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Contribution of detour transitions to internal bremsstrahlung spectrum of ^{204}Tl

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Abstract. The internal bremsstrahlung spectrum of ^{204}Tl has been studied experimentally in a photon energy interval of 0·15 to 1·45 mc^2. The experimental photon distribution is found to agree with some of the earlier experiments in an energy region common with these but disagrees with recent measurements of Singh and Al-Dargazelli in the entire energy region.

The present experiments do not confirm theoretical predictions of Ford and Martin regarding the contribution of detour transitions in the intermediate photon energy region. Direct transition KUB Coulomb corrected Nilsson theory is found to be quite adequate in this region. Excess photon yields in the low and high energy regions of the spectrum observed in these experiments are not explained by the presently available theories. This points to the need for more accurate theories of IB spectra which take into account the Coulomb effects more precisely.

1. Introduction

The internal bremsstrahlung (IB) spectrum of beta active ^{204}Tl with $E_\beta^{\max} = 766$ keV has been studied by a few workers. The earliest measurements of Ricci (1958) covered an energy region from 80 to 400 keV. His experimental results were compared with the theories of Knipp and Uhlenbeck (1936), Bloch (1936) and the Coulomb corrected distribution of Nilsson (1956). The experimental results were found to agree with the Coulomb corrected theory of Nilsson in the energy region from 150 to 400 keV while they showed an excess of photons in the energy region below 150 keV. The KUB theory, on the other hand, was found to be lower than experiment in the entire energy region. Later on, Narasimhamurty and Jnanananda (1967) studied the IB spectrum of ^{204}Tl from 100 to 550 keV and found general agreement in an energy region from 300 to 550 keV with the Nilsson theory but in the energy region below 300 keV the experimental results were higher than theory by 60% at 100 keV to 35% at 250 keV. Except for the recent work of Singh and Al-Dargazelli (1971), no attempts have been made to study the IB spectrum of ^{204}Tl in an energy region close to $E_\beta^{\max}$. These workers investigated the IB spectrum of this isotope from 108 to 760 keV, but they have not taken proper precautions against contribution of external bremsstrahlung (EB) due to the geometry of experimental arrangement and, furthermore, they have not applied all the necessary corrections which play an important part in these measurements. Unfortunately, these authors have not used the correct procedure to evaluate the necessary theoretical results which are therefore not appropriate for comparison with their experiment. Consequently, their conclusions that their results are lower than the theory are wrong. These points are further discussed in § 5.

The beta decay of ^{204}Tl is a first forbidden unique transition ($\Delta J = 2$, yes). Longmire (1949), Horowitz (1952) and more recently Ford and Martin (1969) have pointed out that in the case of a first forbidden beta decay, transitions through virtual states contribute appreciably, particularly in the intermediate energy region of the IB spectrum. In these so called detour transitions the parent nucleus first emits the photon and goes to a virtual intermediate excited state from which it subsequently decays to the final state through beta emission. The process can happen in the reverse order as well. Ford and Martin found theoretically that the detour transitions in ^{90}Y (also unique first forbidden) give an expected maximum contribution in the middle of the spectrum and of the order of 25 % of the direct contribution.

In the light of the above situation it was decided to investigate the IB spectrum of ^{204}Tl up to the end-point energy so as to provide an extensive and rigorous check on the KUB theory corrected for Coulomb effects and test the predictions of Ford and Martin in the case of this unique first forbidden beta emitter.

2. Experimental details

A sufficiently strong carrier free source of ^{204}Tl(2·44 mCi) supplied as thallous sulphate in aqueous solution of specific activity 200 mCi g^{-1} Tl, by BARC Bombay, was prepared on a thin polythene film (1·47 mg cm^{-2}) supported on an aluminium ring of inner diameter 5·1 cm. The source material was uniformly distributed on a circular area of 3 mm diameter with the help of a plain insulin solution.

The IB spectrum was recorded with a single channel gamma ray spectrometer using a 7D 8 NaI(Tl) crystal of 1·75 in diameter and 2 in thickness, supplied by the Harshaw Company, coupled to a 6292 Dumount photomultiplier. The detector was placed at a distance of 10 cm from the source and shielded with a 4 cm thickness of lead, lined inside with an aluminium sheet. Beta particles emitted by the source were absorbed in a perspex disc, 600 mg cm^{-2} placed at a distance of 2·8 cm from the source. The detector was also shielded against x rays originating in the lead collimator by placing a brass disc in such a manner that scattering was reduced to a minimum. The geometrical arrangement (figure 1) was worked out on the lines suggested by Berenyi and Varga (1969) so as to minimize the contribution of external bremsstrahlung. On studying the variation in the factor $(w_\beta \times w_{\mathrm{EB}})/w_{\mathrm{IB}}$, where w_β, w_{EB} and w_{IB} are solid angles for beta particles, EB and IB respectively, as a function of absorber to detector distance (as shown by the curve in figure 2). It is clear that for this factor to be a minimum the optimum position for the absorber is 2·8 cm below the source. The actual EB contribution under these conditions, though small, was obtained experimentally.

