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Internal bremsstrahlung from ^{147}Pm

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Abstract. The shape of the continuous internal bremsstrahlung accompanying the beta decay of the radioactive nuclide ^{147}Pm is experimentally measured with a NaI(Tl) scintillation spectrometer and compared with the Knipp, Uhlenbeck and Block (KUB) theory as well as with the Coulomb-corrected theories due to Lewis and Ford, and Nilsson. The experimental distribution is found to agree fairly well with the KUB theory in the energy region from 60 to 85 keV and beyond 85 keV the experimental distribution is in excess over KUB theory, the excess being 13% at 95 keV, 32% at 130 keV and 80% at 155 keV. However, we find very good agreement between the experimental distribution and the theoretical one due to Lewis and Ford throughout the investigated energy region.

1. Introduction

Internal bremsstrahlung (IB) is a very low-intensity and low-energy continuous electromagnetic radiation originating as a second-order process following the primary beta emission. The field of IB is quite interesting and has been the object of investigation for more than four decades. An exhaustive survey of the field was published recently by Persson (1968) in a review article. As can be seen from the literature, this field is rather controversial in the sense that there have been found discrepancies among various individual experimental results as well as between theory and experiment in the case of IB from many beta-decaying isotopes. For instance, if we consider IB from ^{147}Pm which was investigated several times earlier (Boehm and Wu 1954, Starfelt and Cedurlund 1957, Singh and Dargazelli 1971), even in this case the different individual experimental results do not agree with one another. Thus the experimental results of Boehm and Wu (1954) were found to agree with the theory of Knipp and Uhlenbeck (1936) and also of Block (1936) (usually referred to as the KUB theory). On the other hand the results of Starfelt and Cedurlund (1957) reveal a disagreement between theory and experiment; the experimentally measured IB yield being higher than the KUB theory as well as the Coulomb-corrected theory (Nilsson 1956), the experimental excess over theory increasing with increasing energy throughout the investigated energy range. In contrast to these latter results, the recent results of Singh and Dargazelli (1971) indicate that the experimental IB distribution is lower than the theoretical distribution throughout the investigated energy region from 68 keV to 230 keV, the discrepancy increasing with increasing photon energy. It is very interesting to note that the experimentally-measured IB distributions in the above two investigations (Starfelt and Cedurlund 1957, Singh and Dargazelli 1971) look like mirror images of each other relative to the theoretical distributions. Therefore it appears quite interesting and worthwhile to undertake the investigation of IB from ^{147}Pm again to ascertain at least the correct general trend of the experimental IB distribution relative to that of the theory. With this aim, in the present investigation, the intensity spectral shape of IB from ^{147}Pm has been experimentally measured and compared with the Coulomb-corrected (Nilsson 1956, Lewis and Ford 1957) and uncorrected theories (KUB theories).

2. Experimental details

The experiment to measure IB was rather delicate. The shielding and geometrical arrangement employed was the same as that used by Narasimhamurty and Jnanananda (1966, 1967). The IB photon distribution was experimentally measured by means of a single-channel scintillation spectrometer consisting of a Harshaw NaI(Tl) crystal of $1\frac{1}{2}$ in diameter and 1 in thickness. The ^{147}Pm source was prepared by uniformly evaporating a radioactive solution of ^{147}Pm in the required quantity over a circular area of 5 mm diameter on a thin polythene film of thickness 1 to 2 mg cm^{-2}. The prepared source had an approximate strength of the order of 100 μCi. The beta particles from the source reaching the detector were completely stopped by means of a perspex disc of thickness 50 mg cm^{-2} (corresponding to the range of ^{147}Pm beta particles), placed half way between the source and the detector which are 19 cm apart. Different runs of the experimental pulse-height distributions were taken over long periods (of the order of days) and during these periods the electronic equipment was maintained satisfactorily stable by providing sufficient cooling to it. The observed pulse-height distribution of IB resulting from different runs was found to be consistent within 2–3 %. The experimental distribution was an average of distributions resulting from three runs (while scanning the IB pulse-height distribution, the number of counts at each energy setting was collected in three different trials and the average of those three trials was taken to be the final value of the number of counts at that energy setting, this procedure was repeated throughout the energy of IB under investigation) and it is shown in figure 1 along with the background. The statistical error was made smaller than the size of the points throughout the investigated energy range by collecting a sufficient number of counts. A statistical variation

