Rev. 50, 272 (1936).

[10] C. S. Wang Chang and D. L. Falkoff, Phys. Rev. 76, 365 (1949).

[11] Madansky, Lipps, Bolgiano, and Berlin, Phys. Rev. 84, 596 (1951).

[12] P. Morrison and L. Schiff, Phys. Rev. 85, 24 (1940).

rigorous second-order perturbation method. However, Knipp and Uhlenbeck introduced an alternate method in which the joint probability for the over-all process was simply the product of the probabilities of all the steps. This simplification could be justified if the two processes were independent of each other. Although there is no basis for assuming these processes as independent, the result obtained with this simple approach for the allowed β transition turns out to be exactly the same as that from the more rigorous second-order perturbation method. Therefore, the acceptance of its use for the allowed transition invoked no arguments.

However, the question was then raised as to the validity of the application of this simple method to forbidden transitions. It was pointed out by Morrison and Schiff[12] that the close agreement between the calculations by second-order and by first-order perturbation methods for allowed transitions results from the fact that the electron-neutrino coupling does not depend explicitly on the momenta of these particles. This is certainly not true in forbidden cases. Chang and Falkoff[10] carried out calculations with both the second-order perturbation method and the simple method for first and second forbidden transitions of scalar interactions. They found that, although the results from these two methods are not exactly the same, the close agreement between them justifies the adoption of the simple method. Under the assumption of independence of these two processes, the probability for the emission of a γ quantum with energy k can be expressed[8,9] as

$$S(k) = \int dW_e P(W_e) \Phi(W_e, k),$$

where $P(W_e)dW_e$ is the distribution of the β spectrum and $\Phi(W_e, k)$ is the probability of an electron coming from the nucleus with energy W_e. The function $\Phi(W_e, k)$ was calculated to be

$$\Phi(W_e, k) = \frac{\alpha p_s}{\pi p_e k} \left\{ \frac{W_e^2 + W_s^2}{W_e p_s} \ln(W_s + p_s) - 2 \right\},$$

where "W_e and W_s" and "p_e and p_s" are the energies and momenta of the electron before and after the emission of the photon k. It can be seen that one obvious advantage of using the simple expression is that one may use the experimentally measured β spectrum for $P(W_e)dW_e$, thereby avoiding the difficulty of dealing with the lack of uniqueness in the type of interaction in β decay. This is particularly so in the case of RaE. The β spectrum of RaE has an unique forbidden shape with a great excess of low-energy electrons. The only reasonable theoretical fitting to the observed spectrum was attempted by Petschek and Marshak[13] by using a mixture of interactions (tensor

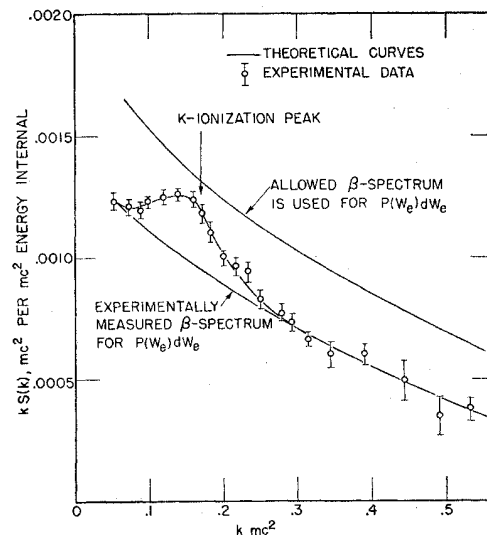[13] A. G. Petschek and R. E. Marshak, Phys. Rev. 85, 698 (1952); C. S. Wu, Physica 18, 989 (1952).

FIG. 1. Internal bremsstrahlung and K-ionization radiation from RaE (measured on the NaI scintillation spectrometer).

and pseudoscalar). However, no calculations using this particular mixture of interactions have been made for the internal bremsstrahlung. By applying the simple method, one could use the observed β spectrum of RaE. Nevertheless, it is interesting to examine how sensitive the shape of the bremsstrahlung is to changes in the distribution of the β-particle energy in the case of RaE by calculating its spectrum, assuming first the observed β spectrum, then an allowed spectrum. The result of this comparison is shown in Fig. 1 on an absolute scale. Both curves 1 and 2 are normalized per β disintegration. It is interesting to note that the difference in shape of these two curves is rather insignificant. However, the absolute probabilities in the low-energy region of these two calculations do differ from one another by a factor of 1.3 with the higher cross section for the curve with allowed β distribution. This is quite understandable since the measured β spectrum has an excess of electrons with energy less than 0.2 Mev, which contribute less in the production of internal bremsstrahlung.

