

## Effect of Nuclear Charge on Internally Produced Pairs

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(Received April 12, 1954)

The effect of nuclear charge on internally converted pairs is investigated using the relativistic Sommerfeld-Maue solutions of the iterated Dirac equation. The orthonormal properties of these wave functions are studied and time-dependent perturbation theory is adapted to allow for their nonorthogonality. The integrals involved are studied using the Fourier transforms of the wave functions. The matrix elements are obtained in terms of one fundamental integral, evaluated by using an integral representation of Butler. The procedure is very simple and promises generalizations. The matrix elements are shown to be complex only through  $n = \mp iZ/(137v_{\pm})$ , where  $v_{\pm}$  is the velocity of the positron or the negatron and  $Z$  is the nuclear charge. The following conclusions are drawn. Firstly, the first Born approximation results when multiplied by the well-known Sommerfeld factors of the negatron and positron are accurate to a term proportional to  $|n|^2$ . For 5-percent resolutions,  $Z \leq 20$ , the kinetic energy must be more than 100 kev. Secondly, this result is valid for all electric and magnetic multipole transitions. Finally, this result is shown to apply to all transitions taking place between unbound states and to hold for higher order perturbation terms.

### I. INTRODUCTION

IT is well known that the angular distribution in internal pair production<sup>1</sup> provides a method of distinguishing between different types of nuclear transitions. This method is particularly useful at high transition energies. In this sense, it is alternative to those procedures which are more effective at lower energies, such as the use of the ratios of internal conversion coefficients for the  $K$ ,  $L$ ,  $\dots$ , etc., atomic shells. The angular distribution in internal pair production has been studied theoretically by Horton<sup>1</sup> and Rose<sup>2</sup> using the first Born approximation. In view of the recent improvements in the experimental techniques, and the subsequent applications of the theory,<sup>3</sup> a re-examination of the region of validity of the first Born approximation is overdue. For external pair production and Bremsstrahlung, the improvement of the first Born approximation, in the relativistic case, has recently been carried out by Bethe, Davies, and Maximon.<sup>4</sup>

<sup>1</sup> This possibility was first suggested to one of the present authors by R. E. Peierls. See G. K. Horton, *Proc. Phys. Soc. (London)* **60**, 457 (1948).

<sup>2</sup> M. E. Rose, *Phys. Rev.* **76**, 678 (1949). For an interesting related paper see G. Goldring, *Proc. Phys. Soc. (London)* **66**, 341 (1953). See also M. E. Rose and G. E. Uhlenbeck, *Phys. Rev.* **48**, 211 (1935), and V. B. Beristetskii and I. M. Shmushkevich, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **9**, 591 (1949).

<sup>3</sup> Internal pair production has been frequently studied experimentally. For the earlier references see G. D. Latyshev, *Revs. Modern Phys.* **19**, 132 (1947). For transitions studied via the total pair formation coefficient see E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, 1952); S. D. Bloom, *Phys. Rev.* **88**, 312 (1952); R. G. Thomas and T. Lauritsen, *Phys. Rev.* **88**, 966 (1952); and also G. Harries and W. T. Davies, *Proc. Phys. Soc. (London)* **65**, 564 (1952). The angular distribution in internal pair creation has been studied by K. Siegbahn, *Arkiv Fysik* **4**, 233 (1952), G. Harries, *Proc. Phys. Soc. (London)* **67**, 153 (1954). Paul, River, and Devons, Imperial College, have kindly informed us that they are also studying the angular distribution in internal pair creation experimentally.

<sup>4</sup> H. A. Bethe and L. C. Maximon, *Phys. Rev.* **93**, 768 (1954); Davies, Bethe, and Maximon, *Phys. Rev.* **93**, 788 (1954).

Internal and external pair production are, however, essentially different processes. Internal pair production, as a tool in nuclear spectroscopy, is of the greatest value for light nuclei and can take place independently of the nuclear electrostatic field.<sup>5</sup> External pair production, on the other hand, depends inherently on the nuclear electrostatic field and is of considerable interest for heavy nuclei. For this reason, the calculations of Bethe *et al.*<sup>4</sup> have to be valid for a much higher nuclear charge than ours. On the other hand the latter are concerned with much higher energies. External pair formation is produced by an incident plane photon wave of high energy. In internal pair formation the perturbation is caused by a relatively low-energy spherical photon wave. This makes the former problem mathematically much simpler. Furthermore, in internal pair formation, the region of large angular separation of the components of the pair is of particular interest,<sup>6</sup> whereas in the external pair formation problem the small angle approximation is of overriding importance. Considerations of this type again make the mathematical problem in the latter case much less formidable.

It is well known that the first Born approximation is valid as long as

$$|n_{\pm}| \ll 1, \quad (1)$$

where

$$n_{\pm} = Ze^2/(i\beta_{\pm}), \quad k_{\pm} = \beta_{\pm}E_{\pm}; \quad (2)$$

$Ze$  is the nuclear charge,  $ze = -e$  for the negatron and  $+e$  for the positron, and  $n \equiv n_{\pm}$  throughout. Condition (1) shows that the first Born approximation is valid,  $|n_{\pm}| \leq 0.05$ , in the shaded region of Fig. 1. We note

<sup>5</sup> It seems, however, that experiments will be carried out for a higher nuclear charge also.  $\text{ThC}''$ ,  $\text{Mn}^{56}$ , and  $\text{Co}^{60}$  have already been investigated by Siegbahn, reference 3. For a discussion see Sec. VII of this paper.

<sup>6</sup> See G. Harries and K. Siegbahn, reference 3; in the latter a  $\beta$ -spectrogoniometer is used, which automatically restricts  $\theta$  to the range  $\pi/2 \leq \theta \leq \pi$ .

that Fig. 1 demonstrates that the first Born approximation is only valid if  $Z \leq 6$  for  $E$ , the total energy, bigger than or equal to  $2mc^2$ . This conclusion is disturbing because experiments have already been performed with nuclei as heavy as  $\text{ThC}''$  with particle kinetic energies well below  $mc^2$ . Furthermore, in the experiments it is sometimes necessary<sup>7</sup> to work with rather slow positrons (negatrons). For instance, if the excited state of the nucleus under investigation is formed through  $\beta$  decay, as in the 1.74-Mev excited state of  $\text{Na}^{24}$ , Siegbahn<sup>7</sup> found it necessary to reject all electron pairs whose negatrons had an energy below the upper energy limit of the accompanying continuous  $\beta$ -ray spectrum. This makes the apparent failure of the first Born approximation (Fig. 1) all the more serious.