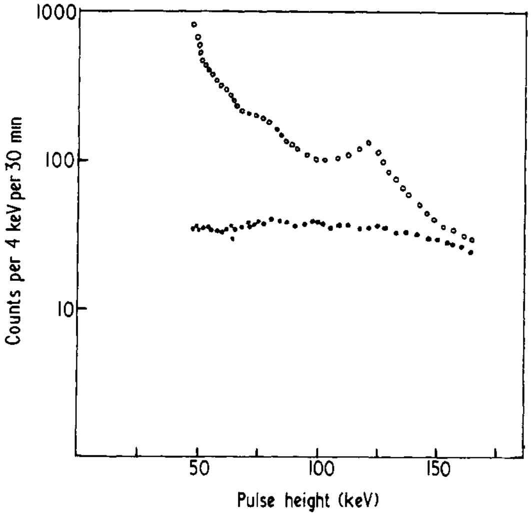

Figure 1. The experimental pulse-height distribution (○) and the background spectrum (●).

of less than 3 % even at high energy was maintained. The energy calibration was made by employing the following gamma-ray lines: 30 keV K x ray (^{133}Ba), 80 keV (^{203}Hg), 145 keV (^{141}Ce) and 280 keV (^{203}Hg). The variation of the energy calibration during the measurement was less than ±1·5 %.

3. Corrections and errors

In order to obtain the true IB photon distribution, the measured IB pulse-height distribution was corrected for factors associated with the process of the scintillation detection such as energy resolution, iodine K x ray escape, Compton electron distribution, geometrical- and gamma-detection efficiency of the crystal as was treated in Liden and Starfelt (1954) as well as for the other factors such as the background, absorption of the radiation in the beta stopper and in the aluminium can of the sodium iodide crystal, external bremsstrahlung produced within the source and the beta stopper.

As regards the calculation of the correction for the Compton electron distribution, if the fraction of photons detected with full energy is $K(E_\gamma)$ at the energy E_γ, it is obvious that the fraction of photons detected with less than the full energy is $(1-K(E_\gamma))$. If the number of photons of energy between E_γ and $E_\gamma+\mathrm{d}E_\gamma$ absorbed in the crystal is $N_\mathrm{a}(E_\gamma)\,\mathrm{d}E_\gamma$, then the number of Compton electrons, having energies ranging from 0 to E_γ^*, where E_γ^* is the maximum Compton electron energy, is $N_\mathrm{a}(E_\gamma)(1-K(E_\gamma))$. The probability that such a Compton electron will have an energy between E and $E+\Delta E$ is a function $C(E, E_\gamma)$. The total number of Compton electrons per unit energy interval at energy E due to all incoming photons from 0 to E_max is

$$N_\mathrm{C} = \int_0^{E_\mathrm{max}} C(E, E_\gamma) N_\mathrm{a}(E_\gamma)(1-K(E_\gamma))\,\mathrm{d}E_\gamma .$$

On the assumption that, in general, the Compton electron distribution for any gamma-ray energy is approximately constant over the energy range from 0 to E_γ^*, the following approximation is made:

$$C(E, E_\gamma) = \begin{cases} C(E_\gamma) = \dfrac{1}{E_\gamma^*}, & \text{for } 0 < E < E_\gamma^* \\ 0 & \text{for } E > E_\gamma^*. \end{cases}$$

In order to obtain the Compton electron distribution a step-by-step integration is performed. Actually, the observed pulse-height distribution is first extrapolated to the end point. By starting out at the highest energy, $N_\mathrm{a}(E_\gamma)$ is taken. Next the corresponding $(1-K(E_\gamma))$ is measured. Then by choosing a suitable value of ΔE_γ (the smaller the better) the ordinate $[\Delta N_\mathrm{C}]_{E'_\gamma}$ of the Compton distribution from 0 to E_γ^* due to photons of energy between E_γ and $E_\gamma+\Delta E_\gamma$, is calculated by the relation

$$[\Delta N_\mathrm{C}]_{E_\gamma} = \frac{E}{E_\gamma^*} N_\mathrm{a}(E_\gamma)(1-K(E_\gamma))\,\Delta E_\gamma .$$