The advantage of a NaI scintillation spectrometer over the ionization chamber method in the investigation of the internal bremsstrahlung is not merely its high detecting efficiency but also the fact that it yields information on its energy distribution. Wu[1] and Stahel and Guillessen[2] found good agreement between their measured total yields of bremsstrahlung and those predicted by the theory by the ionization method. Novey[3] and Bolgiano et al.[4] further confirmed the good agreement with the theory of the shape of the bremsstrahlung spectrum at the low-energy region using a NaI spectrometer. Unfortunately, the analysis of a continuous γ spectrum in a crystal can be carried out only as long as the photoeffect dominates over all other interactions in the crystal. This is approximately
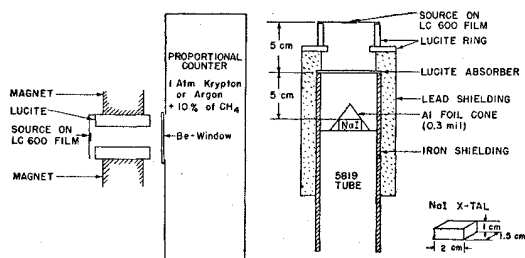
FIG. 2. Experimental arrangements. Right: scintillation counter.
Left: proportional counter.

true for γ energies lower than 200 kev. For higher energies, the Compton effect becomes comparable to the photoeffect and an accurate evaluation of the photospectrum is almost impossible. With this in mind, it seemed interesting to investigate a case of internal bremsstrahlung which has the entire spectrum below 200 kev. Low-energy beta emitters Pm^{147} and S^{35} were chosen because they both yield simple beta spectra with upper energy limits of 227 and 167 kev, respectively and emit no nuclear gamma radiations.

2. Experimental Arrangement

Figure 2 shows the experimental arrangement of the scintillation counter apparatus. The NaI crystal is in optical contact with the photocathode of a 5819 photomultiplier. The crystal is covered with a 200-$\mu g/cm^2$ Al foil. The pulses are amplified by a linear amplifier, and the pulse distributions is analyzed with a single-channel differential pulse-height analyzer. The β particles are absorbed in a Lucite absorber whose thickness is adjusted according to the maximum energy of the β spectrum. The absorber is always placed halfway between the crystal and the source. The amount of external bremsstrahlung which could reach the detector under this geometrical arrangement is not more than 10 percent compared with that of the internal bremsstrahlung. In order to avoid the production of external bremsstrahlung and the excitation of characteristic lead radiation, no lead collimator was used. The sources used were prepared by evaporation of an aqueous solution of the radioactive chemicals on an LC 600 film of a thickness of about $50\mu g/cm^2$. The thickness of the source itself was of the order of 50 $\mu g/cm^2$, and the intensity of the source was of the order of 0.1 to 1 mC.

3. Calibration and Correction

The energy calibration of the scintillation spectrometer was carried out with gamma or x-rays of Cd^{109}, Cs^{137}, and Tl^{204}. With the dimensions of the crystal used (2.0 cm$\times$1.5 cm$\times$1.0 cm), its detecting efficiency is practically 100 percent throughout [in the energy region investigated (below 200 kev)]. The absorption of γ rays in the Lucite absorbers and the thin Al reflector is corrected with known coefficients of absorption.

Another correction for the shifting of pulses to lower energy regions due to the escape of iodine K x-rays from the crystal following photoelectric interaction near the surface was estimated [8] and applied. A small resolution correction similar to that used by Palmer and Laslett[14] (which takes into account the energy dependence of the half-width of the Gaussian distribution of a monochromatic photoline) was applied to the measured spectrum. However, the channel width of the analyzer was always set several times smaller than the line width of the Gaussian distribution.