In this paper, we make a detailed study of the region of applicability of the first Born approximation. It will, in fact, be shown that the first Born approximation has a much greater range of validity than is suggested by (1). This is discussed in Sec. V by using the solutions of the iterated Dirac equation in the Sommerfeld-Maue approximation given in Sec. II. We use first order perturbation theory. It has been pointed out<sup>8</sup> that it is not necessarily correct to assume that first order perturbation theory, developed in the nonrelativistic approximation, is valid when using the solutions (4) of Eq. (3). In Sec. III we develop a relativistic perturbation theory adapted to overcoming the non-orthogonality difficulty encountered when using the wave function (4). The integrals that arise have been studied by a number of authors<sup>9</sup> in configuration space. In Sec. IV, we use the Coulomb wave function given by one of us<sup>10</sup> in the momentum representation, combined with an idea of Butler (see Bess<sup>9</sup>) to evaluate the relevant integral in a new way. We have given the evaluation in some detail in the hope that it will be possible to adapt the techniques to the evaluation of similar but as yet unsolved problems.<sup>11</sup> In Sec. VI, we discuss the generalization of our results to any transition taking place between unbound states in a nuclear Coulomb field. In Sec. VII, we conclude with a critical examination of the validity of our results.

<sup>7</sup> See K. Siegbahn, reference 3.

<sup>8</sup> G. K. Horton and E. Phipps, Phys. Rev. **94**, 1402 (1954). See also F. J. Dyson, Phys. Rev. **83**, 713 (1948); A. Sommerfeld, *Atombau und Spectrallinien* (F. Vieweg and Son, Braunschweig, 1939), Vol. 2, p. 412.

<sup>9</sup> A. Sommerfeld, reference 8, pp. 502 and 529. See also A. Nord-sieck, Phys. Rev. **93**, 785 (1954); L. Bess, Phys. Rev. **77**, 550 (1950).

<sup>10</sup> G. K. Horton and R. T. Sharp, Phys. Rev. **89**, 885 (1953).

<sup>11</sup> We refer, for instance, to the Coulomb excitation of nuclei by low-energy charged particles. See G. K. Horton, Birmingham Dissertation (unpublished), and K. Alder and A. Winther, Phys. Rev. **91**, 1578 (1953); K. A. Ter-Martirosyan, J. Exptl. Theoret. Phys. (U.S.S.R.) **22**, 284 (1952); C. J. Mullin and E. Guth, Phys. Rev. **82**, 141 (1951). Similar integrals arise in the theory of  $\beta$  decay and atomic and nuclear scattering.

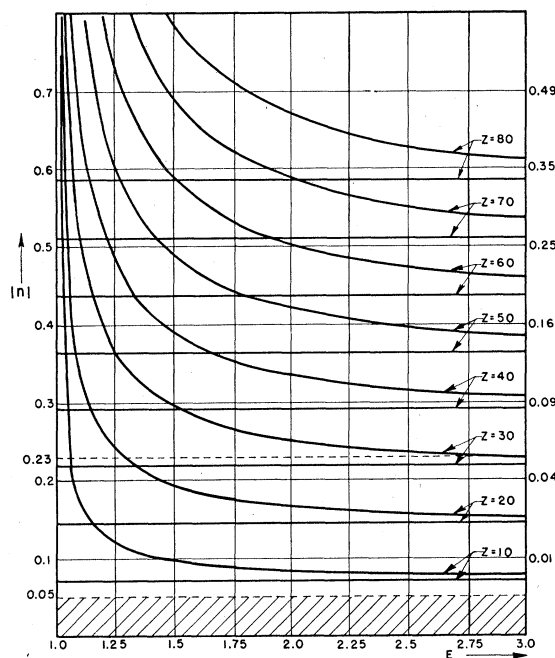


FIG. 1. The vertical scale on the left gives the values of  $|n|$  and  $|n\beta|$ . The scale on the right gives values of  $|n|^2$  and  $|n^2\beta|^2$ . The horizontal scale gives the kinetic energy in units of  $mc^2$ .

## II. WAVE FUNCTIONS AND PERTURBING POTENTIALS

It has been shown by Sommerfeld and Maue<sup>12</sup> that the normalized solution of the iterated Dirac equation,

$$\nabla^2\psi + \left\{ \left( E + \frac{Zze^2}{r} \right)^2 - m^2 \right\} \psi = \frac{zZe^2}{i} \left( \alpha \cdot \nabla - \frac{1}{r} \right) \psi, \quad (3)$$

is

$$\psi(\mathbf{r}) = (2\pi)^{-3/2} N e^{ik \cdot \mathbf{r}} \{ 1 + (i\beta/r) \alpha \cdot \nabla + O(n^2\beta^2) \} \times F(-n, 1; \rho) u(\mathbf{k}), \quad (4)$$

where

$$N^2 = 2\pi |n| / (e^{2\pi|n|} - 1),$$

$$F(-n, 1; \rho) = \frac{1}{2\pi i} \oint_{(0^+, -1^+)} t^{-n-1} (1+t)^n e^{-\rho t} dt, \quad (5)$$

$$\rho = i(kr - \mathbf{k} \cdot \mathbf{r}).$$

We discuss the orthonormal properties of (4), and, in particular, the derivation of the first relation in (4), in the first appendix.

It is well known that if the effect of an electromagnetic field on a relativistic charged particle, obeying Dirac's equation, is taken into account by perturbation theory, then the perturbation term in the Hamiltonian is

$$H' = e(\alpha \cdot \mathbf{A} + V), \quad (6)$$

<sup>12</sup> See Sommerfeld, reference 8, p. 410; W. H. Furry, Phys. Rev. **46**, 391 (1934). See also H. A. Bethe and L. C. Maximon, Secs. II and III, and G. K. Horton and R. T. Sharp, reference 10. We use natural units  $\hbar = c = 1$ .