This process is repeated throughout the spectrum and finally by adding up all the contributions the Compton electron distribution is obtained. The distribution has to be subtracted from the observed pulse-height distribution. Strictly speaking, the energy interval ΔE_γ should be as small as possible, and the experimentally determined shapes of the Compton distributions for mono-energetic photons should be used instead of assuming a Compton distribution of constant amplitude. In the present calculations an energy of 10 keV is used for ΔE_γ. The Compton electron distribution thus calculated for the present experimental distribution was found to be not very large. This correction becomes important only when there are photons of higher energy present. As regards the correction for back-scattering resulting from the photomultiplier glass window, source-backing material and the crystal container, and also from the surrounding material used as shielding, it was found that for the present experimental arrangement, no back-scattering occurred for incident photon energies less than 412 keV and since the present investigated energy range is much below 412 keV the correction for this effect does not exist.

The significant correction of the measurements in the energy range of interest is that for the escape of iodine K x rays from the top and cylindrical surfaces of the crystal. This effect and that of energy resolution were corrected as follows.

In effecting the iodine K x ray escape correction, for the K x ray escape fraction use is made of the calculated values of Axel (1954) for the case of good geometry. After correcting for K x ray escape, the distribution $N_3(E_\gamma)$ of the scintillations in the crystal gives the distributions $N_4(E_\gamma)$. A first approximation $N'_4(E_\gamma)$ of $N_4(E_\gamma)$ was estimated. The corresponding distribution $N'_3(E_\gamma)$ was then calculated. After that, using the experimentally-determined resolving power, the resulting pulse-height distribution $N'_2(E)$ was calculated according to Liden and Starfelt (1954). When $N'_2(E)$ thus obtained deviates only slightly from the experimental curve, $N_2(E)$, the following equation is valid:

$$N_4(E_\gamma) = N'_4(E_\gamma)N_2(E)/N'_2(E).$$

Otherwise $N'_4(E_\gamma)$ was improved upon and the calculations were repeated. The experimentally measured linewidths at half height at 80 and 145 keV were 22 % and 16 % respectively. The correction for the geometrical and gamma detection efficiency of the detector was arrived at and applied in exactly the same way as was done by Narasimhamurty and Jnanananda (1966, 1967). This correction was less than 2 % for energies below 80 keV.

Correction was also made for absorption in the perspex beta absorber and in the aluminium can (thickness of Al = 0·013 cm) of the NaI(Tl) crystal by using absorption coefficients given in the tables of Hubbel (1969). The correction to IB due to scattered radiation from the perspex beta absorber and from the hole of the lead collimator was calculated by the Klein–Nishina formula, on the assumption of single scattering, and it was found to be negligible. The IB pulse-height distribution included the external bremsstrahlung (EB) emitted non-isotropically in the source itself and also from the perspex beta absorber. An investigation of IB with two sources of different thicknesses showed that the EB in the source itself is negligible. The disturbance of the measurement of IB caused by the EB from the perspex absorber was determined by the extrapolation of the experimental determinations of the EB in different elements Al, Cu and Sn. At the low energies covered by this investigation, the EB was calculated under the assumption that it was emitted isotropically. The EB disturbance from the perspex absorber was found to be less than 2 % at all energies studied.

A fairly accurate estimate was made of the effect of the sum pulses resulting from the finite resolving time of the electronics that might distort the measured distribution of IB at the high-energy end; it was found to be not at all significant.

