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# Effects of Radioactive Disintegrations on Inner Electrons of the Atom

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The probability that nuclear emission of an alpha- or beta-particle causes ionization of a $K$ or $L$ electron of the atom is calculated by time-dependent perturbation theory using nonrelativistic Coulomb wave functions. Beta-emission (electron or positron) causes an ionization probability of $0.64/Z^2$ and $2.1/Z^2$ per beta in the $K$ and $L$ shells, respectively. (The $K$ shell result agrees with Migdal and Feinberg; the $L$ shell result disagrees with Migdal.) The use of nonrelativistic wave functions causes an appreciable underestimate in the ionization probability for $K$ electrons of heavy atoms. Screening corrections for the use of Coulomb wave functions would increase the ionization probabilities by a factor of 1.4 for $K$ electrons and by a factor of 3 or 4 for $L$ electrons. Migdal's result for dipole electronic transitions caused by nuclear alpha-decay are reduced by a factor 25 (for the case of $Po^{210}$) because of nuclear recoil. Quadrupole matrix elements such as $(r^{-3})_{1s,\,n'd}$ are evaluated by a new method developed by H. A. Bethe. This method uses the Sommerfeld integral representation for the continuum $n'd$ wave function. Quadrupole transitions are negligible for $K$ electrons, but are the predominant effect for $L$ electrons. The calculated ionization probabilities for $Po^{210}$ are $10^{-7}$ and $1.1\times10^{-4}$ per alpha in the $K$ and $L$ shells, respectively. For alpha-decay, screening corrections and higher multipole transitions would both increase the ionization probability for $L$ electrons. Madansky and Rasetti's measurements of photons from RaE are consistent with our calculations, but Bruner's measurements on $Sc^{44}$ are not. Grace's interpretation of $K$ x-rays from $Po^{210}$ is consistent with the calculation of this paper, while Barber and Helm's interpretation is not. Rubinson and Bernstein find 8 times the $L$ x-ray yield from $Po^{210}$ we have calculated for Coulomb wave functions.

## I. INTRODUCTION

WHEN a radioactive nucleus disintegrates by emission of an alpha- or beta-particle, the radioactive disintegration perturbs the electrons of that atom, and may cause electronic excitation to an unoccupied discrete level, or ionization to the continuum. A hole in an inner electronic shell produced in this manner will be filled either by the emission of a characteristic x-rays, or by the emission of an Auger electron. Similar processes may be caused by other types of nuclear disintegrations: $K$-capture, neutron emission (or nuclear recoil due to neutron scattering), or fission. The probability of electronic excitation or ionization can be calculated by time-dependent perturbation theory. In this paper we shall calculate the probability of ionization for $K$ or $L$ electrons, for the cases of nuclear beta-decay (electrons or positrons) and nuclear alpha-decay.

We shall not concern ourselves here with calculations of nuclear decay processes involving interaction with the electromagnetic field, such as internal conversion or emission of inner bremsstrahlung. Frequently internal conversion masks the processes that we wish to discuss in this paper, since it may occur with very much higher probability. The emission of inner bremsstrahlung makes more difficult the detection of characteristic x-rays produced in beta-decay.

Recently Madansky and Rasetti[1] and Novey[2] have studied electromagnetic radiation emitted in $P^{32}$ and in RaE beta-decay. The characteristic sulfur x-ray following $P^{32}$ beta-decay is of too low an energy to be detected in this experiment. The RaE measurements show that characteristic $K$ x-rays of Po are less frequent than inner bremsstrahlung: the upper limit for $K$ x-rays is about one photon per $10^4$ betas. However, Bruner[3] finds a very much larger yield of negative electrons associated with the positron and $K$-capture activity of $Sc^{44}$, in contradiction to the very low negative electron yield found by Porter and Hotz[4] for $Fe^{55}$ ($K$-capture).

Characteristic lead $K$, $L$, and $M$ x-rays associated with the alpha-decay of $Po^{210}$ have been studied by Curie and Joliot,[5] Rubinson and Bernstein,[6] Grace *et al.*,[7] Barber and Helm,[8] and Riou.[9] The observed yields of photons per alpha are: about $10^{-6}$ for the $K$ shell,[7-9] about $3\times10^{-4}$ for the $L$ shell,[5,6,9] and considerably higher for the $M$ shell.[5] On the other hand, Macklin and Knight[10] found a very much higher yield of $L$ x-rays associated with the alpha-decay of $U^{234}$.

Calculations of the probability of ionization of $K$, $L$, and $M$ electrons in beta- and alpha-decay were first made by Migdal.[11] Migdal treats beta-decay as an example of a sudden perturbation, and alpha-decay as an example of an adiabatic perturbation. Independently Feinberg[12] calculated the effect of beta-decay on $K$ electrons obtaining agreement with Migdal's results based on the sudden change of the nuclear charge. The present author also considered these questions independently some years later,[13] and agreed with the common Migdal-Feinberg result for ionization of $K$ electrons in beta-decay; but there were numerical

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[1] L. Madansky and F. Rasetti, Phys. Rev. **83**, 187 (1951); and Phys. Rev. **89**, 679 (1953).
[2] T. B. Novey, Phys. Rev. **86**, 619 (1952) and Phys. Rev. **89**, 672 (1953).
[3] J. A. Bruner, Phys. Rev. **84**, 282 (1951).
[4] F. T. Porter and H. P. Hotz, Phys. Rev. **89**, 903A, 1953.
[5] I. Curie and F. Joliot, J. phys. et radium **2**, 20 (1931).
[6] W. Rubinson and W. Bernstein, Phys. Rev. **86**, 545 (1952).
[7] Grace, Allen, West, and Halban, Proc. Phys. Soc. (London) **A64**, 493 (1951).
[8] W. C. Barber and R. H. Helm, Phys. Rev. **86**, 275 (1952).
[9] M. Riou, J. phys. et radium **13**, 244 (1952).
[10] R. L. Macklin and G. B. Knight, Phys. Rev. **72**, 435 (1947).
[11] A. Migdal, J. Phys. (U.S.S.R.) **4**, 449 (1941).
[12] E. L. Feinberg, J. Phys. (U.S.S.R.) **4**, 424 (1941).
[13] J. S. Levinger, Ph.D. thesis, Cornell University, 1948 (unpublished).

disagreements for the $L$ shell. The ionization probability due to beta-decay has been considered recently by Schwartz,[14] Winther,[15] and Serber and Snyder.[16] Schwartz's numerical results for the $L$ shell ionization probability agree with those given in this paper; he has also extended the calculations to the $M$ shell. Winther calculates the charge distribution of $Li^6$ recoils from $He^6$ beta-decay, using helium and lithium atomic wave functions. His results are the same order of magnitude as those of this paper, which apply to hydrogen-like wave functions for inner electron of heavier atoms. Serber and Snyder calculate the average energy expended in ionizing the atom as the change in the electrostatic energy of the nucleus at the center of the electronic charge cloud. Their result of about 100 ev for positron or beta-decay in heavy atoms is larger than the energy expended in ionizing $K$ and $L$ electrons as calculated in this paper. We use their result of excitation energy 2 Rydbergs (27.2 ev) per closed shell to check our calculations of the ionization and excitation for the $K$ shell. Recently Primakoff and Porter[17] have calculated the electron ionization due to $K$ capture.

There was a basic diagreement between the methods used by Migdal and by the present author[13] (the latter being incorrect) in the calculations on effects of alpha-decay. Our present methods for alpha-decay represent a modification and extension of Migdal's work. The dipole transition probability is greatly reduced due to nuclear recoil. The quadrupole transition probability is of great significance for the $L$ shell. Our theoretical results are in order of magnitude agreement with the experimental yields of x-rays.

In Sec. II we discuss the case where the perturbation due to the nuclear decay takes place in a time small compared to the periods of the atomic electrons considered. This is represented in nature by beta-decay. In Sec. III we consider screening corrections to the results of Sec. II, which are based on hydrogenic wave functions. In Sec. IV we discuss the case where the perturbation takes place slowly, i.e., adiabatically, as compared to the periods of the atomic electrons being considered. This case is represented in nature by the alpha-decay of Po, if we consider the $K$ or $L$ electrons. Quadrupole matrix elements such as $(r^{-3})_{1s,\,n'd}$ are calculated by a new method developed by H. A. Bethe. In Sec. V we estimate the accuracy of our approximations for alpha-decay. In the last section we shall compare our calculations with experimental data.

## II. SUDDEN PERTURBATION: BETA-DECAY

When a nucleus of atomic number $Z$ decays by emission of a beta-particle, which has a velocity very much larger than that associated with the orbital electrons, the orbital electrons suddenly find themselves in the field of a nucleus of charge $Z+1$. (For positron decay the change is from $Z$ to $Z-1$.) To simplify our formulas we shall limit ourselves to the case of $Z$ much larger than one, in which case beta and positron emission give very nearly the same result, and will be considered together. The amplitude for an orbital electron initially in state 0 to end in state $n$ due to a sudden perturbation is

$$a_{0n}=\int \psi_n^*(Z+1,r)\psi_0(Z,r)d^3r. \tag{1}$$

Here we explicitly write the atomic number of the nucleus to designate the wave functions before and after the beta-decay.

This formulation was first proposed by Migdal.[11] Feinberg[12] discusses the effects of beta-decay on the orbital electrons in some detail and finds that the "shaking term" corresponding to Eq. (1), is in general much larger than the term for direct collisions. (The latter is analogous to the ionization of an atom by an external fast electron.) He also shows that relativistic effects and exchange effects are small. We shall discuss the approximations in Sec. V; in this section we shall evaluate Eq. (1) using nonrelativistic wave functions for a Coulomb field. In this evaluation we shall repeat some of Migdal's and Feinberg's work, as their papers are not sufficiently well known.

First, let us find an upper limit for the sum of the probability of excitation to all discrete states, and of ionization to the continuum. We shall find the probability $P_{00}=|a_{00}|^2$ that the electron remain in the initial state. The difference $1-P_{00}$ represents the probability of excitation to *all* discrete states, together with ionization to continuum states. It therefore overestimates the probability of losing an electron from state 0, since some transitions to discrete states are forbidden by the Pauli exclusion principle. Still, it gives us an upper limit which is the right order of magnitude. Also it is the starting point for our calculations of screening effects.

For a $K$ electron we have, using hydrogenic wave functions,

$$a_{00}=4Z^{\frac{3}{2}}(Z+1)^{\frac{3}{2}}\int_0^\infty \exp(-Zr)\exp[-(Z+1)r]r^2dr$$
$$=\{[(Z+\tfrac{1}{2})^2-\tfrac{1}{4}]/(Z+\tfrac{1}{2})^2\}^{\frac{3}{2}}\approx 1-3/8Z^2. \tag{2}$$

(The angular integrations give unity. Throughout this paper we use Hartree atomic units,[18] e.g., we measure distances in units of the Bohr radius.) The upper limit for loss of an electron from the $1s$ state is

$$1-|a_{00}|^2=3/4Z^2. \tag{3}$$

We have neglected unity in comparison with $Z$, so Eq. (3) holds for either beta or positron emission. To find

[14] H. M. Schwartz, J. Chem. Phys. **21**, 45 (1953).
[15] A. Winther, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, 3 (1952).
[16] R. Serber and H. S. Snyder, Phys. Rev. **87**, 152 (1952).
[17] H. Primakoff and E. T. Porter, Phys. Rev. **89**, 903A (1953).
[18] H. A. Bethe, *Handbuch der Physik* (J. Springer, Berlin, 1933), Vol. 24/1, Introduction.

the probability that a hole is produced in the $K$ shell, one considers the perturbation acting separately on each of the two $K$ electrons, and multiplies the above result by 2. (See Feinberg for a detailed justification.)