where  $\mathbf{A}$  and  $V$  are the classical vector and scalar potential of the perturbing electro-magnetic field. For electric dipole transitions:

$$\mathbf{A} = \mathbf{B}e^{iqr}/r, \quad V = \mathbf{B} \cdot \nabla_r (e^{iqr}/iqr), \quad (7)$$

where  $\mathbf{B}$  is a unit vector oriented in the direction of the dipole and  $q$  is the frequency (or energy) associated with the nuclear transition. For other types of transitions see Sec. VI. The number of quanta emitted by an electric dipole is, in our units,

$$4q/3. \quad (8)$$

### III. PERTURBATION THEORY

We shall follow the Born approximation calculations,<sup>1,2</sup> by using first order perturbation theory in evaluating our transition probabilities. It is well known that in deriving the transition probability for a process by the usual nonrelativistic perturbation theory, the fact that the unperturbed wave functions form a complete orthonormal set is an essential requirement. It is, however, clear (see the first Appendix) that our wave functions (4), solutions of Eq. (3), do not form a complete orthonormal set of unperturbed wave functions but, as is shown in the first Appendix, satisfy

$$\int_{\Delta_-} d\mathbf{k}_- \int d\tau \psi^*(E_-, \mathbf{r}) \left[ 1 + \frac{2Ze^2}{(E_+ + E_-)r} \right] \times \psi(E_+, \mathbf{r}) = 1 \text{ or } 0, \quad (9)$$

according as  $\Delta_-$  includes  $k_+$  or not. This means that the conventional perturbation theory must be amended accordingly. Let

$$\lambda = 2Ze^2/i(E_- + E_+) = 2Ze^2/iq. \quad (10)$$

We require the solution of the time-dependent Schrödinger equation

$$i\partial\Psi/\partial t = H\Psi, \quad \text{where } H = H_0 + H', \quad (11)$$

and

$$H_0\psi(E^{(n)}, \mathbf{r}) = E^{(n)}\psi(E^{(n)}, \mathbf{r}). \quad (12)$$

Let

$$\Psi(\mathbf{r}) = \mathfrak{S}_{(n)} a_n(t) \psi(E^{(n)}, \mathbf{r}) \exp(-iE^{(n)}t), \quad (13)$$

where  $\mathfrak{S}_{(n)}$  indicates summation over the discrete and integration over the continuous states, respectively. Substituting (13) into (11) we have

$$\mathfrak{S}_{(n)} i(\partial a_n / \partial t) \psi(E^{(n)}, \mathbf{r}) \exp(-iE^{(n)}t) = \mathfrak{S}_{(n)} a_n(t) H' \psi(E^{(n)}, \mathbf{r}) \exp(-iE^{(n)}t). \quad (14)$$

Multiply (14) on the left-hand side by

$$\psi^*(E^{(m)}, \mathbf{r}) [1 + i\lambda/r] \exp(iE^{(m)}t)$$

and integrate over all space. We find

$$\partial a_m(t) / \partial t = -i \mathfrak{S}_{(n)} H_{m,n}' a_n(t) \exp[-i(E^{(n)} - E^{(m)})t], \quad (15)$$

where

$$H_{m,n}' = \int d\tau \psi^*(E^{(m)}, \mathbf{r}) [1 + i\lambda/r] H' \psi(E^{(n)}, \mathbf{r}). \quad (16)$$

Equation (16) has been given before without proof.<sup>8</sup> From this stage the theory follows the usual nonrelativistic case. Here we only give the final result. For dipole transitions, the number of pairs per nuclear transition in which  $E_+$  lies between  $E_+$  and  $E_+ + dE_+$  and hence  $E_-$  lies between  $E_-$  and  $E_- + dE_-$ , the direction of  $k_+$  lies in the element of solid angle  $d\Omega_+$ , and the direction of  $k_-$  lies in the element of solid angle  $d\Omega_-$ , is

$$P(E_+, E_-, \Omega_+, \Omega_-) dE_+ dE_- d\Omega_+ d\Omega_- \delta(E_+ + E_- - q) = [3/(32\pi^5 q)] \sum |H_{i,f}'|^2 k_+ k_- E_+ E_- \times dE_+ dE_- d\Omega_+ d\Omega_- \delta(E_- + E_+ - q), \quad (17)$$

where

$$H_{i,f}' = \int \psi_{\text{initial}}^* H' [1 + i\lambda/r] \psi_{\text{final}} d\tau. \quad (18)$$

$H'$  is given by (6) and  $\lambda$  by (10).  $E$  is the total energy of the transition. That (17) and (18) are more plausible with  $\lambda \neq 0$  may also be seen as follows. Consider<sup>13</sup> a constant perturbation  $H'$ . For this case (17) will give zero, which is reasonable physically, whereas (18) will not be zero if  $\lambda = 0$ , but proportional to  $\lambda$ .

It could be argued, of course, that it would be desirable to change as little of the exact Dirac theory as possible, and that the requirement of the orthogonality of the initial and final state is not essential. As evidence for this point of view, we note that in many calculations it has simply not been possible to orthogonalize the initial and final wave functions. Similarly, it does not follow that just because a certain form of the transition probability, (17) and (18), seems more plausible than another [(17) and (18) with  $\lambda = 0$ ], therefore we will get better agreement with experiment; especially when first order perturbation theory is used. We refer, for instance, to the work of Jackson and Schiff<sup>14</sup> on the electron capture by protons in hydrogen. In a first order perturbation calculation, these authors use initial and final state wave functions which are not orthogonal. In addition, they take into account a term in the interaction Hamiltonian which, for physical reasons, should not contribute to the transition probability and yet gives a large contribution. Their agreement with experiment is excellent.

Another procedure, possibly the most consistent of those outlined in this section, would be to use the iterated Dirac equation, including the electromagnetic potentials representing the nuclear multipole. From this equation one would then have to take the perturbing term  $H'$ , equivalent to (6) and, after devising a perturbation theory for a second order Schrödinger

<sup>13</sup> We are indebted to Dr. Harry Schiff for this remark.

<sup>14</sup> J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953); see also H. Schiff, Can. J. Phys. **32**, 393 (1954).

equation, evaluate the matrix elements with Sommerfeld-Maue wave functions. The equivalence of the two procedures has been discussed by Pauli.<sup>15</sup>

#### IV. EVALUATION OF AN INTEGRAL

The following integral will be required in Sec. V:

$$I(q) = \lim_{\delta \rightarrow 0} N_+ N_-^* \int d\tau \exp[(iq - \delta)r + i(\mathbf{k}_+ - \mathbf{k}_- + \boldsymbol{\sigma}) \cdot \mathbf{r}] F^*(-n, 1; \rho_-) \times F(-n, 1; \rho_+)/r. \quad (19)$$

Equation (19) was first evaluated by Sommerfeld<sup>16</sup> by contour integral methods. He introduced the integral representation,

$$F(-n, 1; \rho) = \frac{1}{2\pi i} \oint_{(0^+, -1^+)} dt t^{-n-1} (t+1)^n e^{-\rho t}. \quad (20)$$

The same integral was subsequently evaluated by Bess<sup>17</sup> who introduced the integral representation

$$F(-n, 1; \rho) = \frac{e^\rho}{\Gamma(1+n)} \int_0^\infty e^{-t} t^n J_0[2(\rho t)^{1/2}] dt, \quad (21)$$

with  $\text{Re}(\rho) > 0$ .