As mentioned above, the main aim of the experiment was to study the high energy part of the IB spectrum where the yield is extremely small. Therefore, a proper study involved collection of data over a long period of time. This required the electronic apparatus to be extremely stable. This was checked throughout the experimental observations and procedures were framed for recording the data to eliminate effects of peak shift and electronic drift.

The experimental runs for the spectrum were arranged by dividing the entire energy region into four parts. The channel width was varied from 5 to 30 keV to ensure that the channel width at any energy was smaller at least by a factor of three than the line resolution at that energy. Data were collected in steps extending from 75 to 150 keV, 150 to 400 keV, 400 to 600 keV, and 600 to 750 keV with intermediate runs for checks

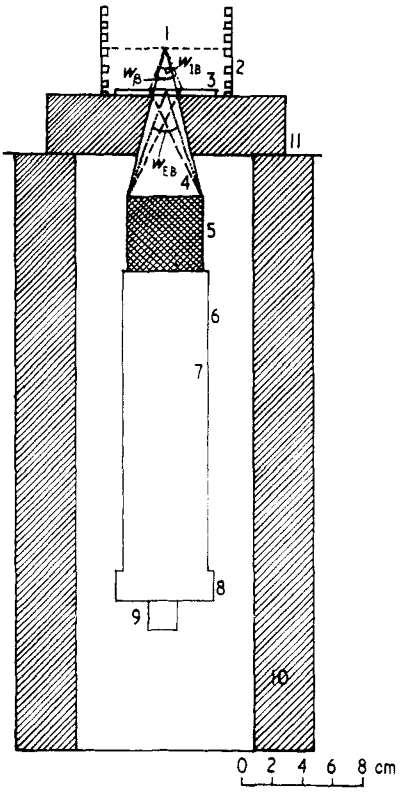

Figure 1. Experimental arrangement. 1 Source of ^{204}Tl on polythene film; 2 perspex source stand; 3 perspex disc for beta absorption; 4 aluminium can containing NaI(Tl) crystal; 5 1·75 × 2 in^2 NaI(Tl) crystal; 6 black adhesive tape; 7 photomultiplier tube; 8 cathode follower; 9 adjustable support; 10 4 cm thick lead shield; 11 brass plate.

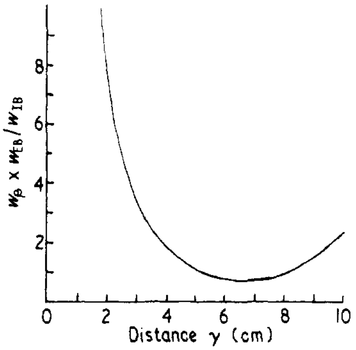

Figure 2. Variation of $(w_\beta \times w_{EB})/w_{IB}$ as a function of detector to absorber distance for a fixed source to detector distance of 10 cm.

on the stability by using standard gamma emitters, namely, ^{141}Ce, ^{203}Hg, ^{51}Cr, and ^{137}Cs to cover the various energy intervals. These were followed by runs for room background. Sufficient counts were recorded so as to ensure counting statistics better than 3 % even at 750 keV.

Spectra were obtained with the same source at different intervals of time extending over a period of five months to ensure that there was no contamination. Also sources of different strengths were employed to check the effect of source strength on the IB spectrum, if any. The strengths of various sources were measured by an end-window GM counter using a standard geometrical set up.