We have made analogous calculations for the $2s$ and $2p$ electrons of the $L$ shell: we find an upper limit of $2.25/Z^2$ per $2s$ electron, and $1.25/Z^2$ per $2p$ electron.

Let us now calculate the probability that a $K$ electron end up in a continuum state. This will be a lower limit for the production of holes in the $K$ state; and should be a good approximation to it, as the more likely transitions to discrete states are forbidden by the Pauli principle. This calculation can be done in several different equivalent ways; we shall follow the method used by Feinberg.[12] We use an integral representation for the hydrogenic continuum wave functions[19] for an $s$ electron (the angular integrations give the selection rule $\Delta l=0$):

$$R_W=-2Z^{\frac{1}{2}}(2kr)^{-1}(1-e^{-2\pi n'})^{-\frac{1}{2}}(2\pi)^{-1} \times \oint e^{-2ikr\xi}(\xi+\tfrac{1}{2})^{-in'-1}(\xi-\tfrac{1}{2})^{in'-1}d\xi. \quad (4)$$

The integral over $\xi$ is taken around a contour that encloses the branch points at $\xi=\frac{1}{2}$ and $\xi=-\frac{1}{2}$.

This wave function is normalized per unit energy. Here

$$n'=Z/k=(E_K/W)^{\frac{1}{2}}. \quad (5)$$

The binding energy of a $K$ electron is, in atomic units ($e^2/a_0=27.2$ ev),

$$E_K=\tfrac{1}{2}Z^2. \quad (6)$$

The positive energy $W$ of the continuum electron is given by

$$W=\tfrac{1}{2}k^2=\tfrac{1}{2}Z^2/n'^2, \quad (7)$$

where $k$ is the wave number.

We use the continuum wave function of Eq. (4) in Eq. (1) for the transition amplitude. [For convenience we use $\psi_0(Z+1,r)$, and $\psi_n{}^*(Z,r)$.]

$$a_{0n'}=NJ, \quad (8)$$

$$N=4Z(1-e^{-2\pi n'})^{-\frac{1}{2}}, \quad (9)$$

$$J=2\pi^{-1}\int\oint e^{-(Z+1)r}e^{-2ikr\xi}(2kr)^{-1} \times(\xi+\tfrac{1}{2})^{-in'-1}(\xi-\tfrac{1}{2})^{in'-1}d\xi r^2dr. \quad (10)$$

The double integral $J$ is evaluated as was done by Sommerfeld in calculations of the atomic photoeffect,[20] i.e., we first perform the $r$ integration,

$$\int_0^\infty e^{-\beta r}rdr=1/\beta^2, \quad (11)$$

where

$$\beta=2ik(\xi-in'/2-i/2k). \quad (12)$$

[19] Reference 18, Eq. (4.22). Note that we have corrected a misprint by multiplying by $2^{\frac{1}{2}}$. See Feinberg, reference 12.

[20] Reference 18, Sec. 47; also see reference 12.

The contour for the $\xi$-integration is transformed to infinity enclosing the pole at $\beta=0$. The result is $2\pi$ times the residue at the double pole at $\beta=0$, or $\xi=in'/2+i/2k$.

$$J=(2k)^{-3}\{(\xi^2-\tfrac{1}{4})^{-2}[(\xi-\tfrac{1}{2})/(\xi+\tfrac{1}{2})]^{in'} \times(in'-2\xi)\}_{\xi=in'/2+i/2k} = 2k^{-4}(n'^2+1)^{-2}\exp(-2n'\cot^{-1}n'). \quad (13)$$

(Note that we check the orthogonality of $\psi_0(Z)$ and $\psi_{n'}{}^*(Z)$ if we put $\xi=in'/2$ in the $(in'-2\xi)$ term. However, we can use this approximation for $\xi$ in the other terms, and we shall also put $Z+1=Z$ in the term $N$.) We combine this value for $J$ with the normalization factor of Eq. (9), and square the amplitude, in order to find the transition probability in terms of the energy $W$ of the continuum electron:

$$P(n')dW=2^6Z^{-4}(1-e^{-2\pi n'})^{-1}n'^8(n'^2+1)^{-4} \times\exp(-4n'\cot^{-1}n')dW, \quad (14)$$

where the variable $n'$ is given by Eq. (5). The emitted electron is likely to have a value around $n'=1$, or continuum energy $W$=binding energy $E_K$, as is shown in Fig. 1 where we plot $P(W)$ *vs* $W$. The probability of ionization to all continuum states is found by numerical integration of Eq. (14), using $dW=-Z^2dn'/n'^3$ [see Eq (7)]. We find

$$P=\int_0^\infty P(n')dW=0.32/Z^2. \quad (15)$$

The probability of ionization as a function of $n'$, and its integral for the ionization probability per $K$ electron per $\beta$-decay, is in agreement with the calculations of Migdal and of Feinberg.

The calculations for ionization of $L_{\text{I}}$ electrons and $L_{\text{II}}$ or $L_{\text{III}}$ electrons is performed in an analogous manner. For $L_{\text{I}}$ electrons the amplitude is

$$(a_{0n'})_1=N_1J_1, \quad (16)$$

$$N_1=2^{\frac{3}{2}}Z^2(1-e^{-2\pi n'})^{-\frac{1}{2}}, \quad (17)$$

$$J_1=(2\pi)^{-1}(2k)^{-1}\oint\int e^{-\gamma r}[1-\tfrac{1}{2}(Z+1)r]rdr \times(\xi+\tfrac{1}{2})^{-in'-1}(\xi-\tfrac{1}{2})^{in'-1}d\xi, \quad (18)$$

$$\gamma=2ik(\xi-in/4-i/4k). \quad (19)$$

The $r$ integration gives a double pole, and a triple pole; the $\xi$-integration is done by evaluating the residues at these poles. We find for the probability of the transition $2s$ to continuum:

$$P_1(n')dW=2^{11}Z^{-4}(1-e^{-2\pi n'})^{-1}n'^8(3n'^2+4)^2(n'^2+4)^{-6} \times\exp[-4n'\cot^{-1}(\tfrac{1}{2}n')]dW. \quad (20)$$

The ionization probability of $0.47/Z^2$ is found from Eq. (20) by numerical integration.

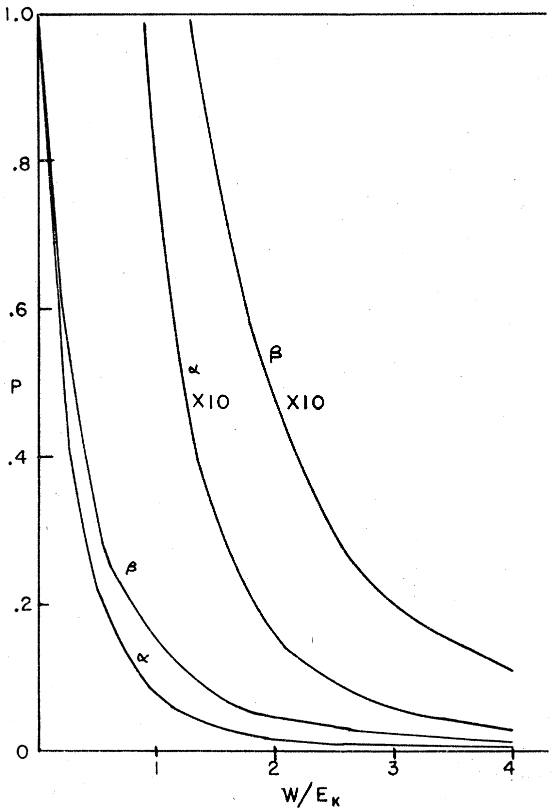


FIG. 1. Calculated energy spectrum for $K$ electrons ionized by beta- or alpha-decay [see Eq. (14) and Eq. (45)]. $W$ is the positive energy of the emitted electron; $E_K$ is the $K$ binding energy. The vertical scale is arbitrary.

For $L_{II}$ or $L_{III}$ electrons the amplitude for a transition to the continuum is

$$(a_{0n'})_2=N_2J_2, \qquad (21)$$

$$N_2=Z^3 6^{-\frac{1}{2}}(1-e^{-2\pi n'})^{-\frac{1}{2}}(1+n'^2)^{\frac{1}{2}}, \qquad (22)$$

$$J_2=(2\pi)^{-1}(2k)^{-2}\oint\int e^{-\gamma r}rdr(\xi+\tfrac{1}{2})^{-in'-2}(\xi-\tfrac{1}{2})^{in'-2}d\xi. \qquad (23)$$

[Note that here we use a $p$ wave function for the continuum electron. $\gamma$ is defined by Eq. (19).] The probability for a transition from $L_{II}$ or $L_{III}$ to continuum is

$$P_2(n')dW=2^{15}3^{-1}Z^{-4}(1-e^{-2\pi n'})^{-1}n'^{10}(n'^2+1) \times(n'^2+4)^{-6}\exp[-4n'\cot^{-1}(\tfrac{1}{2}n')]dW. \qquad (24)$$

The ionization probability of $0.21/Z^2$ is found from Eq. (24) by numerical integration.

The results of our calculations for ionization probability by beta or positron decay are collected in Table I where we have neglected unity in comparison with $Z$. As noted above, we agree with Migdal and Feinberg for the $K$ electron but obtain smaller values than Migdal's for the ionization probability of $L$ electrons. In our thesis[13] we obtained a different result for the $L_I$ electrons. Schwartz's results[14] are in agreement with our present results and extend the calculation to the $M$ shell.

Another type of sudden perturbation is the sudden change of the nuclear velocity: the nucleus suddenly recoils with velocity $V$ in the $z$ direction, owing to neutron emission, or owing to elastic scattering of a neutron. (There is of course a nuclear recoil in beta-decay; but we show below that its effect on the inner electrons is much less than the effect of the nuclear charge change. The nuclear recoil in the case of alpha-decay is considered in the next section.)

Initially the electron has a wave function $u_0$. After the nucleus recoils, an electron with wave function $u_{n'}$ relative to the nucleus has a wave function $e^{iKz}u_{n'}$ in the laboratory system, where $K=mV/\hbar$ is the wave number of an electron with the nuclear recoil velocity $V$. The amplitude for a transition from state 0 to state $n'$ is

$$a_{0n'}=\int u_{n'}{}^*e^{-iKz}u_0d^3r=-\int u_{n'}{}^*iKzu_0d^3r, \qquad (25)$$

where in the last expression we have expanded the exponential for $K$ small.[21]

The probability $P_{0n'}$ for a transition is proportional to the square of the dipole moment. This quantity has already been summed over all continuum states.[22] Using these sums, and the appropriate factors for the angular integrations[23] ($\frac{1}{3}$ for transitions from $s$ states to $p$ states), we have for the probabilities summed over all continuum states:

$$\begin{aligned} &P_{1s}=0.28K^2/Z^2, \quad P_{2s}=0.90K^2/Z^2,\\ &P_{2p}=0.69K^2/Z^2, \quad \text{for } m=0, \\ &P_{2p}=0.48K^2/Z^2, \quad \text{for } m=\pm1.\end{aligned} \qquad (26)$$

The wave number $K$ is expressed in atomic units. It is numerically equal to the nuclear recoil velocity $V$ divided by the characteristic electron velocity $e^2/\hbar$.