The integral will be evaluated in this section by means of the Fourier transform of

$$\psi(\mathbf{r}, \mathbf{k}) = N(2\pi)^{-3/2} e^{i\mathbf{k} \cdot \mathbf{r}} F(-n, 1; \rho), \quad (22)$$

$$= (2\pi)^{-3/2} \int d\mathbf{k}' \exp(i\mathbf{k}' \cdot \mathbf{r}) \phi(\mathbf{k}', \mathbf{k}), \quad (23)$$

where

$$\phi(\mathbf{k}', \mathbf{k}) = -\frac{N}{2\pi^2} \lim_{\epsilon \rightarrow 0} \frac{\partial}{\partial \epsilon} \frac{[\kappa^2 + (\epsilon - ik)^2]^n}{[\epsilon^2 + |\mathbf{k} - \mathbf{k}'|^2]^{1+n}}. \quad (24)$$

The method<sup>18</sup> is essentially different from that of Sommerfeld or Bess, although it has the powerful contour integral approach in common with the former.

It is well known that integrals similar to (19), occurring in related fields, have so far defied evaluation, and this new technique may suggest ways of reducing them.

<sup>15</sup> W. Pauli, *Handbuch der Physik* (Edwards Brothers, Ann Arbor, 1943), Vol. 24, No. 1, p. 239.

<sup>16</sup> See A. Sommerfeld, reference 8. The correct asymptotic behaviour for our wavefunctions demands the substitution  $n_\pm \rightarrow -n_\pm$  and  $k_\pm \rightarrow -k_\pm$  in  $I(q)$ .

<sup>17</sup> See L. Bess, reference 9.

<sup>18</sup> One of us is greatly indebted to Dr. S. T. Butler for pointing out the value of (27) when combined with (24), as well as for a most enlightening discussion on which much of the work in this section is based. We are also grateful to Dr. P. G. Rooney for a discussion on the mathematical aspects of this section.

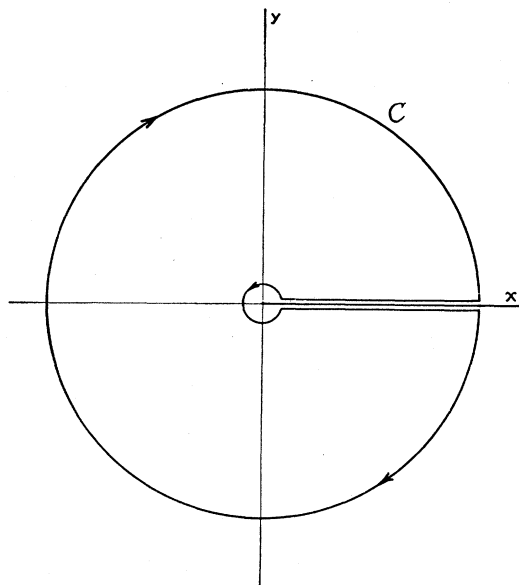


FIG. 2. Integration path used in Eq. (29).

Equation (19) can be written<sup>19</sup>

$$I(q) = \lim_{\delta \rightarrow 0} \int \psi^*(\mathbf{r}, \mathbf{k}_-) \exp[iq\mathbf{r} - \delta\mathbf{r} + i\boldsymbol{\sigma} \cdot \mathbf{r}] \times \psi(\mathbf{r}, \mathbf{k}_+) d\mathbf{r}. \quad (25)$$

By introducing (22) and (23) and carrying out the integration over configuration space,

$$I(q) = \frac{1}{(2\pi)^3} \int d\mathbf{k}' \int d\mathbf{k}'' \phi^*(\mathbf{k}') \phi(\mathbf{k}'') \frac{4\pi}{|\mathbf{k}'' - \mathbf{k}' + \boldsymbol{\sigma}|^2 - q^2}. \quad (26)$$

In order to carry out the integrations in (26) we use<sup>20</sup>

$$a^n/b^{n+1} = -(1/\pi) \sin \pi n \int_0^\infty u^n du / (a + bu), \quad (27)$$

$$a > 0, \quad b > 0, \quad -1 < \text{Re } n < 0. \quad (28)$$

It is easy to deduce from (27) that

$$a^n/b^{n+1} = -\frac{e^{-in\pi}}{2\pi i} \oint_C u^n du / (a + bu), \quad (29)$$

where  $C$  is the contour described in Fig. 2. In order to combine (29) with (24), subject to (28), it is necessary to assume that the  $n$ , defined in (2), contains an in-

<sup>19</sup> We have inserted a factor  $\exp(i\boldsymbol{\sigma} \cdot \mathbf{r})$  in  $I$ , because the trick used by Sommerfeld, Bess, and Bethe *et al.* of keeping  $\mathbf{k} = \mathbf{k}_- - \mathbf{k}_+$  together throughout the integration is not possible here. This approach was previously outlined by E. Guth and C. J. Mullin, *Phys. Rev.* **83**, 667 (1951). Their application of the method to the evaluation of quadrupole transition matrix elements in Coulomb excitation of nuclei was, however, incorrect. This was first noticed by H. McManus (private communication).

<sup>20</sup> W. Gröbner and N. Hofreiter, *Integral Tafel* (Springer Verlag, Wien und Innsbruck, 1950), Vol. II, p. 32, Eq. (9a) with  $m = \frac{1}{2}$ .

finitesimal negative real part which eventually is allowed to end to zero. The interchange of the integration and limit process is justified by the uniform convergence of (29). The result of combining (29) with (24) is

$$\phi(\kappa) = \frac{N e^{-i n \pi}}{4 \pi^3 i} \lim_{\epsilon \rightarrow 0} \frac{\partial}{\partial \epsilon} \times \oint \frac{u^n du}{[\kappa^2 + (\epsilon - i k)^2] + [\epsilon^2 + |\kappa - \mathbf{k}|^2] u}. \quad (30)$$

Equation (30) is a far more convenient form than (24) for the Fourier transform of (22), because the branch points in (24) are innocuous in (30). Conditions (28) are obviously satisfied by (30). Inserting (30) into (22), we find

$$I(q) = \frac{N_+ N_-^*}{8 \pi^6} \exp[-i \pi (n_+ - n_-)] = \frac{1}{2 \pi i} \oint u^{-n} du \frac{1}{2 \pi i} \oint v^{n+d} W(u, v), \quad (31)$$

where

$$W(u, v) = \lim_{\epsilon' \rightarrow 0} \frac{\partial}{\partial \epsilon'} \int d\kappa' \frac{V(\kappa', v)}{[\kappa'^2 + (\epsilon' + i k_-)^2] + [\epsilon'^2 + |\kappa' - \mathbf{k}_-|^2] u}, \quad (32)$$

and

$$V(\kappa', v) = \lim_{\epsilon'' \rightarrow 0} \frac{\partial}{\partial \epsilon''} \int d\kappa'' \frac{1}{\{|\kappa'' - \kappa' + \sigma|^2 - q^2\} \{[\kappa''^2 + (\epsilon'' - i k_+)^2] + [\epsilon''^2 + |\kappa'' - \mathbf{k}_+|^2] v\}}. \quad (33)$$