In the pulse-height distribution of IB shown in figure 1 there appears a peak at 121 keV which may be the gamma line of ^{147}Pm. This line was also observed by the previous investigators (Starfelt and Cedurlund 1957, Langevin-Joliot and Lederer 1956, Hansen 1972). To correct for this gamma-ray line of energy 121 keV, the complete pulse spectrum of the 145 keV gamma-ray line from ^{141}Ce was recorded in the same geometry and the resulting distribution was normalized to the photopeak of 121 keV and subtracted from the IB pulse-height distribution of ^{147}Pm. This correction was found to be not very significant and, as such, the IB distribution is smoothed out by simply subtracting the 121 keV x ray peak.

As well as the way in which the true IB spectrum was obtained from the measured pulse-height distribution by applying individually various corrections as described above, an alternative method can be used for the same purpose: this method is known as the unscrambling method and is due to Starfelt and Koch (1956); it was employed by Starfelt and Cedurlund (1956) in their experimental investigation of IB from ^{147}Pm. According to the unscrambling method the measured pulse-height distribution corrected for background counts as well as for discrete gamma-ray and x ray lines, if any, is related to the photon spectrum $N(E)$ of the radiations incident on the spectrometer crystal as follows:

$$P(E') = \int_0^{E_{\max}} M(E', E)(1 - \mathrm{e}^{-\mu L})N(E)\,\mathrm{d}E. \tag{1}$$

Where $P(E')$ is the measured pulse-height spectrum, $N(E)$ is the incident photon distribution, $M(E', E)$ is the response function that gives the probability of observing a pulse height E' from an incident photon energy E in the crystal under consideration. μ and L are the total absorption coefficient and the thickness of the crystal respectively. Calculation of $M(E', E)$ requires knowledge of the iodine K x ray fraction, the photofraction and the absorption coefficient of the crystal.

As described by Starfelt and Koch, equation (1) can be transformed into a matrix form with a suitable number of rows and columns covering the energy region of interest and can be solved.

Next we come to the survey and estimation of the errors involved in the measurement of the IB distribution as well as in the calculations of the various corrections, the errors due to energy calibration and channel width were very small—not more than 2%. The statistical error, as already pointed out, was less than or equal to 3% throughout the energy range of interest in the present experiment. The errors in the corrections, such as Compton electron distribution, escape electrons and back-scattering effect, were not of any consequence since those corrections themselves were found to be negligibly small. The error in making correction for the 121 keV x ray line that appears in figure 1 was estimated and found to be less than 4% in the energy region where the 121 keV photopeak appears. Thus, in the table of errors (table 1), this error was also taken into account in arriving at the compound error at the energy 120 keV alone as it existed only in that energy region though it was not specifically shown in table 1. The error in the determination of the crystal detection efficiency was of major importance and was partly due to the uncertainties in the values of the absorption coefficients for sodium iodide used in the calculations of the intrinsic efficiencies and partly due to the inaccuracy in the experimental determination of the peak to total ratio, the latter being caused mainly by the radiation scattered from the collimator and the photomultiplier window. Owing to the above errors the maximum uncertainty in the detection efficiency was of the order of 4%. The error in the correction for the EB disturbance from the perspex beta stopper was mainly due to the uncertainty caused by the assumption of an isotropic distribution of the EB. The compound error (RMS value) arrived at from the various errors involved in making these corrections turned out to be less than about 8% at 200 keV and it was found to decrease with decreasing energy. The different possible errors mentioned above are given in table 1 in which the error in the determination of the total activity of the source is not included because this error affected only the total photon energy yield but not the shape of the IB spectrum, as pointed out in several IB spectral-shape studies (Hakeem and Goodbrich 1962, Berenyi and Varga 1969).

Table 1. Calculated errors (%).

