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L shell ionization in the α decay of ^{210}Po

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Abstract. The number of L x-rays of lead emitted per α disintegration of ^{210}Po has been measured, in terms of the known emission of neptunium L x-rays by ^{241}Am, using a thin Ge(Li) x-ray detector, a low geometry α particle counter and a weak (~4 μCi) source of ^{210}Po. The result, $(1{\cdot}41 \pm 0{\cdot}12) \times 10^{-4}$ L x-rays per α, is substantially lower than the results of previous measurements but is still in marked disagreement with the predictions of calculations based on the theoretical approach of Migdal. The result is in fair agreement with a recent calculation, using a special formulation of the binary encounter approximation, by Hansen. Previous theoretical and experimental work is reviewed and the results of a simple calculation of the L shell ionization probability, using the sudden approximation, are given.

1. Introduction

The development of a new generation of high resolution x-ray detectors of Ge(Li) and Si(Li) type has led to renewed interest in the study of atomic effects in nuclear transitions. When the nuclear charge changes in a reaction, the fact that the atomic electrons contribute to the process through changes in their binding energy is properly accounted for by the use of atomic masses in the calculation of the energetics. In the case of nuclear β decay, for instance, it has long been realized that the change in electronic binding energy as the nuclear charge increases by one unit is available for sharing between the electron and anti-neutrino (apart from a very small amount accounted for by the fact that the final atom is singly ionized in an outer shell), so that there is no cut-off of the electron energy spectrum below a value corresponding to the difference between the initial and final electronic binding energies. The essential 'one-step' nature of β decay has been emphasized by Stephas and Crasemann (1967) in their calculation of the probability of the higher-order process whereby an inner shell electron is emitted during the β decay process, and more recently by Law and Campbell (1972) who have modified the theory of this effect by giving a proper treatment of the indistinguishability of the emitted electrons. The contribution to the ionization arising from a direct collision of the emitted electron with an inner shell electron appears to be negligible as long as the average energy of the β particle is very much greater than the binding energy of the inner shell electrons.

There is also a probability of inner shell ionization accompanying α decay but, as will be shown below, theory and experiment are in very poor agreement and there have been no measurements of the effect performed with solid-state x-ray detectors. The work described here consists of a measurement of the number of lead L x-rays emitted per α decay of ^{210}Po, using a high resolution Ge(Li) detector and a very much weaker source than those used in previous measurements (μCi compared with mCi).

^{210}Po is well suited to this type of investigation since it has a 138·4 d half-life and a particularly simple decay scheme (Lederer *et al* 1967) consisting of a predominant 5·305 MeV ground state to ground state α transition and an extremely weak (0·001 %) feed to the 0·803 MeV level of ^{206}Pb. The number of inner shell vacancies arising from internal conversion of the γ ray can be estimated as $1{\cdot}2 \times 10^{-7}$ and $0{\cdot}3 \times 10^{-7}$ per decay, for the K and (L + M) shells respectively, from the total conversion coefficient measurement of Ovechkin and Tsenter (1957) and the K/(L + M) ratio of Alburger and Pryce (1954). It therefore appears certain that almost all of the lead K, L and M x-rays which are observed in the decay of ^{210}Po arise from the act of α decay itself. All the experimental work which has so far been done on the atomic consequences of α decay has used ^{210}Po and, in the review which follows, all calculated quantities refer to this particular decay.

2. Review of theory and previous experiments

The probability of ionization of the inner electronic shells during α decay was first calculated by Migdal (1941) and the theory has since been refined by Levinger (1953, 1955), Schwartz (1955), Rubinson (1963), Ciocchetti and Molinari (1965) and Hansen (1974). With the exception of Hansen's work all the calculations are elaborations of the original approach of Migdal, who treated the Coulomb interaction between the α particle and the atomic electrons as a small perturbation and applied time-dependent perturbation theory in the adiabatic approximation, assuming the α particle not to be deflected by the electrons and ignoring nuclear recoil. The calculation involves, unfortunately, a power series expansion in terms of successive time derivatives of the perturbation, the terms giving rise to monopole, dipole, etc, transition probabilities with appropriate selection rules, and a difficulty arises in deciding where to terminate the series. Migdal considers the dipole term only, as do Schwartz and Rubinson, whereas Levinger includes the quadrupole term as well, and Ciocchetti and Molinari (whose approach is slightly different since they use an integral expansion of the perturbation) find that the monopole term is the most important one for the K shell, being some 3·5 times larger than the dipole term. Their dipole result is close to those of Levinger and Rubinson. Hansen's approach is radically different because the ionization probability is derived as the special case of zero impact parameter in a generalized impact parameter formulation of the binary encounter approximation. This approximation is much used in calculations of the inner shell ionization produced during ion–atom collisions and is described in a recent review by Garcia *et al* (1973).