Equation (33) is most easily evaluated by noticing that the integrand is the product of two factors whose separate Fourier transforms are well known. If we use the convolution theorem of Fourier integrals, Eq. (33) becomes

$$V(\kappa', v) = \lim_{\epsilon'' \rightarrow 0} \frac{\partial}{\partial \epsilon''} \frac{\pi}{2(1+v)} \int \frac{d\tau}{r^2} \exp \left\{ i \left[ (ip - q)r + \left[ \kappa' - \sigma - \left( \frac{v}{1+v} \right) \mathbf{k}_+ \right] \cdot \mathbf{r} \right] \right\}, \quad (34)$$

where

$$p^2 = [k_+^2 v + (\epsilon'' - i k_+)^2 + \epsilon''^2 v] / (1+v) - v^2 k_+^2 / (1+v)^2. \quad (35)$$

As (34) depends on  $\epsilon''$  only through  $p$ , we only require

$$\lim_{\epsilon'' \rightarrow 0} p = i k_+ / (1+v), \quad \lim_{\epsilon'' \rightarrow 0} \partial p / \partial \epsilon'' = -1. \quad (36)$$

Carrying out the differentiation and limit for  $\epsilon''$  in (34) and evaluating the integral over  $\mathbf{r}$  yields

$$V(\kappa', v) = -2 \pi^2 (1+v)^{-1} \{ |\kappa' - \sigma - \mathbf{k}_+ v / (1+v)|^2 - [k_+ / (1+v) + q]^2 \}^{-1}; \quad (37)$$

$W(u, v)$  may be evaluated in an analogous manner. We only give the final result:

$$I(q) = \frac{1}{2 \pi^2} e^{-i \pi (n_+ - n_-)} N_+ N_-^* \frac{1}{2 \pi i} \oint u^{-n} (1+u)^{-1} du \times \frac{1}{2 \pi i} \oint v^{n+d} (1+v)^{-1} \left\{ \left| \mathbf{k}_+ \left( \frac{v}{1+v} \right) - \mathbf{k}_- \left( \frac{u}{1+u} \right) + \sigma \right|^2 - \left[ q - \frac{k_-}{1+u} + \frac{k_+}{1+v} \right]^2 \right\}^{-1}, \quad (38)$$

$$= \frac{1}{2 \pi^2} e^{-i \pi (n_+ - n_-)} N_+ N_-^* \times \frac{1}{2 \pi i} \oint u^{-n} \{ u [|\mathbf{k}_- - \mathbf{k}_+ - \sigma|^2 - q^2] - [(k_- - q)^2 - |\mathbf{k}_+ + \sigma|^2] \}^{-1} du \frac{1}{2 \pi i} \oint \frac{v^{n+d} dv}{v - v_0}, \quad (39)$$

where

$$v_0 = \frac{u \{ (k_+ + q)^2 - |\mathbf{k}_- - \sigma|^2 \} + \{ (k_+ - k_- + q)^2 - \sigma^2 \}}{u \{ |\mathbf{k}_- - \mathbf{k}_+ - \sigma|^2 - q^2 \} - \{ (k_- - q)^2 - |\mathbf{k}_+ + \sigma|^2 \}}. \quad (40)$$

It can be shown that  $v_0$  does not lie on the positive real axis, provided  $u$  does not lie on the negative real axis between

$$u_1 = - \{ |\mathbf{k}_+ + \sigma|^2 - (k_- - q)^2 \} / \{ |\mathbf{k}_- - \mathbf{k}_+ - \sigma|^2 - q^2 \}, \quad (41)$$

and

$$u_2 = - \{ (k_+ - k_- + q)^2 - \sigma^2 \} / \{ (k_+ + q)^2 - |\mathbf{k}_- - \sigma|^2 \}.$$

The integration over  $v$  can, therefore, be carried out by evaluating the residue of the integrand at  $v_0$ .

We remember that the integrand tends to zero along the large circle (see Fig. 2) because of the small real negative part of  $n_+$ . Hence,

$$I(q) = (2 \pi^2)^{-1} N_+ N_-^* \exp \{ -i \pi (n_+ + n_-) \} \frac{1}{2 \pi i} \oint \frac{\{ u [ (k_+ + q)^2 - |\mathbf{k}_- - \sigma|^2 ] + (k_+ - k_- + q)^2 - \sigma^2 \}^{n+d} du}{u^{-n} \{ u [ |\mathbf{k}_- - \mathbf{k}_+ - \sigma|^2 - q^2 ] - [(k_- - q)^2 - |\mathbf{k}_+ + \sigma|^2] \}^{n+1}}. \quad (42)$$

In (42), make the transformation

$$u = u_1 (1 - i) + u_2 i. \quad (43)$$

Hence,

$$I(q) = \frac{N_+ N_-^*}{2\pi^2} \frac{[|\mathbf{k}_- - \boldsymbol{\sigma}|^2 - (k_+ + q)^2]^{n_+} [|\mathbf{k}_+ + \boldsymbol{\sigma}|^2 - (k_- - q)^2]^{n_-}}{[|\mathbf{k}_- - \mathbf{k}_+ - \boldsymbol{\sigma}|^2 - q^2]^{n_+ - n_- + 1}} \frac{1}{2\pi i} \oint^{(1+, 0+)} dt (1-zt)^{-n_-} (1-t)^{n_+} t^{-n_+ - 1}. \quad (44)$$

The contour does not include the singularity at  $t=1/z$ ,  $z < 0$ , where

$$z = 1 - \frac{u_2}{u_1} = 1 + \frac{\{|\mathbf{k}_- - \mathbf{k}_+ - \boldsymbol{\sigma}|^2 - q^2\} \{(k_+ - k_- + q)^2 - \sigma^2\}}{\{|\mathbf{k}_- - \boldsymbol{\sigma}|^2 - (k_+ + q)^2\} \{|\mathbf{k}_+ + \boldsymbol{\sigma}|^2 - (k_- - q)^2\}}. \quad (45)$$

$$I(q) = \frac{N_+ N_-^*}{2\pi^2} \frac{[|\mathbf{k}_- - \boldsymbol{\sigma}|^2 - (k_+ + q)^2]^{n_+} [|\mathbf{k}_+ + \boldsymbol{\sigma}|^2 - (k_- - q)^2]^{n_-}}{[|\mathbf{k}_- - \mathbf{k}_+ - \boldsymbol{\sigma}|^2 - q^2]^{n_+ - n_- + 1}} {}_2F_1(n_-, -n_+, 1; z). \quad (46)$$