In the case of beta-decay, $K$ is very much less than unity, so the yield of vacancies in the $K$ or $L$ shells due to nuclear recoil is very much less than the yield of about $1/Z^2$ due to the nuclear change of charge. Nuclear

TABLE I. Ionization probability in beta-decay.[a]

| | Upper limit | Our calculation | Migdal's calculation |
|---|---|---|---|
| $K$ electron | $0.75/Z^2$ | $0.326/Z^2$ | $0.32/Z^2$ |
| $K$ shell | $1.5/Z^2$ | $0.65/Z^2$ | $0.64/Z^2$ |
| $L_I$ electron | $2.25/Z^2$ | $0.47/Z^2$ | $1.9/Z^2$ |
| $L_{II}$ or $L_{III}$ | $1.25/Z^2$ | $0.21/Z^2$ | $0.5/Z^2$ |
| $L$ shell | $12/Z^2$ | $2.1/Z^2$ | $6.8/Z^2$ |

[a] The upper limit for ionization of orbital electrons due to beta-decay is given by Eq. (3) and following. The results of our calculations are given in Eqs. (15), (20), and (24), respectively. See reference 11 for Migdal's calculations.

[21] This approach to the problem of nuclear recoil was suggested by R. P. Feynman.
[22] Reference 18, Table XV.
[23] Reference 18, Eq. (39.7).

recoil is of importance in the cases of neutron or alpha emission.

### III. APPROXIMATIONS FOR BETA-DECAY

The calculations in the previous section are approximate in three respects: (1) the use of Coulomb wave functions; (2) neglect of relativistic effects, and (3) treatment of beta-decay as a sudden perturbation. In this section we shall correct the first approximation by calculating the screening corrections for Hartree wave functions[24] for Hg. (These functions do not include relativistic effects, or exchange effects.) We shall estimate the errors introduced by the other approximations. We shall also check our calculations by comparison with the average excitation energy calculated by Serber and Snyder.[16]

The probability $P_{00}$ that an electron remain in its initial state when the nuclear charge changes suddenly by one is given in Table I. The results given there are based on Coulomb wave functions, but for $K$ and $L$ electrons they should be reasonably accurate for Hartree wave functions. (For the $1s$ and $2p$ states of Hg there is excellent agreement between Hartree and Coulomb wave functions, provided we use Slater's screening constants to obtain the effective charge[25] for the latter.)

We shall calculate the probability that due to the sudden change of charge the electron be excited from its initial state 0 to some discrete state $n$. (We are considering a hypothetical atom where the states $n$ are unoccupied, so that the Pauli principle has no effect.) The probability $P$ of ionization to the continuum states is then

$$P=1-P_{00}-\sum_n P_{0n}. \qquad (27)$$

While the sum over discrete states $n$ should go to infinity, we find that for Hartree functions we can break off the sum after several terms.

Instead of calculating the matrix element $a_{0n}$ by

$$a_{0n}=\int \psi_n^*(Z+1,r)\psi_0(Z,r)d^3r, \qquad (28)$$

we express it in another form:

$$a_{0n}\cong(E_n-E_0)^{-1}\int \psi_n^*(Z,r)\Delta H\psi_0(Z,r)d^3r, \qquad (29)$$

where $\Delta H=e^2/r$ is the change of the Hamiltonian due to the sudden beta-decay.[26] $E_0$ and $E_n$ are the single electron energies estimated using Slater's effective $Z$.[27] The results of numerical integrations evaluating Eq. (29) for Hartree wave functions for Hg are given in Table II and compared with the matrix elements calculated using Coulomb functions. The first row $1-P_{00}$ gives the results of Table I for Coulomb functions, which should be a good approximation for Hartree functions. The last row $P$ (ionization probability to the continuum) is copied from Table I for Coulomb wave functions. For Hartree functions it is estimated as $P=1-P_{00}-P_{02}-P_{03}-P_{04}-P_{05}$.[28] The screening correction is the ratio of $P$ for Hartree and Coulomb wave functions. The factor $1/Z^2$ (or $1/Z_L^2$ for $L$ electrons) is omitted in Table I. Only the principal quantum number is given in the subscript and 0 means the initial state, e.g., $P_{03}$ means $P_{1s,3s}$ or $P_{2s,3s}$ or $P_{2p,3p}$ for $K$, $L_{\mathrm{I}}$ or $L_{\mathrm{II}}$ (or $L_{\mathrm{III}}$) electrons, respectively.

TABLE II. Screening corrections in $\beta$-decay for Hg. $1-P_{00}$ is the upper limit for the ionization probability, while $P_{01}$, $P_{02}$, etc., are the probabilities of excitation to discrete levels. The ionization probability $P=1-P_{00}-P_{01}-\cdots-P_{05}$. The screening correction is the ratio of $P$ for Hartree functions to $P$ for Coulomb functions.

| | $K$ electrons | | $L_{\mathrm{I}}$ | | $L_{\mathrm{II}}$ or $L_{\mathrm{III}}$ | |
|---|---|---|---|---|---|---|
| | Coulomb | Hartree | Coulomb | Hartree | Coulomb | Hartree |
| $1-P_{00}$ | 0.75 | ∼0.75 | 2.25 | ∼2.25 | 1.25 | ∼1.25 |
| $P_{01}$ | ... | ... | 0.312 | 0.28 | ... | ... |
| $P_{02}$ | 0.312 | 0.24 | ... | ... | ... | ... |
| $P_{03}$ | 0.059 | 0.038 | 1.08 | 0.47 | 0.783 | 0.37 |
| $P_{04}$ | 0.022 | 0.007 | 0.207 | 0.060 | 0.137 | 0.050 |
| $P_{05}$ | | | | | | 0.006 |
| $P$ | 0.326 | 0.46 | 0.47 | 1.44 | 0.21 | 0.82 |
| Screening corr. | 1.4 | | 3.1 | | 3.9 | |

This calculation shows that for heavy atoms the screening effects cause an appreciable increase in the ionization probability following $\beta$ decay: for $K$ electrons the factor is 1.4, while it is 3 or 4 for $L$ electrons.[29]

Screening corrections have been calculated by Reitz[30] for the beta-decay spectrum and for internal conversion coefficients using relativistic wave functions for the atomic field. He did not go to low enough energies to compare in detail with our results above. Thus, for Po he calculated only down to energies of 25 kev, or $W/E_K=0.3$. At this energy screening effects amounted to only 5 percent. Presumably the appreciable fraction of emitted electrons which have a continuum energy less than 0.3 of the $K$ binding energy (see Fig. 1) have quite large screening corrections, so that the screening correction averaged over the electron energy distribution of Fig. 1 amounts to an increase of 40 percent for $K$ electrons of mercury. The large increase in the screening

[24] D. R. Hartree and W. Hartree, Proc. Roy. Soc. (London) **A149**, 210 (1935).

[25] J. C. Slater, *Quantum Theory of Matter* (McGraw-Hill Book Company, Inc., New York, 1951), Appendix 13.

[26] D. Bohm, *Quantum Theory* (Prentice-Hall, Inc., New York, 1951), Sec. 20.6.

[27] Then the energy difference is appreciably greater than that calculated using Coulomb wave functions. If we used the atomic energies for the two different electron states, the energy difference would be appreciably less than that for Coulomb functions, due to the change in the screening of all the other electrons by the one electron making the transition. *Note added in proof:*—We now believe that the above was a poor choice for the energy difference, and gives too large screening corrections.

[28] The $P_{05}$ term is omitted for $K$ and $L_{\mathrm{I}}$ electrons.

[29] Screening effects are presumably of great importance in other problems involving the overlap integral of the wave function of an inner electron with that of an electron of low positive energy, e.g., ionization due to alpha-emission (Sec. V); calculation of absorption edges in the atomic photoeffect; or calculation of the stopping power of $L$ electrons.

[30] J. R. Reitz, Phys. Rev. **77**, 10 (1950).

correction for low continuum energies is substantiated by the much larger screening correction for $L$ electrons (3.1 and 3.9) which have in general much lower continuum energy than the electrons emitted from the $K$ shell.

Serber and Snyder's (S.S.) calculation[16] of the average excitation energy of the atom for beta-decay provides a valuable check on our calculations. They calculate the excitation energy of the whole atom as $\Delta E = 22.85Z^{2/5} = 132$ ev for Hg, using a Fermi-Thomas charge distribution. They also calculate an average excitation energy of 2 Ry$=27.2$ ev per closed electronic shell, assuming hydrogenic wave functions. These results are reached both by calculating the change of the nuclear electric potential energy due to the electronic cloud; and by a direct calculation using sum rules.

Their result of 2 Ry can be checked directly against the average excitation energy for $K$ hydrogenic wave functions, calculated from the second column of Table II. We also need the average positive energy given to a continuum electron: this is determined as $1.03Z^2$ Ry, by numerical integration using Eq. (14) for their energy spectrum. (Also see Fig. 1.) The average excitation energy $\Delta E$ for the $K$ shell is

$$\Delta E = 2Z^2\,\mathrm{Ry}[0.312Z^{-2}(0.75)+0.59Z^{-2}(0.889) +0.022Z^{-2}(0.938)+0.031Z^{-2}(0.97) +0.326Z^{-2}(2.03)] = 1.998\ \mathrm{Ry}. \quad (30)$$

The numbers in the bracket give the probability times the energy expenditure (factor of $Z^2$ Ry taken outside) for final states $2s$, $3s$, $4s$, $ns$ ($n>4$), and continuum $s$, respectively. The agreement with S.S. is within the accuracy of our numerical integration.

Now this calculation neglected the exclusion of transitions to occupied discrete states. The Pauli exclusion principle forbids, for example, a $1s$ to $2s$ transition in which the atom gains energy from the nuclear decay, but it also forbids a $2s$ to $1s$ transition in which the atomic system loses energy to the nuclear decay. The energies involved are exactly equal in magnitude. Thus the excitation energy *for the entire atom* is unaffected by the Pauli principle. However, the Pauli principle causes a smaller excitation energy than 2 Ry for the $K$ shell, and a correspondingly larger excitation energy for the higher shells.[31] (For hydrogen wave functions the excitation energy for the $K$ shell of Hg would be 1.38 Ry.)

Since we have not yet calculated the energy spectrum of continuum electrons for Hartree Hg wave functions, we cannot make a precise comparison between our $\Delta E$ and that of S.S. There will in general be a change of the excitation energy of the $K$ shell due to the use of atomic wave functions. This change in wave functions corresponds to a change in the S.S. treatment for closed shells, since they use the virial theorem for a Coulomb field to find the potential energy from the binding energy.