Hansen considers that nuclear recoil can be neglected and Migdal ignores it, but the other authors all consider this to be important, although the details of the various treatments are different and the results do not always agree. The effect is as follows. Migdal gives the ionization probability for an electron in a shell (n, l) as

$$\left(\frac{2v_\alpha}{v_K}\right)^2 \frac{C_{nl}}{Z^2}$$

where v_α is the speed of the α particle, v_K is the speed of a K electron in the initial nucleus of charge Z and the C_{nl} are sums of squares of hydrogen dipole matrix elements. Levinger (1953) on the other hand, gives

$$\left(\frac{Zv_{cc}}{v_K}\right)^2 \frac{C_{nl}}{Z^2}$$

where v_{cc} is the speed of the centre of charge of the α particle and the recoiling nucleus, centre of charge being defined in a manner exactly analogous to centre of mass, and the result of Rubinson (1963) is almost the same as this. Thus the probability is reduced by a factor of about 25 by introducing recoil. Schwartz's version of the theory gives a result rather closer to Migdal's, reducing it to about two thirds of its value, but Rubinson (1963) implies that this development of the theory is incorrect. Ovechkin and Tsenter (1957) argue, not very convincingly, that the effect of recoil is to increase slightly the ionization probability. The effect of recoil on the quadrupole term is very small but, according to Levinger (1953), this term dominates in the L shell.

Although our concern is with the L shell ionization probabilities, theoretical and experimental results for the K, L and M shells are given in tables 1 and 2. The predicted quantities (ionization probabilities) and those measured (x-ray yields) are related by the various fluorescence yields and Coster–Kronig transition probabilities and these have been taken from the review of Bambynek *et al* (1972). It should be pointed out that comparison of theory and experiment is not entirely straightforward because the theoretical formulae contain Z explicitly and the question of screening arises. Also, especially for the M shell, the choice of fluorescence yield might be difficult if, as is suggested by Rubinson (1963), there are substantial numbers of vacancies in the outer shells. Since the selection rules governing Auger electron emission are less restrictive than those for the radiative filling of inner shell vacancies, the fluorescence yield might be altered when there are vacancies in the outer shells but the effect is not readily predictable since it will obviously depend upon the details of the vacancy distribution, in particular, on the relative extent to which the sub-shells which supply the electrons for radiative filling are denuded. It is hardly likely, however, that any reasonable allowance for screening and outer shell vacancies could bring the theoretical and experimental results displayed in tables 1 and 2 into agreement.

Table 1. Summary of theoretical predictions of the x-ray yields of Pb associated with the α decay of ^{210}Po.

Reference	K x-rays	L vacancies	Total L x-rays	M vacancies	Total M x-rays
Migdal (1941)	$2{\cdot}6 \times 10^{-6}$	2s: $0{\cdot}84 \times 10^{-5}$ 2p: $1{\cdot}56 \times 10^{-5}$	$0{\cdot}87 \times 10^{-5}$	3s: $1{\cdot}4 \times 10^{-5}$ 3p: $3{\cdot}7 \times 10^{-5}$ 3d: $3{\cdot}0 \times 10^{-5}$	$2{\cdot}4 \times 10^{-6}$
Levinger (1955)	10^{-7}	2s: $0{\cdot}036 \times 10^{-5}$ $2p_{1/2}$: $0{\cdot}99 \times 10^{-5}$ $2p_{3/2}$: $3{\cdot}98 \times 10^{-5}$	$1{\cdot}76 \times 10^{-5}$		
Schwartz (1955)	$1{\cdot}8 \times 10^{-6}$				
Rubinson (1963)	$1{\cdot}36 \times 10^{-7}$	2s: $0{\cdot}85 \times 10^{-6}$ $2p_{1/2}$: $0{\cdot}64 \times 10^{-6}$ $2p_{3/2}$: $1{\cdot}27 \times 10^{-6}$	10^{-6}	3s: $4{\cdot}9 \times 10^{-6}$ 3p: $17{\cdot}5 \times 10^{-6}$ 3d: $33{\cdot}5 \times 10^{-6}$	$2{\cdot}22 \times 10^{-6}$
Ciocchetti and Molinari (1965)	$0{\cdot}85 \times 10^{-6}$				
Hansen (1974)	$1{\cdot}96 \times 10^{-6}$	2s: $2{\cdot}3 \times 10^{-4}$ $2p_{1/2}$: $0{\cdot}76 \times 10^{-4}$ $2p_{3/2}$: $2{\cdot}86 \times 10^{-4}$	$1{\cdot}83 \times 10^{-4}$	3s: $17{\cdot}4 \times 10^{-4}$ 3p: $56{\cdot}6 \times 10^{-4}$ 3d: 116×10^{-4}	$0{\cdot}57 \times 10^{-3}$