It is understood that the small negative real parts in the  $n_-$  and  $n_+$ , occurring in (46) and (45), are now allowed to tend to zero. This result agrees with that obtained previously by Sommerfeld.<sup>21</sup>

#### V. PROBABILITY FOR ELECTRIC DIPOLE TRANSITIONS

In this section we use the evaluation of  $I(q)$  presented in Sec. IV in order to evaluate the  $H_{i,f}'$  defined in (18). We need the following results:

$$\nabla_r F(-n_{\pm}, 1; \rho_{\pm}) = -(k_{\pm}/r) \nabla_{\pm} F(-n_{\pm}, 1; \rho_{\pm}), \quad (47)$$

where  $\nabla_{\pm}$  means  $\nabla_{k_{\pm}}$ .

$$\begin{aligned} -i^{\mu} I_{\mu}(q) &= \int d\tau \exp(iqr) \mathfrak{F}(\mathbf{r})/r^{\mu+1} \\ &= -i^{\mu} \{\Gamma(\mu)\}^{-1} \int_q^{\infty} (q-q')^{\mu-1} I(q') dq', \end{aligned} \quad (48)$$

where<sup>22</sup>

$$I_0(q) = I(q) = \lim_{\delta \rightarrow 0} \int d\tau \exp(iqr - \delta\tau) \mathfrak{F}(\mathbf{r})/r, \quad (49)$$

and

$$\begin{aligned} \mathfrak{F}(\mathbf{r}) &= (2\pi)^{-3} N_-^* N_+ \exp\{i(\mathbf{k}_+ - \mathbf{k}_- + \boldsymbol{\sigma}) \cdot \mathbf{r}\} \\ &\quad \times F^*(-n_-, 1; \rho_-) F(-n_+, 1; \rho_+), \end{aligned}$$

$\mu \geq 0$ , the definition of  $\mathfrak{F}(\mathbf{r})$  follows from (19). Relations (48) and (49) are easily established by an interchange in the order of integration and a use of the definition of  $\Gamma(\mu)$ . Introducing (19), (45), (46), (47), and (48) into (18), we find

$$H_{i,f}' = u^*(\mathbf{k}_-) [\boldsymbol{\eta} + \boldsymbol{\alpha} \cdot (\boldsymbol{\chi} + i\rho\mathbf{v})] eu(\mathbf{k}_+), \quad (50)$$

$$\begin{aligned} \sum |H_{i,f}'|^2 &= (E_+ E_-)^{-1} \left\{ (|\boldsymbol{\chi}|^2 + |\mathbf{v}|^2 - |\boldsymbol{\eta}|^2) (E_+ E_- + m^2 + \mathbf{k}_+ \cdot \mathbf{k}_-) + 2 \operatorname{Re}[(\boldsymbol{\eta}^* E_- + \boldsymbol{\chi}^* \cdot \mathbf{k}_-) (\boldsymbol{\eta} E_+ - \boldsymbol{\chi} \cdot \mathbf{k}_+)] \right. \\ &\quad \left. + (\mathbf{v} \cdot \mathbf{k}_-) (\mathbf{v}^* \cdot \mathbf{k}_+) + (\mathbf{v} \cdot \mathbf{k}_- \times \mathbf{k}_+) \boldsymbol{\eta}^* + (\mathbf{v} \times \boldsymbol{\chi}^* \cdot \mathbf{k}_+) E_- + (\mathbf{k}_- \times \boldsymbol{\chi} \cdot \mathbf{v}^*) E_+ \right\}. \end{aligned} \quad (54)$$

Since  $\boldsymbol{\eta}$ ,  $\boldsymbol{\chi}$ , and  $\mathbf{v}$  are complex only through  $n_+$  and  $n_-$ , it follows that (54) only contains even powers of  $n_+$

<sup>21</sup> See A. Sommerfeld, reference 8. We note the difference between the normalization of our wave function and those used by Sommerfeld. Sommerfeld neglected certain terms in his evaluation as he did not require them for his calculations. They have subsequently been given by A. Nordsieck, reference 9, whose results agree with ours also after allowing for a change in notation.

<sup>22</sup> The  $I_{\mu}$  defined by (48) and (49) does not converge for  $\mu > 1$ . However, it occurs only in conjunction with  $\boldsymbol{\sigma}$ -gradients when  $\mu > 1$  and always in such a way that the resultant integrals do converge.

where

$$\begin{aligned} \boldsymbol{\eta} &= -\mathbf{B} \cdot \{[\beta_+ \nabla_+ - \beta_- \nabla_- - (\beta_+ + \beta_-) \nabla_{\sigma}](I_1 - \lambda I_2) \\ &\quad - \nabla_{\sigma}[I_2/q - I_1 - \lambda(I_3/q - I_2)]\}, \end{aligned} \quad (51)$$

$$\begin{aligned} \boldsymbol{\chi} &= -\mathbf{B}(I_0 + \lambda I_1) - \{\beta_- \nabla_- - \beta_+ \nabla_+ + (\beta_+ + \beta_-) \nabla_{\sigma}\} \mathbf{B} \\ &\quad \cdot \nabla_{\sigma}[I_3/q - I_2 - \lambda(I_4/q - I_3)], \end{aligned} \quad (52)$$

$$\mathbf{v} = \mathbf{B} \times [-(\beta_+ \nabla_+ - \beta_- \nabla_-) + (\beta_+ + \beta_-) \nabla_{\sigma}](I_1 - \lambda I_2); \quad (53)$$

$$\rho = \begin{bmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{bmatrix}.$$