Relativistic effects will be of most importance for $K$ electrons of heavy atoms. We have calculated with relativistic Coulomb wave functions the probability $P_{00}$ that an electron remain in the $1s$ state for a change of one in the nuclear charge. The result for the upper limit for the ionization probability is, for $Z\gg 1$,

$$1-P_{00} = Z^{-2}[(2\gamma+1)/4+\alpha^2Z^2+\psi'(2\gamma)\alpha^4Z^4] = Z^{-2}(0.70+0.36+0.06) = 1.12Z^{-2}, \quad (31)$$

where

$$\gamma = (1-\alpha^2Z^2)^{\frac{1}{2}}, \quad (32)$$

and[32]

$$\psi'(x) = d^2(x!)/dx^2; \quad (33)$$

also $\alpha = e^2/\hbar c$. The numerical result in Eq. (30) is given for lead, $Z=82$, $\gamma=0.80$. Comparing with $1-P_{00} = 0.75/Z^2$ for nonrelativistic Coulomb functions [Eq. (3)], we see that for $K$ electrons of lead relativistic corrections might lead to an appreciable increase of the ionization probability. Our work is still preliminary, as we have not yet calculated the probability $P$ of ionization to the continuum. We have shown that relativistic corrections give an upper limit for the ionization probability of $1.12/0.46=2.4$ times that found in Table II for nonrelativistic Hartree wave functions.

In applying the sudden approximation to the case of beta decay, there will be a correction term of order $(v_e/v_b)^2$, where $v_e$ is the velocity of the orbital electron and $v_b$ that of the beta-particle. (See Feinberg's discussion[12] of the relatively low ionization produced by "direct collisions"). The coefficient of this correction term depends on the form of the time rate of change of the Hamiltonian $dH/dt$ [see Eq. (37) in Sec. IV], and this in turn depends on the values of the electron coordinates. For instance, if $dH/dt$ always has the same sign (say positive) we can show that

$$\left|\int_0^\infty (dH/dt)e^{i\omega t}dt\right|_{0n'} \leqslant \left|\int_0^\infty (dH/dt)dt\right|_{0n'}. \quad (34)$$

The right side is proportional to the amplitude for ionization of the $n'$ state for a sudden perturbation. Thus for this case the ionization probability found for the sudden perturbation case sets an upper limit for the ionization probability. But if $dH/dt$ changes sign as $t$ increases (as it does for $z$ positive) it appears that there will be a maximum in the ionization probability as a function of $(v_e/v_b)$ for the velocity of the emitted particle comparable to the velocity of the orbital electrons. Since we then integrate over $z$, we cannot be sure that Eq. (34) applies.

We should note that since $v_b$ always remains smaller than $c$, the parameter $v_e/v_b = Z/137n$ is not very much smaller than unity for $K$ electrons of heavy atoms; so

[31] This is analogous to the shift of oscillator strength from one shell to another in dipole transitions in x-ray spectra. See A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), p. 551.

[32] E. Jahnke and F. Emde, *Table of Functions* (Dover Publications, New York, 1945), p. 17.

the sudden perturbation treatment will not be very accurate for this case.

In summary, our approximations have in general underestimated the transition probabilities. For $K$ electrons the principal approximation was the use of nonrelativistic wave functions, which may lead to an underestimate of the ionization probability by a factor of 2. For $K$ electrons the screening corrections for the use of Coulomb wave functions lead to an increase of 40 percent in the transition probability. For $L$ electrons screening corrections for Hg give an increase by a factor 3.5. The sudden perturbation calculation should be a fair approximation for the case of beta-decay but becomes poorer for high $Z$.

## IV. ADIABATIC PERTURBATION: ALPHA-DECAY

The alpha-particles from radioactive nuclei travel with a velocity rather smaller than that of the inner orbital electrons (e.g., for $Po^{210}$ the ratio $v_a/v_e=0.087$ for $K$ electrons). The electron velocity $v_e=Z/n$ atomic units for $NR$ Coulomb wave functions, with principal quantum number $n$; $v_a$ is the velocity of the alpha-particle. Now the alpha-particle is so heavy that it has an extremely small wavelength, so it can be considered as moving through the atom classically, producing a perturbation on the atomic electrons that varies slowly compared with their motions. From the adiabatic theorem we know that a slowly varying perturbation produces very small effects, so we expect to find a very small ionization probability for the $K$ electrons. However, the $L$ electrons have a smaller velocity, so that the perturbation is not so adiabatic, and the transition probability is much larger.

Migdal[11] starts from the formulation of time dependent perturbation theory with perturbation $V(r,t)$ giving an amplitude for the state $n'$ at infinite time, for the system initially in state 0,

$$a_{n'}=(i\hbar)^{-1}\int_0^\infty V_{0n'}e^{i\omega t}dt$$
$$=(i\hbar)^{-1}[(i\omega)^{-1}e^{i\omega t}V|_0^\infty-\omega^{-2}e^{i\omega t}dV/dt|_0^\infty$$
$$-(i\omega)^{-3}e^{i\omega t}d^2V/dt^2|_0^\infty\cdots]. \quad (35)$$

The last expression is obtained by successive integrations by parts. This type of expansion is useful for a potential which varies slowly with time, so that the higher time derivatives are small. Migdal omits the term $(i\omega)^{-1}e^{i\omega t}V$, without a detailed explanation. This omission is clearer if we use another formulation of time dependent perturbation theory[33] for a Hamiltonian $H$ which is a function of time:

$$\dot{a}_{n'}=\sum_n(a_n/\hbar\omega_{n'n})\left[\exp\left(i\int_0^t\omega_{n'n}dt'\right)\right](dH/dt)_{n'n}. \quad (36)$$

[33] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949), Eq. (31.8).

This equation is exact. If the perturbation is small, we can take $a_n(t)=\delta_{n0}$ and we can take $\omega_{n'n}(t')$ and the wave functions $u_n(t)$ and $u_{n'}(t)$ all as time independent. This gives the amplitude after infinite time for the state $n'$

$$a_{n'}=(\hbar\omega)^{-1}\int_0^\infty e^{i\omega t}(dH/dt)_{0n'}dt$$
$$=(\hbar\omega)^{-1}[(i\omega)^{-1}e^{i\omega t}dH/dt|_0^\infty$$
$$-(i\omega)^{-2}e^{i\omega t}d^2H/dt^2|_0^\infty\cdots]. \quad (37)$$

This expression automatically omits the term involving $V$ (or the change of $H$), justifying Migdal's omission of this term.[34]

The significant difference between our present calculation and that of Migdal's is that he takes $V$ as the perturbation on the orbital electrons due to the alpha-particle, while we take the Hamiltonian $H$, which includes both the effect of the alpha-particle and the effect of the nucleus. Migdal uses

$$V=2e^2[x^2+y^2+(z-v_at)^2]^{-\frac{1}{2}}, \quad t\geq 0. \quad (38)$$

For the dipole term he finds

$$-(i\hbar)^{-1}\omega^{-2}e^{i\omega t}dV/dt|_0^\infty$$
$$=-(i\hbar\omega^2)^{-1}2e^2v_ar^{-2}P_1(\cos\theta). \quad (39)$$

Here the alpha-particle moves with velocity $v_a$ along the positive $z$ axis, starting at $t=0$. $P_1$ is a Legendre polynomial. Since we use the total Hamiltonian $H$ for a nucleus initially of atomic number $Z$, recoiling with velocity $v_n$:

$$H=2e^2[x^2+y^2+(z-v_at)^2]^{-\frac{1}{2}}$$
$$+(Z-2)[x^2+y^2+(z+v_nt)^2]^{-\frac{1}{2}}, \quad t\geq 0, \quad (40)$$

we find for the dipole $dH/dt$ term in Eq. (37):

$$(\hbar\omega)^{-1}(i\omega)^{-1}e^{i\omega t}dH/dt|_0^\infty$$
$$=(i\hbar\omega^2)^{-1}[(Z-2)e^2v_n-2e^2v_a]P_1(\cos\theta). \quad (41)$$

The matrix elements of the expression in Eq. (39) or Eq. (41) can be evaluated easily by a trick used by Migdal based on potential energy $V=Ze^2/r$, and Ehrenfest's relations:

$$[r^{-2}P_1(\cos\theta)]_{0n'}=(z/r^3)_{0n'}=[-\partial/\partial z(r^{-1})]_{0n'}$$
$$=m(\ddot{z})_{0n'}/Ze^2=(m\omega^2/Ze^2)(z)_{0n'}. \quad (42)$$

The probability of ionization due to alpha-decay is then, according to Migdal,

$$P_d=\sum_{n'}|a_{n'}|^2=(4v_a^2/Z^2)\sum_{n'}|z_{0n'}|^2. \quad (43)$$

We are using Hartree atomic units, so $v_a$ is the alpha-particle velocity in atomic units, and $\hbar$ and $m$ are equal to unity. Now this same sum over continuum states of

[34] The physical reason for omitting this term is that it corresponds exactly to a sudden change of the Hamiltonian, which does not occur in this case. This term was erroneously included in our calculation for alpha-decay in reference 13.

the squared dipole moment was met above in calculations by sudden perturbation theory of the ionization due to nuclear recoil; and the results for these probabilities, copied from Bethe's Handbuch article, are tabulated in Eq. (26). They are also tabulated in Migdal's Table II, where he includes the factor of 4 for the squared charge of the alpha particle, and also multiplies by the number of 1$s$, 2$s$, and 2$p$ electrons, respectively. To show the relation to the adiabatic theorem, he uses the electron velocity $v_e=Z$ atomic units, so that the ionization probability for dipole transitions per 1$s$ electron can be written

$$P_d=4(v_a/v_e)^2(0.28/Z^2), \tag{44}$$

with the coefficients for 2$s$ and 2$p$ electrons to replace the 0.28 being given in Eq. (26).

However, we use Eq. (41) for $dH/dt$, and obtain for the ionization probability,[35]

$$P_d=\{[(Z-2)v_n-2v_a]^2/Z^2\}\sum_{n'}|z_{0n'}|^2, \tag{45}$$

to replace Migdal's result of Eq. (43). This suggests the definition

$$Zv_z=(Z-2)v_n-2v_a, \tag{46}$$

so that Eq. (45) can be rewritten in a form analogous to Eq. (26), the ionization probability $P$, for 1$s$ electrons:

$$P_d=v_z^2\sum_{n'}|z_{0n'}|^2=0.28v_z^2/Z^2. \tag{47}$$

The physical interpretation of the last two equations is that $v_z$, as defined in Eq. (46), is the velocity of the *center of charge* of the system composed of alpha-particle with velocity $v_a$ and recoiling nucleus with charge $Z-2$ and velocity $v_n$ in the opposite direction. Further, for the dipole transitions which we are here considering, only the motion of the center of charge is of significance.[36] At time 0, the atomic electrons begin to feel the perturbation of the alpha-particle traveling one way, and the remainder of the charge recoiling in the other direction. Since the electron velocities are large compared to the velocity of separation of alpha and nucleus, the electrons to a first approximation (i.e., the dipole approximation) average out in their motion the fields of alpha and recoiling nucleus, and experience only the sudden change from no motion of the center of charge to motion of the center of charge with velocity $v_z$; thus Eq. (26) is applicable.

If the alpha-particle and recoiling nucleus had identical charge-to-mass ratios, the center of charge of the system would remain at rest, since the center of mass must do so. Using conservation of momentum, and the definition Eq. (46) we have in the general case:

$$Zv_z=2v_a(A-2Z)/(A-4)=2v_a(0.203). \tag{48}$$

The numerical value 0.203 is for the case $Po^{210}$; for $U^{238}$ the numerical coefficient is 0.232. Since $Zv_z$ replaces $2v_a$ used by Migdal [Eqs. (43) and (45)], and these quantities enter squared in calculating the ionization probability, we find for this term an ionization probability of $(A-2Z)^2/(A-4)^2=0.04$ that found by Migdal, where the number 0.04 applies to $Po^{210}$. For $K$ electrons of $Po^{210}$ Migdal finds a probability of $2.5\times10^{-6}$ per alpha, while we find an ionization probability of only $10^{-7}$ per alpha. (These numbers come from Eq. (47) or (43), using $v_a/v_e=0.087$, as given above.) Since $v_a/v_e\ll1$, Migdal calculated only the dipole term.

