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The Present Experimental and Theoretical Status of the Problem  
of Electron Ejection in the Alpha Decay of  $\text{Po}^{210}$  \*

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(Abstract)

MASTER

The experimental measurements of electron ejection in  $\text{Po}^{210}$  decay are discussed, and the theory of the process is outlined. The order-of-magnitude discrepancy between theory and experiment which was evident a decade ago has not yet been definitively resolved. The discrepancy is ascribed to an inadequacy of the theory, in particular to the use of an asymptotic expansion in that theory. Brief mention is made of some very recent unpublished calculations by G. W. Schaefer in which a reasonable estimate of the K ejection probability is obtained by a procedure that avoids the asymptotic expansion.

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The picture of the process we are dealing with is the following. The alpha particle emitted from the nucleus in the alpha decay act moves adiabatically through the cortège of electrons around the nucleus, subjecting these electrons to a time-dependent electrostatic force that is small compared to the force from the daughter nucleus. The interaction between the slowly moving alpha particle and a rapidly moving bound electron excites the latter to a higher bound state or to a continuum state, leaving a hole in one of the electron shells around the daughter nucleus. The subsequent filling of this hole by an electron from a higher shell thus takes place in the field of the daughter nucleus, and so results in the emission of an x-ray (or an Auger electron) characteristic of the daughter atom.

Measurement of the rate of emission of these x-ray photons from a source of known alpha decay rate gives the "photon yield" (i.e., the average number of photons per alpha decay) which can then be compared with the theoretical photon yield, the latter being the theoretical probability of electron ejection from a given shell, multiplied by a known empirical fluorescence yield.

The reason for continued interest in this apparently straightforward problem is that the published theories underestimate the experimental photon yields by one or two orders of magnitude, in surprising contrast to the state of affairs in the corresponding problems of electron ejection in beta decay, and of electron ejection by bombardment with a beam of heavy particles, where there is fair agreement between experiment and theories that make use of approximations similar to those used in the case of alpha decay.

All the studies on electron ejection in alpha decay have been made with  $\text{Po}^{210}$  (138.4 day half life; 5.3 MeV decay energy), which is by far the most convenient nuclide for such studies, and the one least subject to misinterpretation of the experimental results.

An order-of-magnitude discrepancy between theory and experiment in the case of electron ejection in  $\text{Po}^{210}$  decay was evident at the time of the last International Conference on the role of electrons in radioactive decay,<sup>1</sup> in 1954. At that time, the L x-rays<sup>2,3</sup> and K x-rays<sup>3,4,5</sup> from  $\text{Po}^{210}$  had been identified positively as characteristic Pb radiations, their photon yields had been measured,<sup>6</sup> and their connection with electron ejection in the alpha decay act had been recognized.<sup>2,3,5</sup> The theories available at that time were Migdal's original theory<sup>7</sup> and Levinger's<sup>8</sup> formally minor but physically important modification of that theory to take account of nuclear recoil, which Migdal had neglected. Migdal's theory gave a reasonable estimate of the K photon yield, but underestimated the L photon yield by a factor of  $\sim 30$ . The estimates on Levinger's more realistic theory were an order of magnitude lower than those on Migdal's theory.

It was not clear at that time whether the large discrepancy between theory and experiment was to be attributed to an inadequacy of the theory or to a misinterpretation of the experimental results. The few papers that have been published in the intervening nine years have hardly settled the problem in a definitive way, but they have made it quite clear that the cause of the discrepancy lies in the theory. And in some recent unpublished work (described briefly in a recent publication<sup>9</sup>), G. W. Schaefer<sup>10</sup> seems to have located the source of the theoretical difficulty, as will be mentioned below.

On the experimental side the situation is now quite satisfactory:

1. Each of the K, L, and M photon yields in  $\text{Po}^{210}$  decay has been measured in at least two independent investigations with different techniques and widely different source strengths, with results that agree within the claimed experimental errors of  $\pm 15\%$  to  $\pm 33\%$ . These results are listed in Table I.

Table I

Measured Pb x-ray Photon Yields in  $\text{Po}^{210}$  Decay

X-Ray	Photon Yield	Claimed Error	Reference
K	$1.5 \times 10^{-6}$	+33%	a
	2.00	$\pm 19\%$	b
	1.6	$\pm 31\%$	c
	1.5	$\pm 27\%$	d
L	$2.2 \times 10^{-4}$	$\pm 23\%$	c
	2.93	$\pm 15\%$	e
	4	....	f
M	$1.5 \times 10^{-3}$	....	f
	0.91	$\pm 15\%$	g

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2. The identification of the K and L x-rays as characteristic x-radiations of Pb, with negligible admixture of other radiations, has been established by critical absorption studies on the K spectrum,<sup>3,4</sup> and by critical absorption of the L  $\beta$  component of the L spectrum.<sup>2,3</sup> It is not possible to characterize the M rays by critical absorption, but their identity with pure Pb M rays has been established by comparison with known Pb M rays, and by establishing the absence of possible sources of contaminating radiations.<sup>9</sup> It has been shown in this latter work that the relative intensities of the Pb M component lines from  $\text{Po}^{210}$  decay differ appreciably from those observed in a Pb M spectrum excited by bombarding Pb with  $\text{Po}^{210}$  alpha particles. This has led to the inference that the  $\text{Po}^{210}$  decay act may result in so extensive a stripping of the outermost electrons from the atom that the M x-ray fluorescence yield may be considerably enhanced, in which case the theoretical M photon yield would need to be correspondingly increased, perhaps by 50%. A measurement of this fluorescence yield is now under way.