In (51), (52), and (53) it is understood that  $\boldsymbol{\sigma} \rightarrow 0$  as soon as the  $\boldsymbol{\sigma}$ -gradients have been carried out. Terms proportional to  $|n|^2 \beta^2$  have been omitted as is consistent with the approximate solution (4) of Eq. (3). It was shown in Sec. IV that  $I(q)$  is complex only through  $n_+$  and  $n_-$ . It follows from the definition (48) that the same conclusion holds for  $I_1$ ,  $I_2$ , etc. Consequently, (51), (52), and (53) are complex only through  $n_+$  and  $n_-$ . We must now carry out the summation over the spin directions of the electron and positron using the standard spur and closure theorems. These are applicable because our wave functions have the great advantage that their spinor dependence is that of the plane wave solution of Dirac's Equation. We give only the final result:

and  $n_-$ . However, as (50) and consequently (51), (52), and (53) were derived on the assumption that terms proportional to all but the first power of the fine structure constant were negligible, it is consistent with our approximation to neglect similar terms in (54). This yields the final result:

$$\begin{aligned} \sum_{\text{spin}} |H_{i,f}'|^2 &= |N_+|^2 |N_-|^2 \left\{ \sum_{\text{spin}} |H_{i,f}'|^2 \right\}_{\text{first Born approx.}} \\ &\quad + O[\max(|n|^2, |n|^4, \dots; |n|^2 \beta^2)]. \end{aligned} \quad (55)$$

This result has already been assumed, *ad hoc*, by Bloom.<sup>23</sup> The Born approximation calculations now continue as before.

## VI. HIGHER ORDER MULTIPOLE TRANSITIONS AND THE EXTENSION TO OTHER UNBOUND TRANSITIONS

In the first part of this section, we shall show that the results obtained in Sec. V for electric dipole transitions hold for all nuclear electromagnetic transitions.

It is well known that nuclear transitions differ in character, and can be classified as electric and (or) magnetic multipole transitions with different selection rules for angular momentum and parity applying to each type. A thorough discussion of this problem has been given by Blatt and Weisskopf.<sup>24</sup> Our result (55) in the electric dipole case depended essentially on the fact that  $H_{i,f}$  was complex only through  $n_+$  and  $n_-$ . Consider the case of an electric quadrupole transition; it is easily shown that

$$V = \left( \frac{\mathbf{c} \cdot \nabla_r}{iq} \right) \left( \frac{\mathbf{c} \cdot \nabla_r}{iq} \right) \frac{e^{iqr}}{r},$$

$$A = 3\mathbf{c} \left( \frac{\mathbf{c} \cdot \nabla_r}{iq} \right) \frac{e^{iqr}}{r}.$$

In other words, the electric quadrupole potentials are obtained from the electric dipole potentials, apart from a real multiplying constant, by the application of the operator  $\mathbf{c} \cdot \nabla_r / iq$ . It can now be shown very easily, by using (48) and (19), that the  $H_{i,f}$  for electric quadrupole transitions is again complex only through  $n_+$  and  $n_-$ . This argument is easily extended to cover all electric multipole transitions. The same result holds for all magnetic multipole transitions. The proof follows the electric multipole case closely.

Furthermore, we see that if the Born approximation matrix element is real, i.e., the transition takes place between two unbound states, then our result must hold in general. This result is of interest in the estimation of the errors due to the use of the Born approximation in processes like the pair formation by direct nuclear interaction in the 6.05-Mev  ${}^6_0\text{O}^+ \rightarrow 0^+$  transition in  $\text{O}^{16}$ . On the basis of our general result the error caused by the neglect of the nuclear Coulomb field should be of the order of  $\frac{1}{3}$  of 1 percent. This is rather smaller than the present experimental error.<sup>25</sup> Similar considerations apply to the 7.25-Mev  $0^+ \rightarrow 0^+$  transition in  $\text{C}^{12}$ , first observed with certainty by Harris.<sup>24</sup> The error due to the neglect of the nuclear Coulomb field is rather less than  $\frac{1}{3}$  of 1 percent.

<sup>23</sup> See S. D. Bloom, reference 3.

<sup>24</sup> J. M. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952); G. Harries, reference 3.

<sup>25</sup> Devons, Goldring, and Lindsey, Proc. Phys. Soc. (London) **67**, 134 (1954).

## VII. CONCLUSIONS

Apart from the exact calculations of Jaeger and Hulme<sup>26</sup> on the total pair production probability in lead ( $Z=84$ ), the only theoretical calculations available to compare with the growing experimental evidence are based on the first Born approximation. When the latter calculations are compared with the exact results of Jaeger and Hulme, an error of about 20 percent is found. This is not at all in agreement with (1) which suggests that the error should be of the order of 65 percent. It is, however, shown<sup>27</sup> in this paper that the first Born approximation calculation is much better than might have been expected on the basis of (1). We find that the condition for its validity is

$$|n|^2 \ll 1, \quad |n|^2 \beta \ll 1, \quad |n|^2 \beta^2 \ll 1, \quad (56)$$

rather than

$$|n_+| \ll 1, \quad |n_-| \ll 1. \quad (1)$$

The corrections to the first Born approximation are of two types. There are terms proportional to  $|n|^2 \beta^2$ , or  $(Z/137)^2$ , and there are terms proportional to  $|n|^2$  and  $|n|^2 \beta$  (we ignore terms proportional to  $|n|^4$ , etc.). As we are concerned primarily with low-energy transitions, we cannot evaluate the former terms in (55) because the wave functions (4), used in (16), are only accurate<sup>28</sup> to the order of this correction term. The latter terms correspond essentially to higher order Born approximation terms and they may, in principle, be calculated to any order. There is at present, little hope that the very complicated integrals involved can be evaluated in general.<sup>29</sup> In Appendix II we outline a procedure for taking the terms proportional to  $|n|^2$  and  $|n|^2 \beta$  into account.

It is clear that for a given nuclear charge and given positron (negatron) energy,

$$|n|^2 > |n|^2 \beta > |n|^2 \beta^2.$$

The condition  $|n|^2 \beta^2 \ll 1$  is independent of the particle energy, in contradistinction to (1), and so  $|n\beta|$  plotted against the particle energies gives straight lines. These lines, for various values of  $Z$  are shown in Fig. 1. As indicated in (55), our transition probability also contains terms proportional to  $|n|^2$ . The conditions

<sup>26</sup> J. C. Jaeger and H. R. Hulme, Proc. Roy. Soc. (London) **148**, 708 (1935).

<sup>27</sup> It has been suggested by Rose, see reference 2, that in the Born approximation the integral features of the pair production process will yield even better agreement with the exact calculations as the effect of the Coulomb field is to suppress the number of slow positrons and increase the number of fast ones. Upon integration over the energy spectrum the two effects largely cancel out. This might account for the surprising excellence of the first Born approximation. A similar effect was found by Bethe *et al.*, reference 4, for external pair production. In both processes the negatron-positron interaction has so far been ignored. That this is reasonable has been shown by R. H. Dalitz, Proc. Roy. Soc. (London) **A206**, 521 (1951).