In order to compare the absolute cross section for bremsstrahlung production with the theoretical prediction, the absolute intensity of the β source and the efficiency factor of the detector system must be known. The first quantity was measured in a large cylindrical vacuum chamber (the Columbia solenoid spectrometer) using a G-M counter with a thin Formvar window of a thickness of 0.03 mg/cm^2. A suitable antiscattering baffle-system was put in to exclude electrons scattered from the wall. Several measurements were made with varying distances between the source and the detector to insure square distance relation. The efficiency factor of the NaI detector or the proportional counter (including the solid angle subtended) was estimated with the help of a Cd^{109} source which was calibrated in terms of an absolute number of disintegrations per second by measuring the conversion electrons in the mentioned vacuum chamber and G-M counter system and assuming 0.8 K-conversion electron and 1.5 K quanta per disintegration.

4. Results and Conclusion

After all the corrections were applied, the spectrum of the internal bremsstrahlung was plotted with $kS(k)$ vs energy to avoid an abrupt rise at the low-energy end. Figures 1, 3, and 4 represent the experimental results of RaE, S^{35} and Pm^{147}. In calculating $S(k)$, the allowed β spectrum was used for S^{35} and Pm^{147}, but the observed β spectrum was substituted for RaE. Because of the uncertainties involved in the calibration of the β source
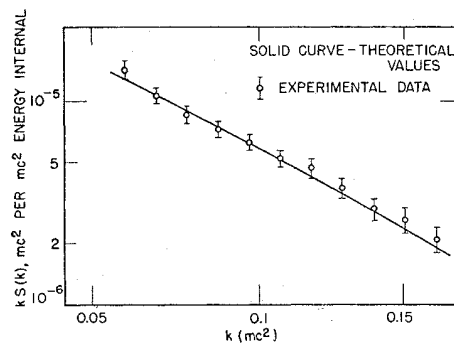

FIG. 3. Internal bremsstrahlung of S^{35}.

[14] J. P. Palmer and L. J. Laslett, U. S. Atomic Energy Commission Report AECU-1220, (March 14, 1951) (unpublished).

and in the detector efficiency, the emphasis is put on comparing the energy distribution of the internal bremsstrahlung rather than the absolute yields. The absolute yield may have an error of the order of 20 percent. Nevertheless, both of these factors agree rather satisfactorily with the theoretical predictions. Recently, Renard[15] investigated the internal bremsstrahlung from P^{32} in an energy region of 3–30 kev in a proportional counter and also obtained good accord with the theory. The excellent agreement between the theory and the experimental findings may imply that the simplification (first-order perturbation) and approximation (Z independent) introduced in the theoretical calculations have very little effect on the distribution or yield of the bremsstrahlung.

III. EXCITATION AND IONIZATION OF THE ATOM

1. Introduction

The excitation and ionization processes of the atomic core often accompany the β decay of a nucleus. As the nuclear charge suddenly changes from Z to $Z\pm1$ and the β particle passes through the atom in a β-decay process, the atomic core may suffer two kinds of perturbations. One is the direct collision with the nuclear β particle. This is analogous to the excitation and ionization of an atom by an external fast electron. The other is a "shaking" effect caused by the sudden change of nuclear charge. Because the time taken for the fast β particle to pass through the atom is much shorter than the period of revolution of the orbital electrons, the atomic core has very little time to make adiabatic readjustments (shrinkage for β^- decay and expansion for β^+ decay) but mostly undergoes nonadiabatic transition, that is, excitation or ionization.

The probability of the ionization of the atom due to β decay was first investigated theoretically by Feinberg[16] and Migdal.[17] Their estimate showed the dominating role played by the shaking process and the insignificant contribution from the direct collision. Therefore, on the basis of the sudden change of nuclear charge alone, they calculated the probability of ionization of K, L, and M electrons in β decay. The K ionization is given as $(2\times0.32)/Z^2$, whereas the L ionization is given as $6.8/Z^2$. Later, Levinger[18] independently studied this problem and reached general agreement with Feinberg and Migdal on the probability of ionization of K-shell electrons $(0.65/Z^2)$ but differed on the results for L-shell electrons $(2.1/Z^2)$. Recently, Schwartz[19] took up the calculation of the probability of ionization resulting from β decay and his L-shell ionization yield is in agreement with that of Levinger. Winther[20] also carried out a detailed calculation of the

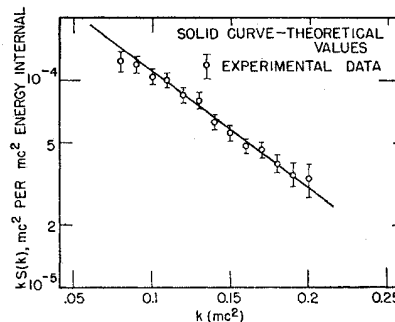FIG. 4. Internal bremsstrahlung of Pm^{147}.