3. There no longer seems any possibility that, as has been suggested, the discrepancy between the measured and theoretical K photon yields may be attributed to internal conversion of the 0.8 MeV  $\text{Po}^{210}$   $\gamma$ , the intensity of which is<sup>11</sup>  $1.22 (\pm 0.06) \times 10^{-5}$  per alpha. To account for the discrepancy the internal conversion coefficient of this gamma would need to be greater than 10%. But  $\alpha$ - $\gamma$  angular correlation measurements<sup>12</sup> have shown that this gamma is probably E2, in which case the theoretical internal conversion coefficient is<sup>13</sup> 0.9%, i.e., less than one-tenth the value needed to account for the discrepancy. The case for a 1% internal conversion coefficient has been much strengthened by a subsequent direct experimental measurement<sup>14</sup> of the ratio of internal conversion electrons to  $\text{Po}^{210}$   $\gamma$ 's, the reported value being  $1(\pm 0.3)\%$ . There remains another possibility proposed by some, namely, that hitherto undetected gammas are converted in the different shells. Such

gammas have been sought<sup>4,9</sup> in the energy range 5 keV to 2.5 MeV, and not found, the limit of detection at a given energy being one-tenth the intensity of the closest known x- or  $\gamma$  ray of  $\text{Po}^{210}$ . Consequently, if the observed x-rays are to arise almost exclusively from the internal conversion of undetected gammas, such gammas must have internal conversion coefficients greater than 10, and thus (as seen from a table of internal conversion coefficients<sup>15</sup>) be of low energy and of higher order electric or magnetic multipolarity. In principle such internal conversion could be detected by the presence of low energy lines in the electron spectrum from a  $\text{Po}^{210}$  source, an experiment that would be formidably difficult in practice. Nevertheless, though internal conversion of undetected gammas (or other imaginable processes) has not been completely eliminated as a possibility, we reject this possibility as highly improbable and attribute the discrepancy between theory and experiment to an inadequacy of the theory.

We outline a recent version<sup>9</sup> of the theory that follows a formulation of the problem due to Schwartz<sup>16</sup> and Grard.<sup>17</sup> Nuclear recoil is introduced in a way alternative to Levinger's by means of a coordinate system in which at times  $t < 0$  the  $\text{Po}^{210}$  nucleus bearing its electron cortège moves along the negative z-axis in the positive z direction with velocity  $u$ , the negative of the recoil velocity. When the moving  $\text{Po}^{210}$  nucleus reaches the origin, at  $t = 0$ , a one-electron state in the  $\text{Po}^{210}$  atom is (we use atomic units)  $e^{iuz} \psi(x, y, z) \rangle$ , where  $\psi(x, y, z) \rangle$  is the corresponding one-electron eigenstate in the field of a  $\text{Po}^{210}$  nucleus fixed at the origin. At  $t = 0$ , the instant the origin is reached, the  $\text{Po}^{210}$  nucleus decays, the  $\text{Pb}^{206}$  daughter nucleus recoiling to a dead stop at the origin, while the emitted alpha particle, which has instantaneously acquired its full terminal velocity  $v$ , moves off along the positive z-axis, interacting with the electron at times  $t \geq 0$  according to the time-dependent potential

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

Let  $E$  be the Hamiltonian of an electron in the field of the  $\text{Pb}$  nucleus of nuclear charge  $Z$  fixed at the origin. The complete set of eigenstates  $|E'\rangle$  of  $E$ , belonging to energy eigenvalues  $E'$ , will constitute our representation for the problem. Designate by  $|E^0\rangle$  the  $\text{Pb}$  eigenstate corresponding

to the Po eigenstate  $\Psi(x, y, z) \rangle$ , and define the first order term  $|1\rangle$  by

$$\Psi \rangle = |E^0 \rangle + |1\rangle$$

Then the initial electron state, i.e., the state at the instant of decay, can be written

$$e^{iuz} \Psi \rangle = (1 + iuz)(|E^0 \rangle + |1\rangle) = |E^0 \rangle + |1\rangle + iuz|E^0 \rangle + \text{higher order terms.}$$

The probability that an electron in this state at  $t = 0$  will be found in state  $|E' \rangle \neq |E^0 \rangle$  at  $t = \infty$  is  $|w(E')|^2$ , where, by first order time-dependent perturbation theory,

$$(2) \quad w(E') = \langle E' | 1 \rangle + iu \langle E' | z | E^0 \rangle - i \int_0^\infty e^{i(E'-E^0)t} \langle E' | V(t) | E^0 \rangle dt + \text{higher order terms,}$$

where  $V(t)$  is given by (1).