Table 2. Summary of previous experimental results.

Reference	Shell	Source strength (mCi)	Detector	Measured yield
Grace *et al* (1951)	K	213	Scintillation and proportional	$(1{\cdot}5 \pm 0{\cdot}5) \times 10^{-6}$
Barber and Helm (1952)		1	Scintillation	$(2 \pm 0{\cdot}4) \times 10^{-6}$
Riou (1952)		8 and 11	Geiger	$(1{\cdot}6 \pm 0{\cdot}5) \times 10^{-6}$
Ovechkin and Tsenter (1957)		7·5	Scintillation	$(1{\cdot}5 \pm 0{\cdot}4) \times 10^{-6}$
Curie and Joliot (1931)	L	27–38	Ionization chamber	4×10^{-4}
Riou (1952)		8 and 11	Geiger	$(2{\cdot}6 \pm 0{\cdot}5) \times 10^{-4}$
Rubinson and Bernstein (1952)		0·5–4	Proportional	$(2{\cdot}93 \pm 0{\cdot}4) \times 10^{-4}$
Curie and Joliot (1931)	M	27–38	Ionization chamber	$1{\cdot}5 \times 10^{-3}$
Rubinson (1963)		0·044–0·44	Proportional	$(0{\cdot}91 \pm 0{\cdot}13) \times 10^{-3}$

Overall, Hansen obtains fair agreement with experiment, but the other calculations give poor agreement, except for the K shell where only the results of Levinger (1955) and Rubinson, that is the dipole results corrected for recoil, are in complete disagreement with experiment. The effect of neglecting nuclear recoil for the L shell can be calculated by adding Migdal's dipole results to the quadrupole results of Levinger (1955), since the quadrupole probabilities are hardly affected by recoil, and this gives the following vacancy distribution per α decay:

$$2s: \quad 0{\cdot}84 \times 10^{-5}$$

$$2p_{1/2}: \quad 1{\cdot}49 \times 10^{-5}$$

$$2p_{3/2}: \quad 4{\cdot}98 \times 10^{-5}$$

and a total L x-ray yield of $2{\cdot}6 \times 10^{-5}$ per disintegration, which is still a long way short of the experimental results. The M shell predictions based on Migdal's method are even further away.

The theoretical L and M shell results are so poor, and not improved much by neglect of recoil, that it seems fair to question the use of the adiabatic approximation and it is useful for this purpose to do some semi-classical, order of magnitude calculations. For the K shell, the adiabatic approximation is justified if we recall that the speed of an electron in a Bohr orbit (Z, n) is Z/n atomic units (au), whereas the speed of the ^{210}Po α particle is 7·3 au, and that the radius of the orbit is n^2/Z au. Let us assume that the α particle only perturbs the electrons in a given shell, n, while it is passing through the region between shells $(n-1)$ and $(n+1)$. Then the K electron has time to make more than seven orbits while the α particle is passing from the nucleus to the L shell radius and the process is one of adiabatic adjustment to a changing potential, which accounts for the very low probability of K shell ionization. An L shell electron, however, makes rather less than two orbits in the time that the α particle takes to travel between the K and M

radii and the adiabatic approximation might be expected to be poor for the L shell, and even worse for the M shell, as indeed appears to be the case.

The requirement for the validity of the other extreme approximation of time-dependent perturbation theory—the sudden approximation—is that the time for which the perturbation acts should be small in comparison with the periods $h/(E_k - E_l)$ corresponding to the eigenfunctions k and l that appear when the hamiltonian changes (Schiff 1955). These times are comparable for the L shell electrons and so this approximation is also of doubtful validity, as is apparent also from the considerations of the previous paragraph. Since, however, the experimental work reported here is concerned with the L shell and no calculations using the sudden approximation seem to have been performed, such a calculation has been carried out to see what the predictions are.