ionization effect for the case of $He^6\rightarrow Li^6$ because of its particular interest for the β-recoil studies. He also pointed out in his paper the importance of the Auger effect in the ionization process which was not taken into account in Feinberg or Migdal's calculation. Serber and Snyder[21] calculated the average excitation energy of the atom for β decay. Their expression for an average excitation energy per closed electronic shell compared favorably for the K shell with Levinger's evaluation.

In Feinberg and Migdal's original calculations of the ionization probability, three approximations were used: (1) no screening corrections for Coulomb wave functions, (2) nonrelativistic treatment, and (3) sudden change of nuclear charge. Levinger first calculated the correction resulting from the screening effect by applying Hartree wave functions for Hg and found that for heavy atoms the screening effects cause an appreciable increase in the ionization probability following β decay. For K electrons, the factor is 1.4 while it is 3 or 4 for L electrons. He also estimated the errors introduced by the other two approximations and found that the use of the nonrelativistic wave functions lead to an underestimation of the ionization probability by a factor of 2. Also, the approximation of sudden perturbtion calculation should not be very accurate for a heavy atom with low energy β decay.

To detect the ionization process caused by β decay, one could either

(1) detect the orbital electrons ejected during electron-capture or the β^+ decay process,

(2) observe the characteristic x-rays emitted in filling the holes in the K or L shells, or

(3) measure the Auger electrons emitted in competition with the x-ray emission.

Bruner,[22] with the aid of a magnetic spectrometer, has applied the first method to Sc^{44}, a radioactive isotope which decays by β^+ emission and electron capture process as well as by emission of nuclear gamma rays. However, the calculated yield of ionization based on the number of electrons detected is much too high to be reconciled with the theory. Recently, Porter and

[15] G. Renard, J. phys. radium 14, 361 (1953).
[16] E. L. Feinberg, J. Phys. (U.S.S.R.) 4, 423 (1941).
[17] A. Migdal, J. Phys. (U.S.S.R.) 4, 449 (1941).
[18] J. S. Levinger, Phys. Rev. 90, 11 (1953).
[19] H. M. Schwartz, J. Chem. Phys. 21, 45 (1953).
[20] A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 27, 2 (1952).

[21] R. Serber and H. S. Snyder, Phys. Rev. 87, 152 (1952).
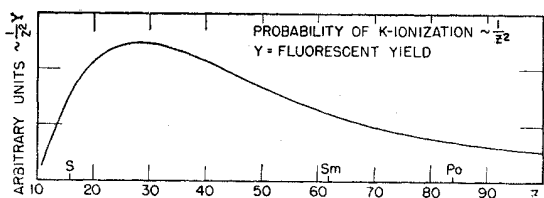[22] J. A. Bruner, Phys. Rev. 84, 282 (1951).

Fig. 5. Variation of fluorescence yield/Z^2 vs Z.

Hotz[23] studied the electrons ejected from Fe^{55} (a pure K-capture radioisotope) in a cloud chamber and found good agreement with the theoretical calculations by Primakoff and Porter.[24]

The method employed in the investigation of the ionization of the atom due to β^- decay as reported in this paper is based on the detection of the characteristic x-rays by a proportional counter or scintillation counter spectrometer. In order to rule out the characteristic x-rays from other effects, a radioactive substance with no nuclear gamma radiation or electron capture is preferred. The substances used in our investigation are RaE and Pm^{147}, whose simple decay schemes are well established. The β decay of Pm^{147} is particularly suited for this investigation. Since the cross section for ionization varies with $1/Z^2$, one would therefore at first suggest the use of β^- radioactive light nuclei. However, the fluorescence yield of the atom decreases rapidly with decreasing Z. One therefore must make a compromise between these two effects. Furthermore, the energy of the characteristic x-radiations of a light atom is low and would require the detection of a weak line on a high background of low-energy internal bremsstrahlung. Figure 5 gives the curve showing the variation of the product of ionization probability times fluorescence yield for K-shell electrons *versus* atomic number Z.