Energy (keV)	Channel width	Energy calibration	Resolving power + K x ray escape	Crystal efficiencies	EB from perspex absorber	Counting statistics	Total RMS
50	2	0·5	2	1	1	0·5	3·0
80	2	0·5	1	2	1	0·5	3·0
100	2	1	1	2	1	1	3·1
120	2	1·5	2	4	1	2	7·0
150	2	1·5	2	5	1	3	6·8

For the purpose of comparison between experiment and theory, since the theoretical IB distributions shown in figure 2 were calculated expressing the different energies, such as the beta-particle energies and IB photon energies, in units of mc^2 and obtained as the number of photons per 1 MeV (in units of mc^2) per beta disintegration, the experimental IB distribution after being corrected for different factors mentioned earlier, was also

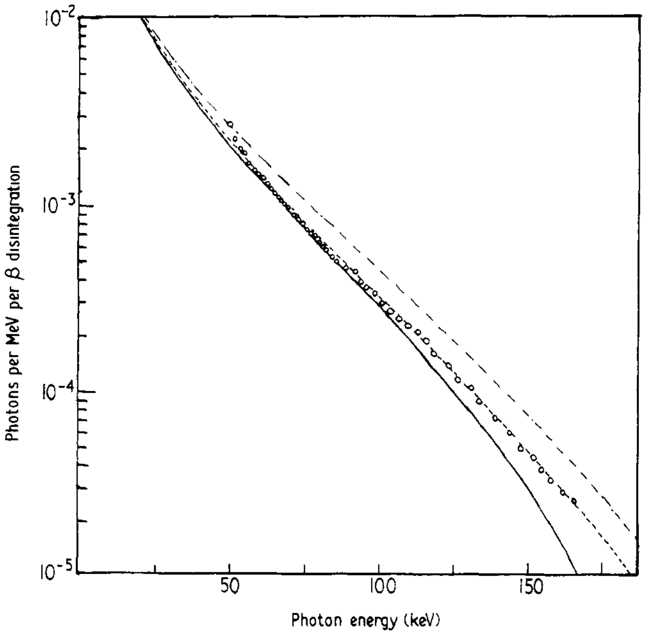

Figure 2. The IB spectral shape along with the theoretical distributions for ^{147}Pm. The full curve is the KUB distribution; the broken curve is the Lewis and Ford distribution; the chain curve is the Nilsson distribution. The open circles give the experimental distribution.

represented as the number of photons per 1 MeV energy interval per beta disintegration against the photon energy. The number of photons per 1 MeV per beta distintegration was obtained firstly by dividing the number of counts (shown as ordinate in figure 1) by the analyser window width ΔK in units of mc^2 and this gave the number of photons per unit energy interval which was then multiplied by 1 MeV in units of mc^2 to get the number of photons per 1 MeV energy interval. Subsequently this number of photons per 1 MeV was divided by the beta strength of the source employed to get the number of photons per 1 MeV per beta disintegration and the experimental distribution expressed as above is shown in figure 2 in the energy range from 50 to 165 keV against the corresponding theoretical distributions due to KUB, Lewis and Ford (1957) and also Nilsson (1956) which were calculated in the usual way.

4. Results and discussion

^{147}Pm is classified according to Langer (1950) as a non-unique first-forbidden beta emitter having an end-point energy of 223 keV and half-life of the order of 2·62 yr. From the figure 2, it can be seen that there is a good agreement between experiment and KUB theory in the energy range 60 to 75 keV and beyond 85 keV there is a gradual deviation between KUB theory and experiment, the experimental results being in slight excess over KUB theory up to 165 keV. The discrepancy between KUB theory and experiment is 13% at 95 keV, 32% at 130 keV and 80% at 155 keV. However, we find a close correspondence between experiment and the theory of Lewis and Ford (1957) throughout the investigated energy region. Nilsson's (1956) theoretical distribution is definitely above the present experimental distribution throughout.

On comparison of the present experimental results with those of an earlier investigation by Starfelt and Cedurlund (1957, to be referred to as SC), the present results are found to lie below those of SC throughout the energy region studied. The experimental IB spectrum of SC was found to be far greater than the theoretical IB distributions due to KUB as well as Nilsson (1956). Concerning the difference between the present results and those of SC it is rather difficult to understand the actual cause, but it can be partly attributed to the following differences between the present investigation and that of SC. Firstly, in the geometrical set-up employed by SC, a collimator channel (cylindrical) with a diameter smaller than that of the spectrometer crystal was used to reduce considerably the back-scatter effect and also to improve the energy resolution. However, such an arrangement presents certain difficulties in the calculation of the solid angle subtended by the spectrometer and, moreover, since the collimator was cylindrical, scattering effects resulting out of multiple scattering may be more than in the present set-up, in which scattering from the walls of the collimator and also from the surface of the photomultiplier uncovered by the crystal was minimized by the use of a conical collimator which limited the beam of radiations strictly to the size of the crystal. Further, the method of evaluation of detector corrections applied to the IB pulse-height distribution in order to get the photon distribution was different in the two cases. As a result of the difference in the methods of evaluation of the experimental IB in the two cases, the compound errors would also be different. Thus SC reported compound errors of 14% at 150 keV and 7% at 60 keV whereas the corresponding errors in the present case were 7% at 150 keV and even less at lower energies.