3. Results of the sudden approximation applied to the L shell

Application of the sudden approximation to this problem requires calculation of quantities of the form

$$P_{n,0} = \left| \int \psi_n^*(Z-2, r)\psi_0(Z, r)\, \mathrm{d}^3 r \right|^2$$

where $P_{n,0}$ is the probability of an electron in state 0 ending up in state n, ψ_0 is the (normalized) radial part of the non-relativistic hydrogen-like wavefunction of Po and ψ_n the appropriate final state wavefunction in Pb. The selection rules are those of a monopole transition. In order to avoid the use of continuum wavefunctions, the probability of at least one electron in any one sub-shell being excited was found by calculating $(1-P^N)$ where $P = P_{2s,2s}$, or $P_{2p,2p}$, is the probability that no transition occurs and N is the appropriate sub-shell population. This overestimates the transition probability because the Pauli principle operates and so the overlaps to the filled states were calculated and subtracted from the quantity $(1-P^N)$, allowance again being made for the sub-shell populations. The calculations were performed as far as the 5s and 5p states, but if there is any significant ejection of electrons from the N and 0 shells then the final results will be rather low. Effective charges $(Z-4)$ were used (Levinger 1953) with the following results for the number of L sub-shell vacancies per α decay:

$$2s: \qquad 1-P^2-2\sum_{n=3}^{5} P_{ns,2s} = 11{\cdot}9\times 10^{-4}$$

$$2p_{1/2}: \qquad 1-P^2-2\sum_{n=3}^{5} P_{np_{1/2},2p_{1/2}} = 3{\cdot}85\times 10^{-4}$$

$$2p_{3/2}: \qquad 1-P^4-4\sum_{n=3}^{5} P_{np_{3/2},2p_{3/2}} = 7{\cdot}7\times 10^{-4}.$$

In using the sudden approximation there is another consideration to be taken into account, namely that the process of ionization and α decay might take place in a single step, as is the case in β decay, and the energy necessary to cause single ionization in the L shell would then have to appear at the expense of the α decay energy with consequent reduction in the probability of α decay by an amount easily calculable by the Gamow theory. Thus, taking the energy loss as equal to the values of the L absorption edges of lead (because the emitted electrons have low energy), the above vacancy distribution is

reduced by factors of 0·81 for the $2s_{1/2}$ and $2p_{1/2}$ levels and 0·84 for the $2p_{3/2}$ level. Multiplying by these factors and converting to total x-ray yield gives the result as 7×10^{-4} L x-rays per α particle. This value is higher than the experimental results, which is to be expected, but is closer than the results based on the adiabatic approximation.

The use of non-relativistic hydrogen-like wavefunctions in this region of high Z might be expected to introduce errors. Carlson *et al* (1968) have carried out extensive overlap calculations for the case of β decay using relativistic Hartree–Fock–Slater wavefunctions and find that, for high Z, the hydrogen wavefunctions give values which are low by about a factor of two. It is therefore possible that the above results would be increased by a factor of two if more realistic wavefunctions were used, and this would make them about as far from experiment in the one direction as the adiabatic results are in the other, although these latter results will be affected by relativistic corrections as well.

Finally, it should be mentioned that the calculation of Hansen (1974) includes relativistic effects associated with the bound electrons, uses screened, hydrogen-like wavefunctions and takes into account the fact that the α particle does not move through the atom at constant speed because of its Coulomb interaction with the nucleus, an effect which is ignored in all the other calculations.

4. Experimental details

4.1. Outline of the method

The x-ray detector used in this work was a thin Ge(Li) detector of (nominal) sensitive surface area 100 mm^2 and 5 mm depletion depth having resolution of 280 eV at 5·9 keV. The efficiency of such a detector shows a discontinuity at the energy of the K absorption edge of Ge(11·103 keV) because of two effects. Firstly, x-rays above this energy can excite Ge K x-rays in the detector, which have a certain probability of escape and, secondly, there is invariably a dead layer of Ge at the front face of the detector, or at least a region of poor charge collection, and this layer naturally gives more absorption at energies just above the K edge. Both these effects tend to reduce the efficiency above the edge and, unfortunately, the L x-rays of lead have energies which straddle the discontinuity. There is also an electrical contact in the form of a thin (100–500 $\mu g\,cm^{-2}$) layer of gold on the front face of the detector and this can give rise to a further discontinuity in efficiency at the energy of the L_{III} absorption edge of gold (11·919 keV).