In the curves showing the continuous internal bremsstrahlung of RaE by Novey[3] and Madansky and Rasetti,[4] an unmistakable peaking could be interpreted as an evidence of the ionization of the atom by β decay. Nevertheless, a similar peak of the same energy is shown also on the bremsstrahlung curve of P^{32}. This suggests that at least a great portion of that peak is likely to be the result of the fluorescence radiation of lead excited by external electrons on the lead shielding and not the result of ionization caused by the sudden change of nuclear charge.

2. Experimental Arrangement

A proportional counter and a scintillation counter spectrometer were used for the measurement of the characteristic radiations. Figure 1 shows the experimental arrangement of the counter setup. The proportional counter is filled with a mixture of one atmosphere of Kr and 7 cm of methane. The counter window consists of a 10-mil Be foil. The scintillation counter is

the same one used for the internal bremsstrahlung. The electrons are bent away with a strong permanent magnet. All metal parts are carefully shielded with lucite in order to avoid the production of any external characteristic radiation. The Pm^{147} was a carrier-free fission product from Oak Ridge National Laboratory. The extremely thin source was deposited on a $50\mu g/cm^2$ LC 600 film. The thickness of a source must be carefully considered as any small amount of carrier would produce a fluorescent radiation which may be mistaken as due to the ionization effect under investigation. The amount of characteristic radiation produced in the source by electron bombardment could be estimated[25] from the total ionization by electrons of an energy equivalent to the mean energy of the β spectrum. It is found that the carrier free 0.1-mC Pm^{147} source of an amount ~ 10 $\mu g/cm^2$ would produce $\sim 6 \times 10^{-7}$ K ionization and 5×10^{-5} L ionization per beta decay. As we shall see, the K ionization is several orders of magnitude smaller than the actually measured ionization probability.

The RaE was first separated from polonium and was then separated from RaD be the dithiazone method. The purity of RaE can easily be assured by checking the absence of the 46.5-kev gamma line of RaD and also by following the half-life. If the separation of RaE from RaD were not complete, a faint L x-ray from such a RaE source due to the remaining RaD could easily be mistaken as the L radiation of RaE due to the ionization effect. The RaE separation was actually carried out with only $2\mu g$ of Bi carrier. Nevertheless, the source prepared is quite visible, probably because of impurities from the reagents used. The source thickness is no more than $100\mu g/cm^2$. The fluorescence yield estimated[25] from this thickness of the source is 1.5×10^{-6} K ionization and 1.5×10^{-4} L ionization per β decay per $100\mu g/cm^2$. The K ionization again is at least two orders of magnitude smaller than the value found in the experiment.

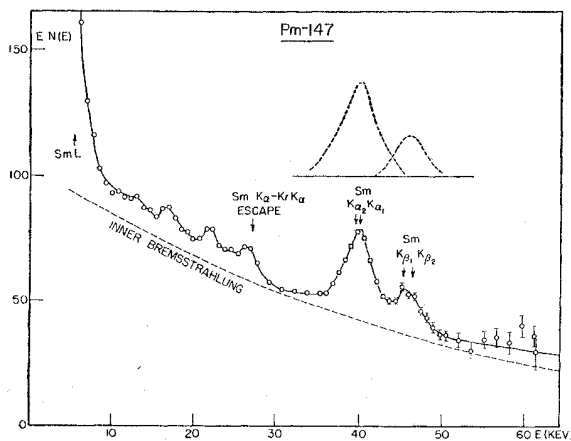The energy calibration of the proportional counter was effected with the x and gamma radiation of Cd^{109},

Fig. 6. K ionization radiations from Pm^{147}.

[23] F. T. Porter and H. P. Hotz, Phys. Rev. **89**, 938 (1953).

[24] H. Primakoff and F. T. Porter, Phys. Rev. **89**, 930 (1953).

[25] E. H. S. Burhop, Proc. Cambridge Phil. Soc. **36**, 47 (1940).

Cs[137], and RaD. The total efficiency calibration was established with a Cd[109] source of known strength.

3. Results

The experimental results for Pm[147] are shown in Figs. 6 and 7. Figure 6 exhibits the region of the K x-rays with the K_α, K_β, and escape peak distinctly shown. Two small peaks appeared between 15 and 25 kev, which may be due to some impurities. Figure 7 shows the photon spectrum in the L-energy region and a pronounced L peak of 5.6 kev is present.