3. Corrections to IB distribution

Spectral distributions corrected for room background, obtained in the above manner, were reduced to a common window interval in energy and plotted. The spectrum recorded by the crystal was transformed to the spectrum emitted by the source through the following steps. The spectra of standard gamma emitters, ^{141}Ce, ^{203}Hg, ^{51}Cr, ^{137}Cs and ^{54}Mn were recorded in the same geometrical set-up and used in calculating energy resolution, photofraction, and back scattering peak ratios. The experimentally recorded pulse-height distribution of the IB spectrum was corrected for the energy resolution of the entire assembly by the procedure given by Novey (1953). The magnitude of the correction was found to vary from 8 % at 150 keV to 2 % at 350 keV and is significant only at low energy. The iodine escape peak correction was applied by using data given by Crouthamel (1960) and its contribution varied from 1 % at 180 keV to 9 % at 94 keV. The correction due to back scattering of photons was also obtained from experimental data and the average ratio of the intensities of back scatter peak to photopeak was found out to be 4 %. This correction was mainly in the energy region from 150 to 200 keV. The spectrum was then corrected for the contribution due to the Compton continuum present in the low energy photon region, due to higher energy photons, by using the experimental data obtained above for the crystal and employing the procedure used by Narasimhamurty and Jnanananda (1967). This contribution was found to be 1 % or less in the entire energy region covered in the present measurements.

Contributions due to counting of EB along with IB were obtained and corrected for using absorbers of perspex, aluminium, tin and copper of the same equivalent thicknesses. Four spectra (EB + IB) were recorded, one for each absorber. The counting rates corresponding to different energies were corrected for the different absorption in the absorbers and then plotted as functions of atomic number (Z). Counting rates corresponding to $Z = 0$ gave the IB contribution. The external bremsstrahlung contribution with the geometrical set-up in question was found out to be 5 % in the energy interval from 150 to 500 keV.

Pile-up effects on the spectra were found to be negligible. All the important corrections are depicted in figure 3. The geometrical and intrinsic efficiencies of the crystal were taken from the data given by Crouthamel (1960). After applying all the corrections the spectrum was reduced to photons per unit energy interval (mc^2) per beta disintegration, by dividing the data by window width (units of mc^2) and the beta strength of the source. These spectra are shown in figure 4 for two sources of ^{204}Tl in an energy interval from 0·15 to 1·45 mc^2.

4. Errors

The most important error is ascribed to uncertainties in the values of intrinsic efficiency and photofraction. The error quoted in the values of intrinsic efficiencies is within 2 to

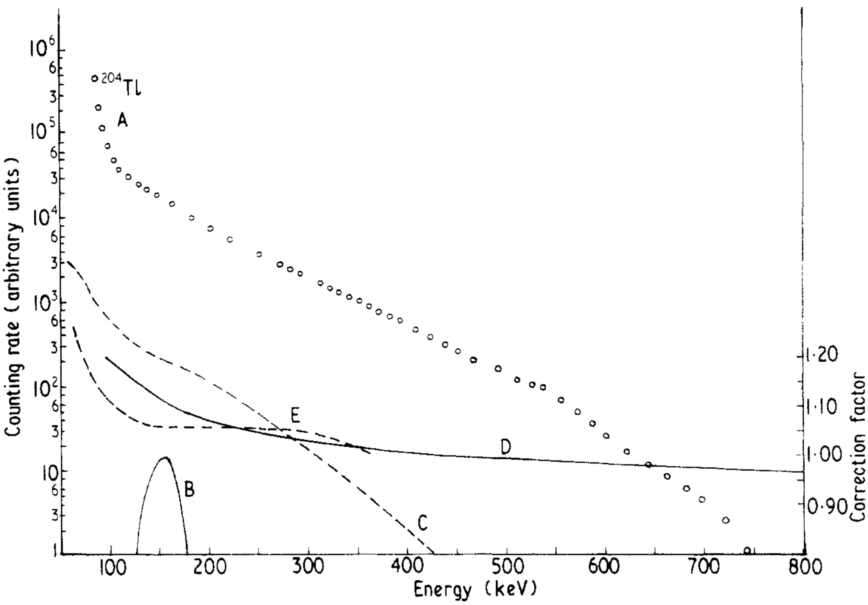

Figure 3. Experimental spectrum and various corrections. A Experimental pulse height distribution of IB photons corrected for background, ^{204}Tl = 2·44 mCi; B back scattered contribution; C Compton electron distribution; D correction factors, absorption in perspex, aluminium can of NaI(Tl) crystal and external bremsstrahlung; E correction factors due to iodine K x-ray escape and energy resolution.