Absolute calibration of the x-ray detector is difficult because accurately calibrated x-ray sources in the appropriate energy range and geometrical configuration are not readily obtainable and therefore the method of measuring the yield of lead L x-rays adopted in this work was one of comparison, specifically a comparison of the number of lead L x-rays emitted from a source of ^{210}Po with the number of L_α x-rays of neptunium emitted from a source of ^{241}Am, with the two sources being made as nearly identical as possible. The x-ray counting of the sources was done in the same geometry and the source strengths were compared in a low geometry α counting arrangement which is described below. The relative efficiency of the x-ray detector was measured by comparing K_β/K_α ratios of elements in the range $Z = 31$ to 37 as measured by the Ge(Li) detector with the latest values in the literature. The details of this procedure are given by Scott (1974).

If the Po and Am α counting rates are P_α and A_α respectively (as measured in the low geometry counter), f and g are the numbers of lead L x-rays and neptunium L_α x-rays

per disintegration, X_{Pb} and X_{Np} the x-ray counting rates, and E_{Np} and E_{Pb} the x-ray detection efficiencies then we have

$$f = g\frac{X_{Pb}}{P_\alpha}\frac{A_\alpha}{X_{Np}}\frac{E_{Np}}{E_{Pb}}.$$

This procedure could be used to generate absolute detection efficiencies over a limited energy range, with the advantage of using only one absolute photon yield from the literature, namely g. The ratio E_{Np}/E_{Pb} was deduced for each lead L x-ray component from the relative efficiency curve obtained from the K_β/K_α measurements.

The quantity g is given by Hansen *et al* (1973) as $0{\cdot}135 \pm 0{\cdot}003$ and this, according to these authors, is the value recommended by IAEA (which agency supplies sets of standardized sources). This value is presumably taken from the work of Magnusson (1957), which appears to be the only absolute measurement, but, according to that author, although the total number of L x-rays per disintegration should be accurate to about 2 %, the error in the relative intensities of the α, β and γ groups is more like 5 % and therefore the uncertainty in the value of g is probably $\pm 0{\cdot}007$.

4.2. Preparation of sources and α counting

Sources of ^{210}Po were made from a stock solution of 1 mCi of Po, obtained as 5 ml of the nitrate in 3M nitric acid from the Radiochemical Centre, Amersham, by dropping, as closely as possible to the centre of the holders, a single drop of stock solution onto holders made of 1·7 mg cm^{-2} mylar sheet cemented to 2·5 cm diameter perspex rings. Autoradiographs showed that the sources were roughly circular and about 4 mm in diameter. They were barely visible to the naked eye. After the sources had dried, they were sprayed with a commercial PTFE solution which provided a covering sufficient to contain the sources whilst being thin enough not to introduce uncertainties into the α counting. Sources of ^{210}Po have a reputation (deserved) for 'wandering' and it is essential frequently to monitor sources and apparatus to make sure that none of the source has escaped. Sources of ^{241}Am were made in the same way from a similar type of stock solution on exactly the same kind of source holders. Typical source strengths were in the range 1–4 μCi.

Although the source strengths are not required for this experiment, it is convenient to be able to measure them so that the x-ray detector can be calibrated absolutely for other work, and a low geometry α counting chamber of precisely known dimensions was therefore constructed. The entire construction was of dural and consisted of a cylindrical chamber some 15 cm high by 8 cm in diameter containing, at the bottom, a block with a recess into which the source holders fitted snugly, then a 5 cm spacer, a plate with a 6 mm diameter hole and finally another block into which was inserted the α detector. Electrical and vacuum connections were brought out through the lid of the chamber. The hole defining the solid angle was spark-cut to a knife edge of some 0·05 mm depth, and this, together with all the relevant dimensions of the apparatus, was measured by the Metrology Group at the National Engineering Laboratory. The geometry factor for α counting was calculated to be 0·00099, with an uncertainty of ± 1 % calculated from the outer limits of the measurements.

The α detector was of silicon surface barrier type connected in the usual way through a pre-amplifier, main amplifier and discriminator either to a simple scaler or to a multichannel analyser, and the counting was performed integrally above as low a bias setting

as possible. Counts were extrapolated to zero bias level by obtaining an energy calibration using an extremely thin source of Pu (^{239}Pu with a small amount of ^{238}Pu), although, with the bias level used, the correction was entirely negligible.