We have found that the dipole $dH/dt$ term is greatly reduced due to the nuclear recoil. However, the quadrupole $d^2H/dt^2$ term of Eq. (37) is changed very little. From Eq. (40),

$$d^2H/dt^2|_0^\infty=-2e^2P_2(\cos\theta)r^{-3}[2v_a^2+(Z-2)v_n^2]. \tag{49}$$

Here, since the small velocity $v_n$ appears squared, the nuclear recoil term can be neglected.

Since the dipole term is made unusually small by the effect of the nuclear recoil, while the quadrupole term is almost unaffected, we see that the type of series Eq. (35) or (37) has the special property that both the dipole and quadrupole terms need to be calculated, but that one can hope to neglect higher multipoles. It turns out that the quadrupole term contains numerical coefficients markedly different from unity, so that it is very small compared to the dipole term for $K$ electrons; but large compared to the dipole term for $L$ electrons. Equation (35) or (37) are actually semiconvergent series, with the general term for the amplitude $a_p$ for

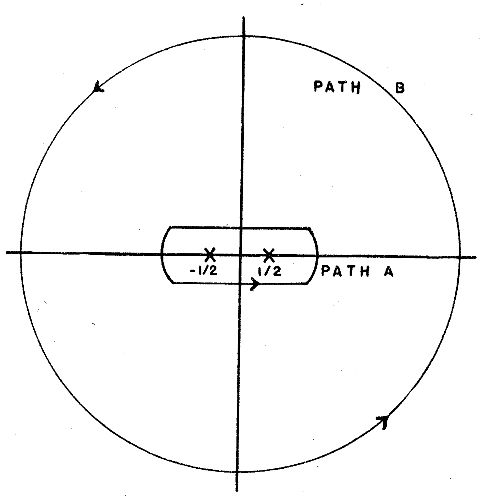


FIG. 2. Contour for Eq. (56). Path A surrounds the branch points at $\xi=\pm\frac{1}{2}$; path B has the large radius $\epsilon^{-1}$.

[35] The shape of the energy spectrum for the emitted electrons is the same as that given by Migdal. It is plotted in Fig. 1, with an arbitrary scale for the ordinate. We see that the emitted $K$ electrons are likely to have a positive energy the order of the $K$ binding energy; but that there are fewer high energy electrons accompanying alpha-decay than beta-decay. Migdal shows that asymptotically the $K$ electrons from alpha-decay fall off as $E^{-9/2}$; while the $K$ electrons from beta-decay fall off as $E^{-7/2}$.

[36] For an analogous use of the concept of the center of charge in the nuclear photoeffect see H. A. Bethe, Revs. Modern Phys. **9**, 71 (1937), Secs. 87 to 90. Dr. Bethe suggested use of the concept "center of charge" in our present problem.

the $2^p$ pole transition being

$$a_p = 2e^2[(\hbar\omega)^{-1}(v_a/i\omega)^p r^{-p-1} p! P_p(\cos\theta)]_{0n'}. \quad (50)$$

Successive terms decrease rapidly for the case of $K$ electrons of $Po^{210}$; but much less rapidly for the $L$ electrons. In this section we shall calculate the quadrupole term for $K$ and $L$ electrons. We discuss the englect of higher terms in the next section.

Migdal's trick of Eq. (42) does not work for the quadrupole term of Eq. (49). Professor Bethe has developed a method to use the same integral representation of the continuum wave functions that we use above[19] for our present matrix element which involves negative powers of $r$. (The angular integration for $P_2(\cos\theta)$ gives the usual quadrupole selection rules for the angular momentum change, and for allowed transitions gives numerical factors which will be included in our final result.) For $K$ electrons we have transitions to continuum states, with the matrix element

$$(r^{-3})_{1s,\,n'd} = N_3 J_3, \quad (51)$$

$$N_3 = -4Z^2(n'^2+1)^{\frac{1}{2}}(n'^2+4)^{\frac{1}{2}}(1-e^{-2\pi n'})^{-\frac{1}{2}}, \quad (52)$$

$$J_3 = (2k)^{-3}(2\pi)^{-1}\oint\int e^{-\beta_1 r} r^{-4} dr (\xi+\tfrac{1}{2})^{-in'-3} \times (\xi-\tfrac{1}{2})^{in'-3} d\xi, \quad (53)$$

$$\beta_1 = 2ik(\xi - in'/2). \quad (54)$$

The integration over $r$ is performed first; and since it diverges at the lower limit, we replace this limit by a small number $\epsilon$, and integrate by parts several times:

$$\int_\epsilon^\infty e^{-\beta_1 r} r^{-4} dr = \epsilon^{-3} e^{-\beta_1\epsilon}/3 - \epsilon^{-2}\beta_1 e^{-\beta_1\epsilon}/6 + \epsilon^{-1}\beta_1^2 e^{-\beta_1\epsilon}/6 - (\beta_1^3/6)\int_\epsilon^\infty e^{-\beta_1 r} r^{-1} dr. \quad (55)$$

Consider any one of the first three terms on the right side, such as $\epsilon^{-3}\exp(-\beta_1\epsilon)/3$. For this term the integral over $\xi$ is

$$I = (3\epsilon^3)^{-1}\oint e^{-\beta_1\epsilon}(\xi+\tfrac{1}{2})^{-in'-3}(\xi-\tfrac{1}{2})^{in'-3} d\xi. \quad (56)$$

The contour integral is to be taken over the path A of Fig. 2, which surrounds the branch points at $\xi=\frac{1}{2}$ and $\xi=-\frac{1}{2}$. This path can be distorted to path B of very large radius without changing the value of the contour integral $I$. On path B, let $\xi=\epsilon^{-1}e^{i\theta}$, so that $\exp(-\beta_1\epsilon)$ becomes $\exp(-2ike^{i\theta}+kn'\epsilon)$ which is finite. The terms $(\xi+\frac{1}{2})^{-in'-3}(\xi-\frac{1}{2})^{in'-3}$ give a factor of $\epsilon^6$, so that $I$ is proportional to $\epsilon^3$ times an integral of a finite quantity over a path of length $2\pi\epsilon^{-1}$, i.e., $I$ is proportional to $\epsilon^2$ and therefore vanishes as we take the limit $\epsilon$ to 0. Similarly the integrals over path B vanish for the second and third terms on the right of Eq. (55). We are then left with the last term, for which we use the

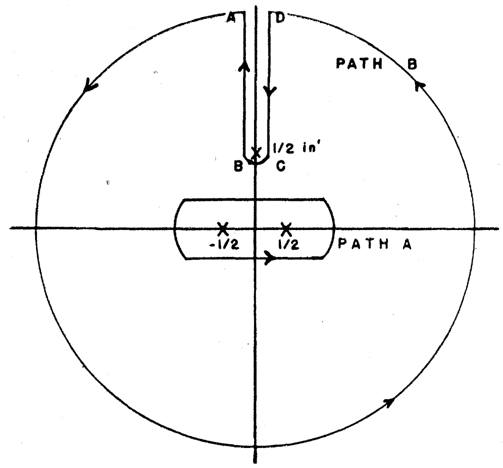


FIG. 3. Contour for Eq. (58). Path A surrounds the branch points at $\xi=\pm\frac{1}{2}$; path B is at infinity except for the portion $ABCD$ surrounding the branch point at $\xi=\frac{1}{2}in'$.

asymptotic expression for Ei$(x)$:

$$(-\beta_1^3/6)\int_\epsilon^\infty e^{-\beta_1 r} r^{-1} dr = (-\beta_1^3/6)(\ln\beta_1 + \ln\epsilon + \text{constant}). \quad (57)$$

The terms with $\ln\epsilon$ and constant vanish on integration over $\xi$ by the same argument as above for $I$. The $\ln\beta_1$ term does not, since it has a branch point at $\beta_1=0$, or $\xi=\frac{1}{2}in'$. The contour integral must be deformed as shown in Fig. 3 in changing from path A to path B, since we must not cross any singularities. Since $\ln\beta_1$ differs by $2\pi i$ on the two linear portions $AB$ and $CD$, and since the remainder of the contour (at infinity and on the small circle) gives zero, the integral over $\xi$ becomes

$$J_3 = (2k)^{-3} i \int_{\frac{1}{2}in'}^{\infty i} (\beta_1^3/6)(\xi+\tfrac{1}{2})^{-in'-3}(\xi-\tfrac{1}{2})^{in'-3} d\xi$$
$$= -(2/3)\int_0^{\cot^{-1}n'} (\cot x - n')^3 \sin^4 x e^{-2n'x} dx$$
$$= [1 - 9\exp(-2n'\cot^{-1}n')]/12(n'^2+4). \quad (58)$$

We obtain the second integral on the right using the substitution $\xi=\frac{1}{2}i\cot x$.

We note that Bethe's method of finding the matrix element for negative powers of $r$ for Coulomb wave functions has been checked by calculating the matrix element of $r^{-2}\cos\theta$ and comparing with Eq. (42) for the relation to the matrix element of $r\cos\theta$.

To find the amplitude for transitions to the $n'd$ state, we combine the matrix element $(r^{-3})_{1s,\,n'd}=N_3J_3$ [with $N_3$ given by Eq. (52) and $J_3$ by Eq. (58)] with the factor $5^{-\frac{1}{2}}$ from the angular integration of $P_2(\cos\theta)$ between $s$ and $d$ states, and with the factor $4e^2v_a2/\hbar\omega^3$

of Eq. (50). Here it is convenient to use

$$\hbar\omega = E_K + W = \tfrac{1}{2}Z^2 + \tfrac{1}{2}k^2 = \tfrac{1}{2}Z^2 n'^{-2}(n'^2+1). \quad (59)$$

We square the amplitude, multiply by $dW = -Z^2 dn'/n'^3$ and integrate to find the ionization probability for the quadrupole term for $K$ electrons

$$P_q = (2^{10}/45)(v_a/v_e)^4 Z^{-2} \times \int_0^\infty n'^9[9\exp(-2n'\cot^{-1}n') - 1]^2 \times (n'^2+1)^{-5}(n'^2+4)^{-1}[1-e^{-2\pi n'}]^{-1}dn' = 0.22(v_a/v_e)^4 Z^{-2}. \quad (60)$$

The final result is obtained by numerical integration. We note that the integrand has a sharp peak at $n' \cong 1$, or continuum energy $W \cong$ binding energy $E_k$.