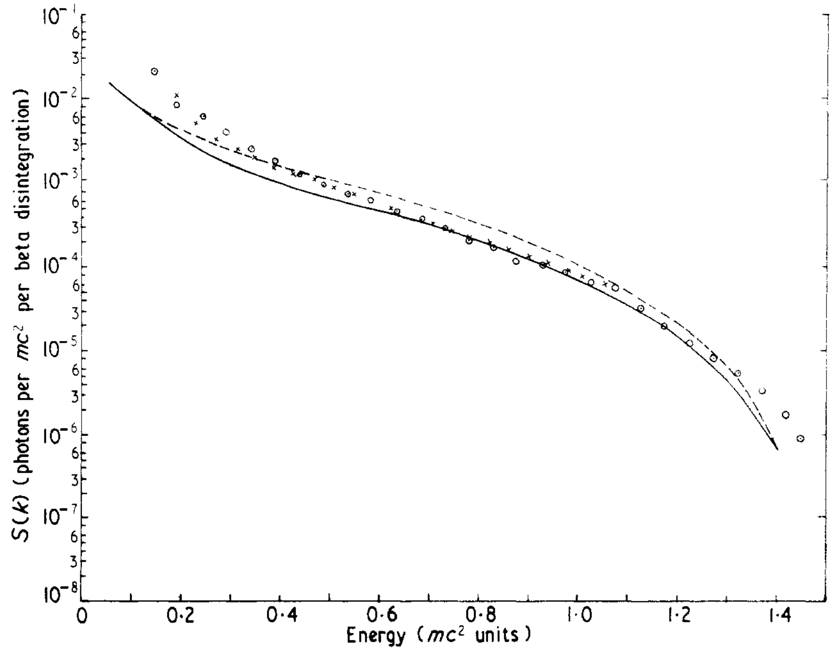

Figure 4. Experimental and theoretical IB distributions for ^{204}Tl. Full curve, direct Nilsson distribution; broken curve, (direct Nilsson + detour) after Ford and Martin. ○ Experimental data ^{204}Tl (2·44 mCi); × experimental data ^{204}Tl (0·46 mCi).

3 % and the experimentally determined photofractions are uncertain by less than 4 %. These two factors contribute an overall error in detection efficiency of not more than 5 %. Uncertainties due to statistics in recording the data are better than 3 % even at 750 keV. Errors involved in the back scattering correction, energy resolution, iodine escape correction and in the correction due to external bremsstrahlung are very small as these contributions by themselves are very small. The error involved in approximating the Compton electron distribution to a straight line for the purpose of calculating the Compton contribution is estimated to be of the order of 5 % by comparing the straight line approximation for the Compton electron distribution for a gamma ray energy to the actual Compton electron distribution obtained from the experimental spectrum. Also, the errors in energy calibration and uncertainty in window width were small. The maximum uncertainty in the determination of the absolute beta activity was of the order of 8 %. The source strength, however, affects the photon yield but not the shape of the spectrum. The results for this isotope are correct within 10 % which is the RMS value of the various errors involved at 700 keV. The errors decrease as the energy decreases because of better statistics at low energy.

5. Theory

Knipp and Uhlenbeck (1936) and Bloch (1936) examined theoretically the problem of internal bremsstrahlung for allowed beta transitions using the polar–vector interaction by the method of second order perturbation theory and obtained the probability per unit time for the emission of continuous radiation due to the sudden change in nuclear charge when a beta particle leaves the nucleus. In an alternative method Knipp and Uhlenbeck considered the radiative beta emission as a two-step process. First they calculated from beta decay theory, the probability $P(W_e)\,\mathrm{d}W_e$ for the creation of an electron with energy W_e and secondly the probability $\phi(W_e, k)$ for the emission of a photon of energy k by the electron, using first order perturbation theory. According to the KUB theory the number of IB photons of energy k per beta disintegration per unit energy interval for a beta emitter with end-point energy W_0 was given by $S(k)$ such that

$$S(k) = \int_{1+k}^{W_0} P(W_e)\,\mathrm{d}W_e\,\phi(W_e, k). \tag{1}$$

The function $\phi(W_e, k)$ is given by

$$\phi(W_e, k) = \frac{\alpha p}{\pi p_e k}\left(\frac{W_e^2 + W^2}{W_e p}\ln(W+p) - 2\right) \tag{2}$$

where p_e, p and W_e, W are the momenta and energies of the electron before and after the emission of the IB photon and α is the fine structure constant. Chang and Falkoff (1949) and Madansky *et al* (1951) examined the case of forbidden transitions and various types of beta interactions. It was found that $S(k)$ depends on the degree of forbiddenness of the beta transition and the type of interaction only at high photon energies.