The experiment was performed using the best ^{210}Po source and the best ^{241}Am source, that is, the sources showing the smallest and most uniform autoradiographs and positioned nearest to the centre of the source holders. (In the interests of safety the sources were made inside a 'glove bag' which was in turn inside a large glove box and it was difficult to place the drops exactly in the centre of the holders.) The decay of the ^{210}Po source used in the experiment was followed over a period of 280 d and the decay curve is shown in figure 1 together with a 138·4 d half-life line for comparison.

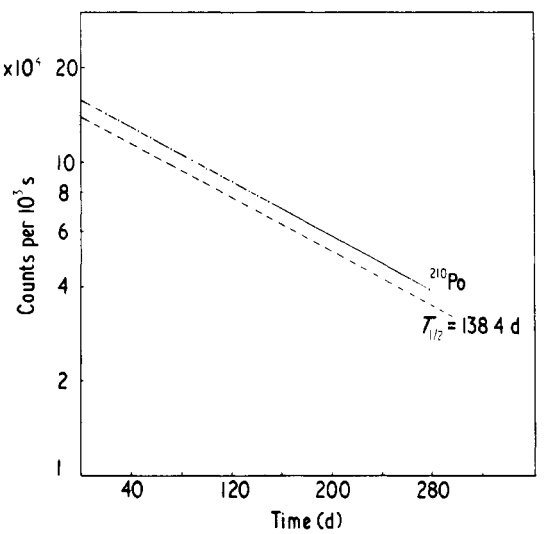

Figure 1. Decay of the ^{210}Po source used in this experiment.

4.3. X-ray counting

The L x-rays from the ^{210}Po and ^{241}Am sources were detected under the same geometrical conditions, the source holder in each case being mounted on a narrow ledge at the bottom of a hole drilled through a cylindrical block of perspex of the same diameter as the detector housing and taped to it. The distance from the source to the 0·1 mm thick Be window of the detector was then about 2 mm and the front face of the detector was (according to the manufacturer) a further 6 mm away. This close geometry arrangement was necessary because of the extremely low counting rate of the ^{210}Po source ($< 1\ s^{-1}$), which also required that the counting channel be stable over long periods of time. Calculations of the solid angle subtended by a point source and by the extended sources used in this work show that effects due to the finite size and small non-uniformity of the sources will be negligible, judging from the similarity of the sources as shown by the autoradiographs.

The electronics in the x-ray detection channel consisted of a pre-amplifier with opto-electronic feedback, amplifier, baseline restorer, pile-up rejector, biased amplifier and stretcher. The pulses were analysed in a computer-based pulse height analysis system consisting of an ADC interfaced to a mini-computer. There were no dead-time

corrections in the ^{210}Po counting but the neptunium L x-ray rate from the ^{241}Am source was much higher and, although the ADC gives compensation for dead time, the presence of the pile-up rejector complicates matters. The counting losses were therefore determined by feeding pulses from a pulse generator at a low rate (100 Hz) into the test input of the pre-amplifier and finding how many of these pulses were lost as sources were placed in front of the detector so as gradually to increase the dead time indicated on the meter of the ADC. It was found necessary to make a 3% increase in the Np L_α peak area.

5. Results

Figures 2, 3 and 4 show respectively the L x-ray spectrum of neptunium, the L spectrum of lead, and a detail of the L_α and L_β lines of lead obtained with the detector. The energies of the neptunium lines were used as a calibration and a linear least-squares fit using these (and the energies of the K_α and K_β lines of manganese from a ^{55}Fe source) showed that the x-rays emitted during the α decay of ^{210}Po are definitely the L x-rays of lead. Figure 3 shows the presence of a small amount of contamination due to ^{241}Am being present in the ^{210}Po stock solution. This was a result of cross-contamination during production of the stock solutions by the manufacturer but the amount of contamination is so small, estimated at about 4 dps in the 4 μCi ^{210}Po source used in the experiment, that it would not normally be noticed except in the course of the long counting times which had to be used. The detail of figure 4 shows that the contamination did not interfere with the calculation of the number of counts in the L_α and L_β peaks, nor did it, in fact, with the L_γ. Figures 3 and 4 show the spectrum obtained after a counting time of 10^6 s and the symmetrical shape of the L_α peak attests to the stability of the equipment.