It is of interest to compare the ionization probability by the quadrupole term with that found for the dipole term in Eq. (47).

$$P_d = (0.203)^2 4 v_a^2 (0.28/Z^2). \quad (61)$$

TABLE III. Ionization probabilities for $Po^{210}$ alpha-decay.[a]

| Electron | Equations | | Numerical results | |
|---|---|---|---|---|
| | Dipole | Quadrupole | Dipole | Quadrupole |
| $K$ | $0.28\,(Zv_z/v_K)^2$ | $0.22\,(v_a/v_K)^4$ | 0.00034 | 0.000013 |
| $LI$ | $0.90\,(Zv_z/v_K)^2$ | $86\,(v_a/v_{LI})^4$ | 0.0011 | 0.17 |
| $LII$ | $0.55\,(Zv_z/v_K)^2$ | $(15+4)(v_a/v_{LII})^4$ | 0.0007 | 0.037 |
| $LIII$ | $0.55\,(Zv_z/v_K)^2$ | $(21+6)(v_a/v_{LIII})^4$ | 0.0007 | 0.075 |

[a] Here $v_K$, $v_{LI}$, $v_{LII}$, and $v_{LIII}$ are the velocities of the $K$, $L_I$, $L_{II}$ and $L_{III}$ electrons, respectively, $v_a$ is the alpha-particle velocity; while $v_z$ is the velocity of the center of charge of the system of alpha-particle and recoiling nucleus: see Eq. (46). The table gives probabilities of ionization per electron; and the factor $(1/Z^2)$ is omitted throughout the table.

The ratio

$$P_q/P_d = 0.196(v_a/v_e)^2/(0.203)^2 = 0.036. \quad (62)$$

The use of $(0.203)^2$ applies to the case of $Po^{210}$, as does the final result using $v_a/v_e = 0.087$. We see that for $K$ electrons the quadrupole term is very much smaller than the dipole term, even though the dipole term is reduced by a factor 25 due to nuclear recoil, while the quadrupole term is unchanged by nuclear recoil. The small numerical factor $0.22/4(0.28) = 0.196$ is of significance in reaching this conclusion; the corresponding numerical factor turns out to be large for the case of ionization of $L$ electrons, and the quadrupole term predominates.

The calculation for the ionization probability of $L$ electrons is carried through in a manner completely analogous to the calculation above for the $K$ electrons, so we will give few details. There are two changes. First putting the binding energy $E_L = \frac{1}{2}Z^2/n^2$, for $n=2$ is a poor approximation, even if we use an effective $Z_L$ as given by Slater. We introduce an empirical parameter $\theta$ to fit the experimental data, defined as

$$E_L = \theta Z^2/8. \quad (63)$$

For Po, $\theta = 0.75$ for $L_I$ and $K_{II}$ electrons, and $\theta = 0.63$ for $L_{III}$ electrons. In place of Eq. (59) for $K$ electrons we have then

$$\hbar\omega = E_L + W = \theta Z^2/8 + \tfrac{1}{2}k^2 = (\theta Z^2/8)n'^{-2}(n'^2 + 4/\theta). \quad (64)$$

We will express our final result in terms of the velocity $v_L$ of the $L$ electron, defined as

$$v_L = \tfrac{1}{2}Z\theta^{\frac{1}{2}} \text{ atomic units.} \quad (65)$$

Second, for $2p$ electrons we obtain different results for the angular integrations for the two cases $m=0$ and $m=1$, which must be weighted appropriately.

The quadrupole contribution to ionization probability per $L_I$ electron is

$$P_2 = (2^{11}/45\theta^4)(v_a/v_L)^4 Z^{-2} \times \int_0^\infty \frac{n'^9[A(n') + B(n')]^2 dn'}{(n'^2+4/\theta)^6(n'^2+4)(n'^2+1)(1-e^{-2\pi n'})} = (86/Z^2)(v_a/v_L)^4 \text{ for } \theta = 0.75. \quad (66)$$

Here

$$A(n') = 8n'^2 - 9n' + 4,$$

and

$$B(n') = (-24n'^6 + 153n'^5 + 196n'^4 + 648n'^3 - 1308n'^2 + 144n' + 576)(n'^2+4)^{-2} \times \exp(-2n'\cot^{-1}\tfrac{1}{2}n').$$

We note that the integrand has a maximum at $n' \cong 6$, corresponding to a continuum energy of only $0.15E_L$.*

Quadrupole transitions for $2p$ electrons can be either to $n'p$ or to $n'f$ continuum states. Also, as noted above, we have different results for the angular integrations depending on the value of $m$ for the $2p$ electron, and we have different values of the parameter $\theta$ for $L_{II}$ and $L_{III}$ electrons. The probability of a transition to an $n'p$ state for $m=0$ is

$$P_3 = (2^{15}/75\theta^4)(v_a/v_L)^4 Z^{-2} \times \int_0^\infty \frac{n'^{11}[1 + 3\exp(-2n'\cot^{-1}\frac{1}{2}n')]^2 dn'}{(n'^2+4/\theta)^6(n'^2+1)(1-e^{-2\pi n'})}. \quad (67)$$

For $L_{II}$ electrons we have $P_3 = (15/Z^2)(v_a/v_{LII})^4$, while for $L_{III}$ electrons $P_3' = (21/Z^2)(v_a/v_{LIII})^4$.

The probability of a quadrupole transitions from a $2p$ state, with $m=0$, to an $n'f$ state is given by

$$P_4 = (2^{11}/75\theta^4)(v_a/v_L)^4 Z^{-2} \times \int_0^\infty \frac{n'^{11}[A_1(n') + B_1(n')]^2 dn'}{(n'^2+4/\theta)^6(n'^2+1)(n'^2+4)(n'^2+9)(1-e^{-2\pi n'})}, \quad (68)$$

* *Note added in proof:*—We have found an error in this calculation, with the help of J. Scandrett. We should have $A(n') = n'^2 + 4$ and change the sixth-order polynomial in $B(n')$ to read $(-45n'^6 - 396n'^4 - 1008n'^2 - 576)$. The number 86 in the result is changed to 0.044. This lowers the quadrupole yield for $L_I$ electrons to $8.7\times10^{-5}/Z^2$ (Table III) and lowers the $L$ shell yield to $0.6\times10^{-4}$ (Table IV).

$$A_1(n') = -3n'^2 - 12,$$

$$B_1(n') = (225n'^6 + 2500n'^4 + 38{,}348n'^2 + 3264) \times (n'^2+4)^{-2} \exp(-2n' \cot^{-1}\tfrac{1}{2}n').$$

For $L_{II}$ electrons $P_4 = (3.6/Z^2)(v_a/v_{LII})^4$ while for $L_{III}$ electrons, $P_4' = (6.2/Z^2)(v_a/v_{LIII})^4$. The peak of the integrands in Eqs. (67) and (68) are at $n'$ about 3, or a continuum energy $W \cong 0.6E_L$.

These results for the dipole and quadrupole contributions to the ionization probability are collected in Table III, together with a numerical evaluation for the case of $Po^{210}$. We have averaged over the results for $m=0$ and $m=1$ cases for the $2p$ electrons. The probabilities are all given omitting the factor $1/Z^2$. The two numbers given for quadrupole transitions for $L_{II}$ and $L_{III}$ electrons are the contributions from transitions to $n'p$ and $n'f$ states, respectively.

In the numerical evaluation we used: the ratio of alpha-velocity to velocity of the $K$ electron, $v_a/v_K = 0.087$ for $Po^{210}$; the ratio of alpha-velocity to that of the $L_I$ or $L_{II}$ electron, $v_a/v_{LI} = v_a/v_{LII} = 0.21$; the ratio of alpha-velocity to that of the $L_{III}$ electron $v_a/v_{LIII} = 0.23$. $v_z$ is the velocity of the center of charge of alpha and recoiling nucleus; from Eq. (48), $Zv_z/v_a = 0.406$.

In calculating the ionization probabilities we combine the dipole and quadrupole terms; we use the effective charges $Z_K = Z - 0.3$, and $Z_L = Z - 4$; and we also multiply by the number of the electrons in each shell. The resulting ionization probabilities for the different shells are given in Table IV.

As stated above, the very large ratio between ionization yields for $L$ electrons and $K$ electrons is due both to the adiabatic theorem, and to the very different numerical factors in the probabilities for quadrupole transitions: e.g., 15 for $2p$ electrons as compared with 0.22 for $1s$ electrons. The quadrupole transition probability for the $L$ shell is about 100 times the dipole transition probability. While a factor of 25 is accounted for by the decrease of the dipole transition probability due to nuclear recoil, the remaining factor of 4 indicates that the semiconvergent expansion of Eq. (37) may not give accurate results for the value $v_a/v_L = 0.2$ for $Po^{210}$. The problem of the accuracy of our semiconvergent expansion is discussed further in the next section.

In the above work we have assumed that the alpha-particle was moving along a radius vector from the nucleus: that is, that we were dealing with an $s$ alpha-particle. The imperfect analogy with the internal conversion effect suggests a strong dependence of ionization probability on the alpha-particle angular momentum. This question is considered in some detail in reference 13, where we have shown that the alpha-particle angular momentum has a negligible effect on the ionization probability. Two possible effects are considered: (1) that the classical path of an alpha-particle depends appreciably on its angular momentum relative to the nucleus; and (2) that an alpha-particle of high angular momentum can penetrate the centrifugal barrier more easily if it first gives up some angular momentum to an electron, by ionizing it. The first effect is found to be extremely small. The second effect is also extremely small, since the loss of energy by the alpha-particle in ionizing the electron greatly decreases its rate of penetration through the Coulomb barrier. (This illustrates the difference between internal conversion and the present process of "internal ionization." In internal conversion emission of an electron is a complete substitute for emission of a nuclear gamma, since the emitted electron takes off both the energy difference and the angular momentum difference between the nuclear states. However, in alpha-particle emission, an alpha-particle must still be emitted to change the nuclear $Z$ and $A$, irregardless of whether an orbital electron is also emitted. The cases of beta or positron emission, or $K$ capture, are similar to that of alpha-emission. The correct analogy for these processes in which particles (alpha, beta, or positron) are emitted from the nucleus is to internal Compton scattering, rather than to internal conversion.)

TABLE IV. Ionization probability for electron shells for $Po^{210}$ alpha-decay.

| Electron shell | Ionization probability per alpha |
|---|---|
| $K$ | $0.98\times10^{-7}$ |
| $L_I$ | $0.53\times10^{-4}$ |
| $L_{II}$ | $0.12\times10^{-4}$ |
| $L_{III}$ | $0.47\times10^{-4}$ |
| $L$ shells | $1.1\times10^{-4}$ |

## V. APPROXIMATIONS IN ALPHA-DECAY

The approximations made in our treatment of the ionization probability of $K$ and $L$ electrons due to alpha-decay are similar to our approximations in the beta-decay treatment. (See Sec. III.) We shall calculate the screening correction for dipole transitions, and estimate the screening correction for quadrupole transitions. We shall also estimate the accuracy of our nonrelativistic approximation, and of our semiconvergent expansion.

The screening corrections for dipole transitions can be calculated in a manner rather similar to that of Sec. III. We calculate the dipole matrix elements to discrete states, using Hartree wave functions[24] for Hg. The sum of the squares of all dipole matrix elements $r_{0n}$ can be found from the sum rule,

$$\textstyle\sum_n (r_{0n})^2 = (r^2)_{00}, \qquad (69)$$

where 0 is the initial state, and the sum goes over both discrete and continuum states. The ionization probability is then proportional to $(r^2)_{00}$ minus the sum over the discrete states of the squared dipole matrix elements.