The above calculations make use of a plane wavefunction for the electron instead of a Coulomb wavefunction for the sake of simplicity. This results in the neglect of the effect of nuclear charge on IB radiation. In the Coulomb corrected theories these effects are taken into account to different extents. In the Nilsson approximation the function $\phi(W_e, k)$ should be replaced by $\phi(W_e, k, Z)$ such that

$$\phi(W_e, k, Z) = \phi(W_e, k)\frac{F(Z, W)}{F(Z, W_e)} \tag{3}$$

where $F(Z, W)$ is the Fermi function for allowed transitions. Lewis and Ford (1957) considered the first order correction in Z by using

$$\phi(W_e, k, Z) = \phi(W_e, k)\frac{1+\pi\alpha ZW/p}{1+\pi\alpha ZW_e/p_e}. \tag{4}$$

The latter approximation gives essentially the same results as Nilsson's correction for low Z values.

In order to calculate $S(k)$ for ^{204}Tl the values of $\phi(W_e, k, Z)$ are calculated from (2) and (3) following the suggestion of Lewis and Ford (1957), at different values of W_e in the range $1+k$ to W_0. The values of $P(W_e)\,dW_e$ are taken from the experimental beta spectrum of ^{204}Tl (Yuasa *et al* 1955). This step results in taking into account automatically the actual nature of the beta interaction, the forbiddenness and also to some extent the Coulomb effects of nuclear charge. The integral (1) for $S(k)$ is then evaluated numerically. The result is shown in figure 4.

Singh and Al-Dargazelli (1971) have calculated the value of ϕ for ^{204}Tl from equations (2), (3) and (4) for k varying from 0·1 to 1·4 mc^2 but did not evaluate $S(k)$. Consequently their comparison with the theory is erroneous and the conclusions are incorrect.

Longmire (1949) and Horowitz (1952) predicted qualitatively that in the case of forbidden beta decays, the contribution of detour transitions to the IB spectrum should be appreciable. These effects should, however, be negligible for allowed transitions. More recently, Ford and Martin (1969) have examined the problem quantitatively and calculated the effect of detour transitions for unique first forbidden beta decays ($\Delta J = 2$, yes). Explicit data for a medium Z nucleus ^{90}Y ($W_0 = 5{\cdot}4\,mc^2$) have been given. Using their expressions for the contribution of detour transitions we have calculated and presented in figure 4, the results for ^{204}Tl ($W_0 = 2{\cdot}5\,mc^2$). We have also replaced the factor $(1+\pi\alpha ZW/p)$ by the Fermi function $F(Z, W)$ and found that the results are not affected to any appreciable extent.

6. Results and conclusions

^{204}Tl is a unique first forbidden beta emitter with a halflife of 3·8 year and $E_\beta^{\max} = 766$ keV (Lederer *et al* 1968). There also exists a weak electron capture branch (2·1 %) in it. The experimental IB photon distribution is plotted in figure 4 for two sources of ^{204}Tl along with theoretical values calculated on the basis of direct KUB Coulomb corrected (Nilsson) theory. The experimental distribution agrees closely with the direct KUB Coulomb corrected theory in the energy region from 0·6 to 1·2 mc^2, whereas an excess of photons is observed both below 0·6 mc^2 and above 1·2 mc^2. The experimental distribution coincides with the detour corrected Nilsson distribution only at 0·3 to 0·5 mc^2 and then at 1·3 mc^2 and lies below it in the entire energy region from 0·5 to 1·3 mc^2. The experimental distribution is in excess of both the theories below 0·3 mc^2 and above 1·3 mc^2. The excess of photons below 0·3 mc^2 is too large to be ascribed to the tail of the Hg K x ray peak and the IB accompanying the electron capture for which the end-point energy is about 400 keV. According to Ricci (1958), IB accompanying the electron capture cannot affect the IB photon distribution at low energy by more than 10 %. The excess of photons above 1·3 mc^2 may be attributed to inadequate Coulomb corrections used in the theory.

We find that our experiments on the internal bremsstrahlung spectrum of ^{204}Tl agree with those of Ricci (1958) and Narasimhamurty and Jnanananda (1967), but disagree with those of Singh and Al-Dargazelli (1971) in the energy regions common to these experiments. The KUB Nilsson theory is found to be quite adequate to describe the experiments in the intermediate energy region of the IB spectrum. No contributions of detour transitions, as predicted by Ford and Martin, are observed in this region. It will be of interest to investigate the IB spectra of other unique first forbidden beta emitters, with low Z and high $E_\beta^{\max}$ values, to test the predictions of Ford and Martin regarding detour transitions more rigorously. The presently available theories of internal bremsstrahlung in beta decay do not explain the observed excess of photons in the low and high energy parts of the spectrum. We stress the need for an accurate Coulomb corrected theory to explain this.

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