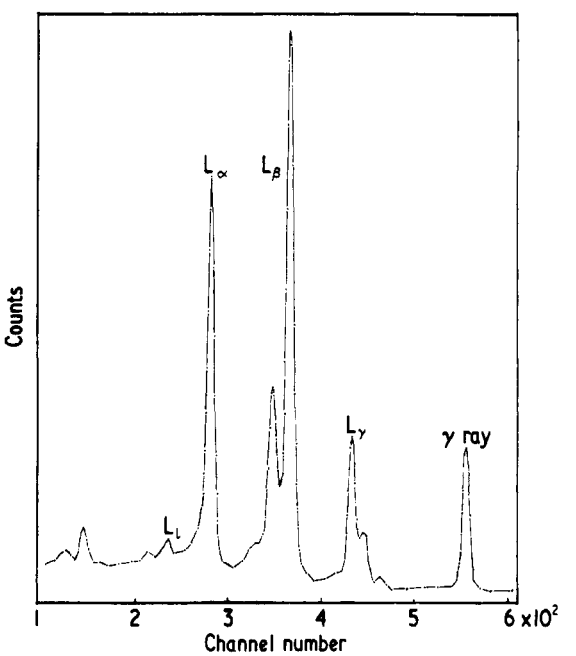

Figure 2. Np L x-rays from a source of ^{241}Am observed with the Ge(Li) detector. The peaks on the low energy side of the L_l peak are escape peaks.

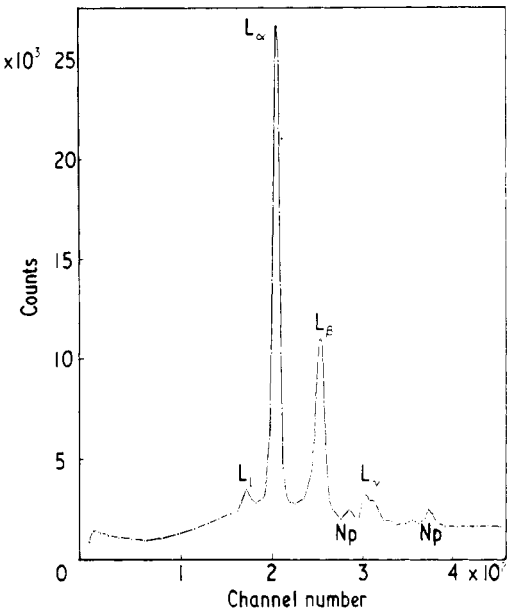

Figure 3. Pb L x-ray spectrum from the ^{210}Po source.

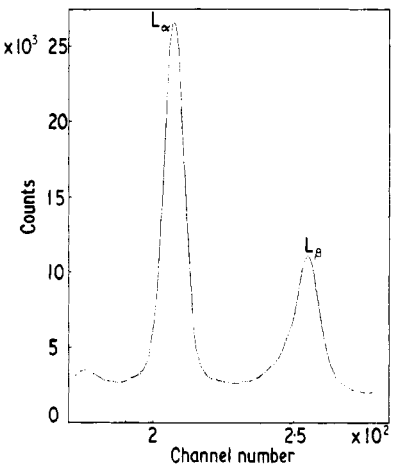

Figure 4. Detail of the Pb L_α and L_β lines.

The results of two experimental runs, one of 10^6 s and the other of $4{\cdot}9 \times 10^5$ s duration are presented in table 3, along with the values of E_{Np}/E_{Pb} derived from the K_β/K_α measurements, the quantity A_α/X_{Np} and the calculated lead L x-ray yields. The numbers C_α are the total number of α disintegrations during a run multiplied by the counting geometry factor, which of course cancels in the final expression for f given in § 4.1, and were calculated using the measured α disintegration rate at the beginning of each run.

The fact that the quantity A_α/X_{Np} is not the same for the two runs requires some explanation, and the reason is as follows. There was found to be a difference between the results of the two runs which was larger than would be expected from an analysis of the

Table 3. The results of two experimental runs: (*a*) 10^6 s duration; $C_\alpha = 1{\cdot}31 \times 10^8$; $A_\alpha/X_{Np} = 0{\cdot}53 \pm 0{\cdot}01$; and (*b*) $4{\cdot}9 \times 10^5$ s duration; $C_\alpha = 7{\cdot}09 \times 10^7$; $A_\alpha/X_{Np} = 0{\cdot}56 \pm 0{\cdot}01$