Next, considering the results of IB from ^{147}Pm obtained by Singh and Dargazelli (1971), their experimental results are found to be far below even the KUB theory throughout the energy region studied. In this investigation, the preparation of the IB source as described appears to be such as to give rise to sizable source bremsstrahlung. Further, it appears that in their experimental arrangement a lot of material surrounding the source and the detector seems to be present. Moreover, they employed a relatively high-Z aluminium absorber to stop completely all the beta particles from the source reaching the detector. All these factors, as a matter of fact, should contribute to an enhancement of the experimental IB distribution but, surprisingly enough, Singh and Dargazelli (1971) reported a very low experimental IB distribution much lower than even the KUB theoretical distribution. This low experimental IB distribution reported by Singh and Dargazelli would only have resulted if too much EB arising from the material around the source and detector (in their experimental arrangement) was subtracted from the experimental distribution. However, Singh and Dargazelli did not mention anywhere in their paper that this was the case. Under these circumstances it appears that it is not possible to properly account for the low value of the IB distribution obtained by Singh and Dargazelli. Here, it may not be amiss to point out that the theoretical estimates reported by Singh and Dargazelli appear to be very large in comparison with the same theoretical values reported by the earlier investigators (Boehm and Wu 1954, Starfelt and Cedurlund 1957). To a large measure this may be the reason for the experimental IB distributions of Singh and Dargazelli (1971) to lie much below the theoretical distributions. Owing to the relatively low intensity of IB in the present investigations necessary precautions are taken to minimize the production of external bremsstrahlung in the source, in the surrounding material and also in the beta stopper by preparing a very-low-intensity source with as small a thickness as possible backed by a very thin and low Z polythene film (1–2 mg cm^{-2}), next by isolating the source and the detector from any external structure (external objects) by a minimum distance of 1 m and by using a perspex beta stopper with a very low Z value of the order of 3.

Finally, it may be mentioned that the general trend of our present experimental IB distribution is comparing favourably with that of SC. Further, it is to be noted that the theories due to KUB and Nilsson (1956) are applicable only to the case of IB from allowed beta decay and the theory due to Lewis and Ford (1957) is applicable even to the case of IB from forbidden beta decays. Again according to the theory of Lewis and Ford (1957) even though a forbidden beta decay and an allowed beta decay with the same W_0 and Z may give a beta spectrum of the allowed shape, the IB spectrum in both the cases in general, need not be the same. In the present case the beta emitter ^{147}Pm is a forbidden one with a beta spectrum of allowed type. So the Coulomb-corrected theory due to Lewis and Ford (1957) for IB is probably the most appropriate theory to be compared with the experimental IB from ^{147}Pm. Actually this is done in the present study and a close correspondence has been observed between this theory and experiment. Also here it is relevant to make mention of the contribution due to the so called detour effect arising out of virtual transitions (Longmier 1949) to the IB spectrum as this effect would generally be present in the case of forbidden beta decays. An estimate of this effect has not been made, since we have already found agreement between experiment and the theory of Lewis and Ford (1957). As the detour effect has not been taken into account, it is not unlikely that the agreement obtained between the present experimental distribution and Lewis and Ford theory is even accidental. Unfortunately an attempt to calculate the effect due to virtual transitions could not be made owing to the lack of satisfactory theories of that effect, although in the literature we find one theoretical paper due to Ford and Martin (1969) concerning that aspect.

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