TABLE V. Screening corrections for dipole transitions of $K$ electrons.[a]

| Matrix element | Coulomb functions | Hartree for Hg | Hartree-Fock for Cu | Hartree-Fock for A |
|---|---|---|---|---|
| $(r^2)_{1s,1s}$ | 3.000 | 2.980 | $\sim$3.0 | $\sim$3.0 |
| $(r_{1s,2p})^2$ | 1.666 | 1.525 | 1.378 | 1.186 |
| $(r_{1s,3p})^2$ | 0.267 | 0.204 | 0.117 | 0.067 |
| $(r_{1s,4p})^2$ | 0.093 | 0.044 | ... | ... |
| $(r_{1s,5p})^2$ | 0.044 | 0.008 | ... | ... |
| continuum probability | 0.849 | 1.19 | $\sim$1.5 | $\sim$1.7 |
| screening correction | | 1.4 | $\sim$1.8 | $\sim$2.0 |

[a] The squared matrix elements are given in units of $a_0^2/Z^2$. The values for Cu and A were calculated by Tuan (reference 37). The value of the continuum probability—i.e., sum of the squared matrix elements over all continuum states—is calculated as $(r^2)_{1s,1s}-(r_{1s,2p})^2-\cdots-(r_{1s,5p})^2$. The screening correction is the ratio of continuum probability for atomic wave functions to that for Coulomb wave functions.

We have made this calculation only for the $K$ electrons, as dipole transitions were found in Sec. IV to be unimportant for higher shells. The results are given in column 3 of Table V. We also include in columns 4 and 5 results for Cu and A, using dipole matrix elements calculated by Tuan[37] for Hartree-Fock wave functions. Column 2 gives the result for Coulomb wave functions.[22]

The screening correction, calculated as the ratio of the ionization probability for atomic wave functions to that for Coulomb wave functions, is 1.4 for Hg, and about 2 for Cu and A. The screening correction for dipole transitions of Hg $K$ electrons is the same as that found in Sec. III for the screening correction in the beta-decay case for the same electrons.

These screening corrections cannot be applied exactly to our calculation of dipole transitions due to alpha-decay, since we originally [Eq. (41)] had the matrix element of $r^{-2}\cos\theta$ which Migdal transformed to a matrix element of $r\cos\theta$, using relations based on there being a Coulomb potential. For the atomic potential the transformation will no longer be exact, but it should be in error by an amount much less than the screening correction of 40 percent.

This method of calculation of screening corrections cannot be applied to quadrupole transitions (which are of interest for the $L$ shell) since we have no sum rule for this case. We can estimate that the screening corrections for quadrupole transitions of $L$ electrons will be somewhat greater than the factor of 3 to 4 found for $L$ electrons in Sec. III for the beta-decay case. Screening corrections became larger for lower energy continuum electrons (e.g., comparison of $K$ and $L$ shells in Sec. III); and the electrons emitted from the $L$ shell in quadrupole transitions [Eqs. (66), (67) and (68)] have somewhat lower energy than those emitted from the $L$ shell in beta-decay [Eqs. (20) and (24)].†

[37] T. F. Tuan, M.S. thesis, Louisiana State University, 1952 (unpublished).

† *Note added in proof:*—Preliminary calculations for quadrupole transitions using the atomic wave functions of Ramberg and of Reitz show only a small screening effect, contradicting our estimate above.

We can make only a tentative estimate on the magnitude of relativistic corrections. Analogies with other processes (ionization by beta-decay, Auger transition rate) suggest a possible increase in the ionization probability of a factor of 2 for $K$ electrons of heavy elements, and a much smaller correction for $L$ electrons.

The accuracy of the adiabatic approximation of expansion into dipole, quadrupole, and higher electric multipole terms is difficult to estimate, since this is a semiconvergent expansion, as noted above. From Eq. (50) the order of magnitude of the ratio of the amplitude for the $2^{p+1}$ pole to $2^p$ pole transition is, in the general case,

$$a_{p+1}/a_p=(p+1)v_a/\omega r\cong(p+1)v_a/v_e. \tag{70}$$

For $Po^{210}$ and $L$ electrons, this gives $a_3/a_2$ about 0.6; which may be changed quite significantly by numerical factors that occur in calculating matrix elements. [These are omitted in our crude expression Eq. (70).] In an attempt to estimate the accuracy of our semiconvergent expansion we have calculated one of the two octupole transitions for $2p$ electrons, namely $2p$ to $n'd$. We find a transition probability, summed over all continuum states, of $(95/Z^2)$ $(v_a/v_{LIII})^6$, which should be compared with the probability for $2p$ to $n'p$ quadrupole (Table III) of $(21/Z^2)(v_a/v_{LIII})^4$. The ratio 4.5 $(v_a/v_{LIII})^2=0.24$ for the case of $Po^{210}$. Our expansion appears to be quite good for $Po^{210}$ and $K$ electrons; and may be satisfactory within 50 percent accuracy for $Po^{210}$ and $L$ electrons. It seems probable that higher multipole terms, such as the octupole, will increase the transition probability over that calculated for dipole and quadrupole terms alone.

Our summary of corrections to the calculations of ionization probability in alpha-decay is similar to that of Sec. III for the beta-decay case. For $K$ electrons of Po, corrections will increase the probability perhaps by a factor of 2; screening corrections give a 40 percent increase. The expansion of the adiabatic approximation holds quite well. For $L$ electrons of Po, relativistic corrections are small; screening corrections lead to an increase in the ionization probability of somewhat more than a factor of 4; and the expansion of the adiabatic approximation is of uncertain validity.

## VI. EXPERIMENTAL RESULTS

Experiments might measure any of three different effects of the ionization of inner electrons due to beta- (or positron) or alpha-decay of the nucleus: (1) the emitted orbital electrons; (2) the characteristic x-rays emitted in filling the holes in the $K$ or $L$ shells; (3) the Auger electrons emitted in competition with the x-ray emission. Very thin, or appropriately diluted sources, must be used to make certain that the effects due to beta- or alpha-decay occur in the same atom, rather than in neighboring atoms. Further, internal conversion of nuclear gammas or emission of inner bremsstrahlung (in beta-decay) may mask or confuse the effects calculated in this paper. The confusion with internal conversion can best be avoided by choosing sources which emit no nuclear gammas. Also, the line spectrum of electrons from internal conversion could in principle be distinguished from the effects discussed in this paper. The emission of inner bremsstrahlung in beta-decay has been treated carefully theoretically, and recently confirmed in detail by experiment[1,2] so that this effect can be subtracted out.

The first effect listed above has been studied by Bruner[3] for the isotope $Sc^{44}$ which emits positrons, and nuclear gammas and also undergoes $K$-capture. He used two different magnetic beta-spectrometers to measure the energy spectrum of electrons of energy from 30 to 150 kev emitted from $Sc^{44}$, and found consistent spectra giving a ratio of electrons/positrons of 4 percent. This high ratio cannot be reconciled with the low yields for the effects calculated in this paper. The yield of $K$ electrons ionized by the sudden perturbation is only $0.64/Z^2$ (increased by perhaps a factor of 2 by screening corrections), and almost all of these are of much smaller energy than that of the electrons measured by Bruner. (See Fig. 1.) He suggests a mechanism of internal conversion of inner bremsstrahlung. This process is very much less probable than Bruner supposes, since the analogy between the internal conversion of nuclear gammas and that of inner bremsstrahlung is poor.[38]

Recently Porter and Hotz[4] have made a cloud-chamber study of electrons from 30 kev to 205 kev accompanying the $K$ capture activity of $Fe^{55}$. They find an upper limit of $0.6\times 10^{-6}$ electron ejected per $K$ capture, in sharp disagreement with Bruner's yield of about 4 percent. Porter and Hotz quote a theoretical prediction[4] of about $10^{-6}$ for the probability of ejection of an electron in this energy range. This figure is consistent with the work of Migdal,[11] Feinberg[12] and the present author[13] for the effects of beta-decay: the probability of ionization is $0.32/Z^2$; but only 0.4 percent of the ejected electrons from iron have an energy greater than 4.3 times the binding energy, or 30 kev [see Eq. (14) and Fig. 1]. We would then have an ionization probability of $2\times 10^{-6}$ per $K$ electron, for beta-decay of Fe. If we treat $K$ capture as another example of a sudden perturbation, the ionization yield per $K$ electron is about half that for beta-decay, since the change in the effective nuclear charge is $1-0.35=0.65$ for $K$ capture instead of one as for beta-decay, and this change enters squared. We then find a yield also about $10^{-6}$ $K$ electron ejected with energy greater than 30 kev per $K$ capture in $Fe^{55}$, in agreement with Primakoff and Porter,[17] and not inconsistent with the experiments of Porter and Hotz.‡

Madansky and Rasetti[1] confirmed the theoretical formulas for the spectrum of inner bremsstrahlung from $P^{32}$ and RaE in the 50- to 200-kev range, using a scintillation spectrometer. Their measurement at 90 kev for the RaE case lies above the smooth theoretical curve for inner bremsstrahlung by three experimental errors. If we interpret this deviation as due to $K$ x-rays from Po (the product atom), we find that the $K$ x-rays are roughly 5 percent as abundant as the inner bremsstrahlung of energy greater than 90 kev. Using their figure of $1.6\times 10^{-3}$ photon ($E\geq 90$ kev) per beta for the inner bremsstrahlung we find an x-ray yield of roughly $8\times 10^{-5}$ per beta, with an error of at least a factor of 2. Novey[2] obtains a similar x-ray yield for RaE. This number is in fortuitously good agreement with the theoretical yield $0.64/Z^2=9\times 10^{-5}$ (see Table I). (The theoretical yield would be increased by a factor of 2 or more by screening and relativistic corrections.)§

Gray[39] has set an upper limit of $10^{-3}$ $L$ x-ray per beta by absorption measurements on the photons from RaE. This is consistent with our calculated $3\times 10^{-4}$ vacancies in the $L$ shell per beta-emission (see Table I). The $L$ x-ray yield will be somewhat smaller since here the fluorescent yield is appreciably less than unity; but screening effects will more than compensate for this, giving $5\times 10^{-4}$ x-ray per beta.

In measurements on characteristic x-rays from beta- or positron-emitters use of a detector with good resolving power is essential to separate the x-rays from the continuous spectrum of inner bremsstrahlung. Ordinary absorption measurements are not good enough and the scintillation spectrometer is just about adequate. Critical absorption techniques or crystal diffraction should prove successful.

Thus for the case of beta- or positron decay, the first two of our three effects have been studied. Bruner's result on the first effect for $Sc^{44}$ is much larger than theoretical results, while Madansky and Rasetti's and Gray's results for the second effect on RaE are not inconsistent with the theoretical calculations.

Many workers have studied the $K$ and $L$ x-rays associated with the alpha-activity of $Po^{210}$. This isotope emits one nuclear gamma of energy 800 kev and intensity about $1.5\times 10^{-5}$ photon per alpha[7–9] which is in coincidence with a correspondingly lower energy alpha.[40] Even a gamma-ray of this low intensity confuses the interpretation of the $K$ x-rays, since some may be due to ionization caused by the alpha-particle, and some due to internal conversion of the nuclear gamma. Since the gamma has much lower intensity than the experimental

[38] See the end of Sec. IV for a discussion of the failure of this analogy. We hazard the guess that Bruner's sources, prepared by evaporation of an aqueous solution, exhibited "clumping"; and that the electrons are principally due to positron-electron scattering in the source.

‡ *Note added in proof:*—Primakoff and Porter include a factor for the correlation of the two $K$ electrons, and also correct a mistake in our calculation that 0.4 percent of the ejected electrons have an energy greater than 30 kev.