The results on RaE are shown in Figs. 1 and 8. The K x-ray region of Fig. 1 was measured with the scintillation spectrometer. The K peak is pronounced, but the line profile is poor because of the broad resolution and high bremsstrahlung background. Figure 8 shows the peak due to L radiation as measured with a proportional counter. L_α and L_β are resolved.

The probabilities of the ionization processes were found by absolute measurements of the source strength and by calibrating the instrument with a calibrated Cd[109] source as described in Sec. A.

The values for the fluorescent yield used for correction of the Auger effect are taken from Burhop's new book on *The Auger Effect*.[26] The K-shell fluorescence yield for Pm[147] is 0.90 and 0.94 for RaE. The L fluorescence yield is 0.19 for Pm[147] and 0.47 for RaE.

The results are summarized in Table I. The second column lists the experimental results on the probabilities of K- and L-shell ionization as calculated from the results of our investigations. The third column gives the probabilities as calculated from the formula of Migdal or Feinberg. The numerical values for the K shells are generally
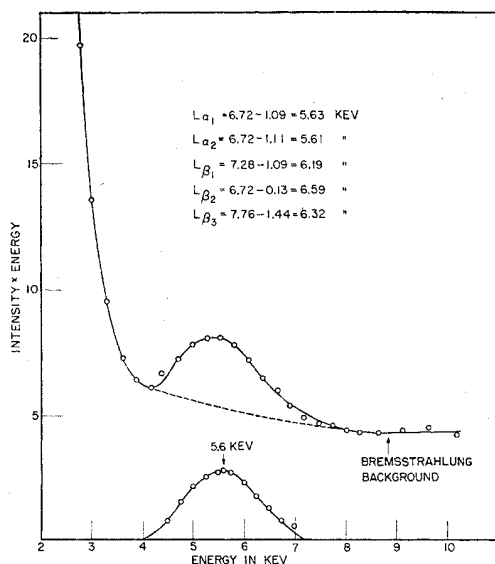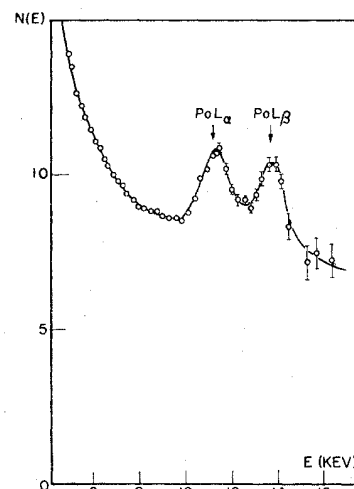

FIG. 8. L ionization radiations from RaE.

smaller than those found in experiments. Levinger applied a screening correction to the Coulomb field of the nucleus and obtained a larger probability of ionization as shown in the fourth column. However, neither relativistic correction nor direct collision was ever taken into account in the theoretical calculations. A preliminary estimate of the relativistic correction made by Levinger gave an increase of a factor of 2 for the K-shell ionization. As for the nonadiabatic approximation, the calculation may be close enough for the case of

TABLE I. Absolute probabilities for K- and L-shell ionization.

	Experimental results	Migdal or Feinberg	Levinger
Pm[147] K	$(3.85\pm0.5)\times10^{-4}$	1.64×10^{-4}	2.36×10^{-4}
L	$(2.05\pm0.3)\times10^{-3}$	1.77×10^{-3}	1.9×10^{-3}
RaE K	$(1.2\pm0.5)\times10^{-4}$	0.95×10^{-4}	1.28×10^{-4}
L	$(0.6\pm0.3)\times10^{-3}$	1.01×10^{-3}	1.04×10^{-3}

RaE, but in the case of Pm[147], the average energy of the β distribution (62 kev) is comparable to the K-binding energy of Pm[147] (45 kev). In other words, the β particles travel with a velocity comparable to that of the inner orbital electrons. Obviously, this is no longer a completely nonadiabatic case. One should therefore expect that the ionization probability would be less than that calculated from the sudden change of charge effect.

In view of the approximate nature of the theoretical calculations and the uncertainties involved in the experimental determinations, the agreement as shown in Table I is at least of the right order of magnitude.

FIG. 7. L ionization radiations from Pm[147].

[26] E. H. S. Burhop, *The Auger Effect*, Cambridge Monographs on Physics (Cambridge University Press, Cambridge, 1952).