Following Migdal, we expand the integral in (2) by successive integrations by parts to obtain an expression that can be manipulated into the form (neglecting higher order terms)

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

with

$$(4) \quad R = -2 \sum_{k=2}^N i^k k! \frac{v^k}{(E' - E^0)^{k+1}} \langle E' | \frac{P_k(z/r)}{r^{k+1}} | E^0 \rangle + \text{remainder,}$$

where  $r$  is the radial variable and  $P_k(z/r)$  is the normalized  $k$ th Legendre polynomial. The  $\langle E' | 1 \rangle$  term of eq. (2) does not appear in eq. (3) because it is cancelled by a term resulting from the first of the integrations by parts (as can be seen from the value of  $\langle E' | 1 \rangle$  computed by time-independent perturbation theory).

From the form of eq. (4) it is seen that our expansion of the integral in eq. (2) is an asymptotic expansion,  $R$  being a sum of all terms after the first two. Despite the fact that  $R$  diverges as  $N \rightarrow \infty$ , we discard it in eq. (3), the justification being that usually one obtains reasonable answers by retaining only the first terms of an asymptotic expansion. In the present case this may well be a bad mistake, as will be seen below.

The matrix element of  $-z/r^3$  in eq. (3) can be simplified by the observation that the quantum mechanical analogue of the classical equation of motion  $\ddot{z} = -Zz/r^3$  ( $Z$  is the charge on the Pb nucleus) gives

$$(5) \quad \langle E' | -z/r^3 | E^0 \rangle = \frac{1}{Z} \langle E' | \ddot{z} | E^0 \rangle = \frac{1}{Z} \langle E' | i[z, E] , E | E^0 \rangle$$

$$= -\frac{(E' - E^0)^2}{Z} \langle E' | z | E^0 \rangle$$

where  $E$  is the Hamiltonian of an electron in the field of  $Z$ , and we have used the convention  $i[a, b] = ab - ba$ .

Substitution of eq. (5) into eq. (3) with  $R$  discarded leads to

$$(6) \quad |w(E')|^2 = (u - 2v/Z)^2 |\langle E' | z | E^0 \rangle|^2 \quad (E' \neq E^0)$$

The total probability of electron ejection in the alpha decay act, i.e., the probability that an electron in state  $|\psi\rangle$  of the Po atom before decay is not in the corresponding state  $|E^0\rangle$  of the Pb atom after decay, is the sum of expression (6) over all final bound and continuum states  $E' \neq E^0$  that are permitted by the selection rules and are not occupied by other electrons. In (6) the sum over the squares of dipole moment matrix elements is readily evaluated from available tabulations of the corresponding values for hydrogen; the magnitudes of  $u$  and  $v$  are, respectively, 0.14 and 7.32 atomic units of velocity, as computed from the 5.3 MeV decay energy of  $\text{Po}^{210}$ ; and the  $Z$ 's are effective nuclear charges computed in a conventional way by means

of screening constants. The x-ray fluorescence yields by which these theoretical electron ejection probabilities must be multiplied to give the theoretical photon yields are available from the literature. Comparison of the theoretical photon yields so obtained with the experimentally measured values reveals the following<sup>9</sup>:

Photons	<u>Experiment</u> <u>Theory</u>
K	13.5
L	270
M	410

These discrepancies are an order of magnitude larger than those obtained with Migdal's original theory which differs essentially from the one outlined above only by the assumption that  $u = 0$ , instead of 0.14 atomic units; for  $Z$ 's corresponding to the Pb nuclear charge, the value of  $(u - 2v/Z)^2$  is about one-tenth the value of  $(2v/Z)^2$ . In fact, it results from eq. (6) that if  $2/Z = u/v$  (by conservation of momentum this would be the case if the charge-to-mass ratio of the alpha particle were equal to the ratio of  $Z$  to the  $Po^{210}$  mass), then the probability of electron ejection is zero, which does not seem physically reasonable. We remark parenthetically that the separate probabilities of K ejection by the alpha particle alone (i.e., assuming  $u = 0$ ) and by recoil alone (i.e., assuming the alpha charge is zero) are both accidentally in fair agreement with the measured K ejection probability, though both underestimate the L and M ejection probabilities by an order of magnitude.

At the present time it appears that the most likely source of the inadequacy of the theory lies in the procedure of successive integrations by parts applied to the integral in eq. (2) to obtain the asymptotic expression

in eq. (3). The first clue to this is Levinger's observation that if the  $k = 2$  term of eq. (4) is retained in the calculations, then the L and, presumably, the M photon yields are increased by an order of magnitude (the K photon yield is not affected). If recourse to the asymptotic expansion is avoided altogether, there is apparently a gratifying improvement in the theory as shown by Schaefer's unpublished calculations (referred to above) of the K ejection probability. If Schaefer proves to be equally successful in calculating the L and M ejection probabilities, then this long-standing problem can be considered to be solved.

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