§ *Note added in proof:*—Howland and Rubinson (private communication) and Boehm and Wu (Bull. Am. Phys. Soc. **28**, No. 1, 40 (1953)) have both measured the yield of characteristic x-rays associated with $S^{35}$ beta-decay; and the latter group have also observed x-rays from $RaE^{210}$ and $Pm^{147}$. The measurements are in good agreement with the calculations of this paper.

[39] J. A. Gray, Phys. Rev. **55**, 586 (1939).

[40] S. De Benedetti and G. H. Minton, Phys. Rev. **85**, 944 (1952).

yield of $L$ x-rays, and its internal conversion coefficient is much less than unity, the $L$ x-rays are due almost entirely to ionization by the alpha-particle.

Curie and Joliot[5] observed $L$ and $M$ x-rays from Po, but did not detect $K$ x-rays, due to their low intensity. Zajac *et al.*[41] observed both the 800-kev gammas and x-rays of energy $84\pm4$ kev. Recently Grace *et al.*,[7] Barber and Helm[8] and Riou[9] have identified these x-rays as characteristic $K$ x-rays of lead ($K\alpha$ of 73 kev): the first group used critical absorption techniques, and the second group used energy measurements by a scintillation spectrometer. The $K$ x-rays have an intensity of $1.7\times10^{-6}$ photon per alpha, averaging the results of the last three papers. Correcting for the fluorescent yield, the vacancies in the $K$ shell are 10 percent greater, or $1.9\times10^{-6}$ per alpha. Grace *et al.* believe that the vacancies in the $K$ shell are produced mainly by internal conversion of the 800-kev gamma, while Barber and Helm believe that internal conversion produces only one-third of the $K$ shell vacancies, the remainder or $1.3\times10^{-6}$ per alpha being produced by alpha-particle ionization.

These arguments depend critically on the internal conversion coefficient of the 800-kev gammas. Grace *et al.* measure a conversion coefficient of $0.067\pm0.017$, by counting the number of conversion electrons and the number of gamma-rays. This would assign the majority of the $K$ x-rays to the internal conversion process. Alburger and Friedlander[42] obtained a rough measurement of the internal conversion coefficient by observing with a magnetic beta-spectrometer both the internal conversion electrons and the photoelectrons from an external converter: their result was 0.01 to 0.05. The angular correlation measurements for alpha and gamma of DeBenedetti and Minton[40] show that the 800-kev gamma is electric quadrupole radiation, which according to Rose *et al.*[43] has a calculated internal conversion coefficient in the $K$ shell of 0.01.

In comparison with the above results, we have calculated here $0.10\times10^{-6}$ electron vacancy per alpha, with perhaps a factor of two or three increase for relativistic and screening effects. Migdal's result[11] is 25 times as large as ours, since he does not include the reduction of the dipole transition probability due to nuclear recoil. Grace's results (a small fraction of $1.9\times10^{-6}$) could agree with our calculation, but Barber and Helm's result of $1.3\times10^{-6}$ is much larger.

An unambiguous separation of the effects of alpha-emission and internal conversion of the nuclear gamma on the $K$ electrons seems quite difficult at the present time. Measurements of electrons in the 50- to 150-kev region, should clarify this issue.[44] One should observe weak lines corresponding to the Auger electrons; and one should also observe a continuous distribution due to the effects of alpha-particle emission. This spectrum could be compared both in intensity and in shape with the theoretical result (Fig. 1).

As noted above, measurements on the $L$ x-rays are not confused by internal conversions of the nuclear gamma[45] of $Po^{210}$; however, an accurate knowledge of the fluorescent yield for the $L$ shell is necessary for their interpretation. Curie and Joliot[5] measured the $L$ x-rays with an electroscope and determined their energy by absorption measurements. They found a yield of about $4\times10^{-4}$ $L$ x-ray per alpha. Rubinson and Bernstein[6] have recently studied the $L$ x-rays with a proportional counter. They identify them by careful energy measurements as $L$ x-rays and determine the yield as: $L\alpha$, $1.67\times10^{-4}$; $L\beta$ $1.06\times10^{-4}$; $L\gamma$ $0.20\times10^{-4}$; or a total of $2.93\times10^{-4}$ photon per alpha, with an experimental uncertainty of 15 percent. They compare a concentrated and a dilute source to show that the $L$ x-rays are not produced in neighboring atoms. This yield is much greater than our calculated yield of $1.1\times10^{-4}$ $L$ vacancy per alpha using Coulomb wave functions. The calculated yield of $L$ x-rays is appreciably less than this. If, following Rubinson and Bernstein, we use Kinsey's values[46] for the fluorescent yields in the $L$ shell, $1.1\times10^{-4}$ $L$ vacancy distributed among the $L_{\text{I}}$, $L_{\text{II}}$, and $L_{\text{III}}$ shells as shown in Table IV gives an $L$ x-ray yield of $0.35\times10^{-4}$, or only 12 percent of Rubinson and Bernstein's experimental value.[47] Also see Riou[9] who confirms these experimental results. This large difference between calculations and experiment should be attributed to screening corrections and to our use of the semiconvergent expansion into multipoles. As discussed in the previous section, screening corrections increase the calculated yield by at least a factor of 3.5 and higher multipole transitions might also lead to an appreciable increase of the ionization probability for the $L$ electrons.

Macklin and Knight[10] observed x-rays from $U^{234}$ with a yield of roughly one photon per alpha-particle. The photons were identified as $L$ x-rays (of thorium or uranium) by absorption measurements. This x-ray yield is several orders of magnitude greater than the experimental x-ray yield for $Po^{210}$ and is also several orders of magnitude greater than the calculated $L$ x-ray yield due to alpha-emission. Goldhaber and McKeown[48]

[41] Zajac, Broda, and Feather, Proc. Phys. Soc. (London) **A60**, 501 (1948).

[42] D. E. Alburger and S. Friedlander, Phys. Rev. **81**, 523 (1951).

[43] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[44] Also H. Halban (private communication) suggests measurements of coincidences between $K$ x-rays and low energy electrons.

[45] But one could make an *ad hoc* assumption of *another* nuclear gamma of energy between the $K$ and $L$ binding energies, which is strongly converted in the $L$ shell. One could make a similar assumption to explain the high yield of $K$ x-rays.

[46] B. B. Kinsey, Can. J. Research **A26**, 404 (1948).

[47] By coincidence, this calculated yield is almost identical with that quoted by Rubinson and Bernstein, based on Migdal's calculations. (Incidentally, we believe Rubinson and Bernstein have misinterpreted Migdal's formula in using $v_e=Ze^2/\hbar n$, rather than as $Ze^2/\hbar$.) *Notes added in proof:*—(1) Correction of our error in the $2s$-$n'd$ calculation lowers the calculated yield to 7 percent of the experimental value. (2) Rubinson (private communication) is now convinced of our interpretation of Migdal's formula. (3) Screening corrections appear to be rather small.

[48] G. Scharff-Goldhaber and M. McKeown, Phys. Rev. **82**, 123 (1951).

studied the radiation from $U^{234}$ with a proportional counter, and identified the 17-kev gammas as $L$ x-rays of thorium. They interpret Teillac's photographic plate measurements[49] of internal conversion electrons (about 0.3 per alpha) as showing the presence of strongly converted gamma-rays of energies from 50 to 90 kev.[50] Recently Bell *et al.*[51] have observed 55-kev gammas from $U^{234}$, using a scintillation spectrometer. This confirms Goldhaber's interpretation of the $L$ x-ray yield from $U^{234}$.

In summary, experiments on RaE are consistent with the $K$ x-ray yield for beta-emission calculated in this paper, but do not verify it. Bruner's experiment on $Sc^{44}$ is inconsistent with our calculations. Grace's interpretation that the $K$ x-rays of $Po^{210}$ are mainly due to internal conversion of the 800-kev nuclear gamma is consistent with the calculations of this paper. Barber and Helm's interpretation of $1.3\times10^{-6}$ $K$ vacancy per alpha as due to alpha-emission, is much higher than the yield for $NR$ Coulomb wave functions of $0.1\times10^{-6}$. Rubinson and Bernstein and Riou's measurements of $L$ x-rays from Po give a yield about 8 times that calculated for Coulomb wave functions, much of the discrepancy being attributed to screening effects. The high x-ray yield from $U^{234}$ is due to internal conversion of a nuclear gamma. The theoretical yields will be increased appreciably by three different corrections: relativistic corrections for the $K$ shell; screening corrections and (for the case of alpha-emission) failure of the adiabatic approximation for the $L$ shell. Due to the provisional nature of some of the present experiments, and the approximate nature of the present calculation, good agreement is not to be expected as yet.

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49 J. Teillac, Compt. rend. **239**, 1056 (1950).
50 Also see reference 13, p. 35.
51 Bell, Davis, Francis, and Cassidy, Oak Ridge National Laboratory Progress Report ORNL 1164, 1951 (unpublished).

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# The Alpha-Particle Induced Phosphorescence of Silver-Activated Sodium Chloride*

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Using as primary excitants ultraviolet light and polonium alpha-particles, the phosphorescent afterglow of the double banded phosphor, silver-activated sodium chloride, has been measured as a function of the time with photosensitive Geiger counters and photomultiplier tubes. The decay curves of the phosphorescent intensity of the two bands can be represented by appreciably different power laws. Decay slopes greater in absolute magnitude than two on a $\log I-\log t$ plot have been frequently observed.

## INTRODUCTION

In several previous communications,[1,2] the writers have discussed the fluorescence and phosphorescence of NaCl-Ag irradiated by nuclear particles. The earlier reports[1,2] constitute mainly an account of how first phosphorescent emission, and later fluorescent pulses, were detected in photosensitive Geiger counters. In the course of qualitative studies of the phosphorescence of the far ultraviolet band as detected with photosensitive Geiger counters, a marked stimulation of the ultraviolet band by long wave light was noted. The soft radiations from the red and green pilot lights on the control panel of the scaling circuit gave rise to a distinct stimulation of the phosphorescent emission. It was immediately suggested[3] that NaCl-Ag might serve as a dosimeter for nuclear radiation, the phosphorescent yield under photostimulation being a measure of dosage received much earlier. However, the present discussion will concern itself mainly with a study of the normal unstimulated phosphorescence induced in samples of NaCl-Ag by alpha-particles and ultraviolet light.

Since the time of the first reports by the writers,[1-3] the study of the phosphorescence of NaCl-Ag has been extended by Furst and Kallmann[4] and by Bittman, Furst, and Kallmann.[5] The light emission has been shown to occur in two bands[2,4] centered, respectively, at 2500A and 4000A. It has also been known for some time that these two bands are emitted when ultraviolet

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1 C. E. Mandeville and H. O. Albrecht, Phys. Rev. **79**, 1010 (1950); **80**, 117, 299, and 300 (1950).
2 H. O. Albrecht and C. E. Mandeville, Phys. Rev. **81**, 163 (1951); Rev. Sci. Instr. **22**, 855 (1951).
3 C. E. Mandeville, privately circulated memorandum (September 26, 1950).
4 M. Furst and H. Kallmann, Phys. Rev. **82**, 964 (1951); **83**, 674 (1951).
5 Bittman, Furst, and Kallmann, Phys. Rev. **87**, 83 (1952).