(*a*)			E_{Np}/E_{Pb}	(*b*)		
Line	Counts	Intensity		Line	Counts	Intensity
L_l	7700 ± 700	$0{\cdot}046 \times 10^{-4}$	$1{\cdot}1 \pm 0{\cdot}1$	L_l	3400 ± 600	$0{\cdot}040 \times 10^{-4}$
L_α	231500 ± 800	$0{\cdot}71 \times 10^{-4}$	$0{\cdot}56 \pm 0{\cdot}04$	L_α	105200 ± 500	$0{\cdot}63 \times 10^{-4}$
L_β	110600 ± 700	$0{\cdot}60 \times 10^{-4}$	$1{\cdot}0 \pm 0{\cdot}07$	L_β	51600 ± 500	$0{\cdot}55 \times 10^{-4}$
L_γ	20800 ± 500	$0{\cdot}113 \times 10^{-4}$	$1{\cdot}0 \pm 0{\cdot}07$	L_γ	9900 ± 400	$0{\cdot}105 \times 10^{-4}$

likely uncertainties and this was eventually traced to the fact that a rotation of the source holder in its own plane caused changes in the x-ray counting rate from the ^{241}Am source. Now the centre of the ^{241}Am source did not coincide with the centre of the source holder (nor did that of the ^{210} Po source, but both sources were off-centre by the same amount, about 1 mm) but such a change in counting rate could only occur if the detector was in some way asymmetrical. This was indeed the case, as was checked by tracing out the sensitive area of the detector using a 10 μCi source of ^{241}Am looking through a 1 mm hole pierced in a lead sheet. It was found that the most efficient part of the detector was definitely off-centre with respect to the Be window, and, not only that, almost half of the supposed sensitive area was essentially dead. This was probably a result of mis-alignment of the detector during manufacture.

As a final check that this effect could explain the discrepancy between the results of the two long runs, two further, but shorter, counts of $2{\cdot}5 \times 10^5$ s were performed with the ^{210}Po source deliberately orientated first in the position of expected high sensitivity and then in the position of low sensitivity. The results of these runs confirmed the above explanation, giving values of $0{\cdot}68 \times 10^{-4}$ and $0{\cdot}70 \times 10^{-4}$ for the lead L_α yield (when the appropriate values of A_α/X_{Np} were used) with uncertainties somewhat greater than those quoted in table 3 since these runs were performed almost two half-lives after the two long runs. A background run of $2{\cdot}4 \times 10^5$ s duration showed no trace of any lead L x-rays.

The uncertainties quoted in table 3 are derived, in the case of the integrated peak counts, on the basis of varying the method of allowing for the background under the peaks, the purely statistical uncertainty being small. The uncertainty in the efficiency ratios arises from that in the measured K_β/K_α ratios, and in the quantities A_α/X_{Np} from the spread in the measured values as a function of source orientation. A series of values of A_α/X_{Np} was found by doing numerous counts with the ^{241}Am source in different angular positions and these could be divided obviously into a set of high values, occurring when the source was furthest from the most sensitive part of the detector, and a set of low values. There was a 10% difference between the highest and lowest values found.

The final values adopted are the weighted means of the results of the two long runs, weighted 2:1 in favour of the longer:

L_l: $(0{\cdot}044 \pm 0{\cdot}01) \times 10^{-4}$

L_α: $(0{\cdot}68 \pm 0{\cdot}05) \times 10^{-4}$

L_β: $(0{\cdot}58 \pm 0{\cdot}05) \times 10^{-4}$

L_γ: $(0{\cdot}11 \pm 0{\cdot}01) \times 10^{-4}$

giving a total L x-ray yield of $(1{\cdot}41 \pm 0{\cdot}12) \times 10^{-4}$ per disintegration.

6. Conclusions

Reference to table 2 shows that the results presented here are much lower than those previously reported. The best of the early experiments is that of Rubinson and Bernstein (1952) in which proportional counters were used. Our results differ in detail as well as in total yield. The relative intensities of the components are given by Rubinson and Bernstein as

$$L_\alpha : L_\beta : L_\gamma = 1 : 0{\cdot}635 : 0{\cdot}12$$

whereas our result is

$$L_l : L_\alpha : L_\beta : L_\gamma = 0{\cdot}065 : 1 : 0{\cdot}85 : 0{\cdot}16.$$

It is difficult to see how the discrepancy in total yield can arise unless it is a result of the difference in source strengths employed, although the generation of, say, Po x-rays by α particles passing through the source, or by aggregate recoils, seems to be ruled out by the critical absorption experiments reported by Rubinson and Bernstein.

There is clearly a need for some more experimental work on this topic and for a theoretical treatment which does not rely on the adiabatic or sudden approximations.

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