<sup>28</sup> In the case of external pair creation, Bethe *et al.* were able to concentrate on the case of large primary energy, and so calculate terms we must neglect.

<sup>29</sup> Far less complicated integrals have so far defied evaluation; see reference 11.

$|n_+|^2 \ll 1$ ,  $|n_-|^2 \ll 1$ , give curves which depend on the energy of the particle and the nuclear charge. They are also given in Fig. 1. (right hand ordinate scale) We note that because electrons have a small rest mass the curves and the straight lines in Fig. 1 diverge only at small energies. For particle kinetic energies above one Mev and the nuclear charge less than or of the order of 30, the corrections to the first Born approximation are of the order of 5 percent ( $|n|^2 \lesssim 0.05$ , i.e.,  $|n| \lesssim 0.23$ ). In view of the status of the experimental techniques, this is probably satisfactory at present. If the particle kinetic energy is above 100 kv, a five-percent error requires the nuclear charge to be less than or of the order of twenty.

Clearly this situation is again improved if ratios of transition probabilities, say, at various angle are considered. The error term may then be expected to be of order  $\text{Max}(|n|^4, |n^2 \beta^2|)$ .

In the first Born approximation, the transition probabilities are symmetrical between the positron and negatron. As soon as the nuclear electrostatic field is no longer neglected, by the inclusion of the "Sommerfeld factors," this symmetry is absent. It is only through the presence of these factors that our results differ from those obtained in the first Born approximation calculation. It is known that they occur in many other similar processes such as the external pair production.<sup>30</sup> These factors are known to affect the transition probabilities considerably for low negatron or positron energies. In Sec. VI, we proved that condition (56) gives the region of validity for all first Born approximation calculations, in which the initial and final states concerned belong to the continuous spectrum. This is clearly not the case for the internal conversion process for which, therefore, condition (1) and not (56) is applicable.<sup>31</sup> This fact explains clearly why the first Born approximation gives results with a very much more restricted range of validity in the internal conversion than in the internal pair production process.<sup>32</sup>

Our work uses a transition matrix element that differs from that usually used by a term that arises from the non-orthonormal properties of the Sommerfeld-Mauer wave functions. We emphasize that our conclusions in this paper are independent of whether  $\lambda$  in (18) is different from zero or not. The question of what is the correct matrix element clearly requires further study.

#### APPENDIX I

Let  $\psi_+$  and  $\psi_-$  be two solutions of (3) for  $E = E_+$  and  $E_-$ , respectively. Then<sup>33</sup>

$$\psi_+^* \nabla^2 \psi_- - \psi_- \nabla^2 \psi_+^* + (E_-^2 - E_+^2)(1 + i\lambda/r) \psi_+^* \psi_- = 0. \quad (\text{A1})$$

<sup>30</sup> See. W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1947).

<sup>31</sup> S. M. Dancoff and P. Morrison, *Phys. Rev.* **55**, 122 (1939).

<sup>32</sup> Rose, Goertzel, Spinrad, Harr, and Strong, *Phys. Rev.* **76**, 1882 (1949).

<sup>33</sup> A similar procedure is sketched by Sommerfeld, reference 8, p. 412, for the case of the discrete spectrum.

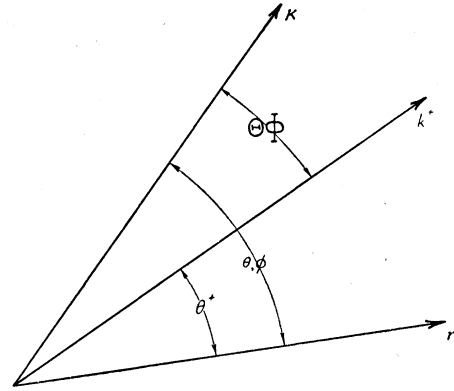


FIG. 3. Definition of angular variables in Appendix I.

Integrating over coordinate space and applying Green's Theorem, we obtain

$$\int_{\Delta_-} d\mathbf{k} \int \psi_+^* (1 + i\lambda/r) \psi_- d\tau = f(E_+, E_-), \quad (\text{A2})$$

where

$$f(E_+, E_-) = \text{Limit}_{r_0 \rightarrow \infty} \int_{\Delta_-} \frac{d\mathbf{k}}{E_-^2 - E_+^2} \times \int \left( \psi_+^* \frac{\partial}{\partial r} \psi_- - \psi_- \frac{\partial}{\partial r} \psi_+^* \right) d\sigma. \quad (\text{A3})$$

Since we are concerned with wave functions in the continuous spectrum, an integration over a spherical shell of thickness  $\Delta_-$  in  $\mathbf{k}$  space has been introduced.  $\sigma$  represents a sphere of radius  $r_0$ . Our task is to evaluate  $f(E_+, E_-)$ . The asymptotic form of the wave function (4) may be shown to be

$$\psi = \frac{N e^{\frac{1}{2} i \pi n}}{(2\pi)^{\frac{1}{2}} \Gamma(1+n)} \exp\{i\mathbf{k} \cdot \mathbf{r} + n \ln(kr - \mathbf{k} \cdot \mathbf{r})\} \times [1 + O(1/r)]. \quad (\text{A4})$$

We have omitted the spinor  $u(\mathbf{k})$ , since its effect on the orthonormalization integral would simply be to introduce an additional Kronecker delta. Inserting (A4) into (A3) and neglecting terms of order  $1/r_0$  gives

$$\begin{aligned} f(E_+, E_-) &= \text{Lim}_{r_0 \rightarrow \infty} \int_{\Delta_-} d\mathbf{k} N_+^* N_- \exp\{\frac{1}{2} i \pi (n_+ + n_-)\} \\ &\times [(2\pi)^2 \Gamma(1-n_+) \Gamma(1+n_-)]^{-1} r_0^2 \\ &\times \int_{\sigma} d\varphi d\theta \cos\theta i (k_+ \cos\theta_+ + k_- \cos\theta_-) \\ &\times \exp[i(\mathbf{k}_- - \mathbf{k}_+) \cdot \mathbf{r} + (\gamma_- - \gamma_+)], \quad (\text{A5}) \end{aligned}$$

where  $\gamma^+ = n_+ \ln(k_+ r - \mathbf{k}_+ \cdot \mathbf{r})$ ,  $\theta_+$  is the angle between  $\mathbf{k}_+$  and  $\mathbf{r}$ ,  $\gamma^- = n_- \ln(k_- r - \mathbf{k}_- \cdot \mathbf{r})$  and  $\theta_-$  is the angle between  $\mathbf{k}_-$  and  $\mathbf{r}$ . The integration over  $\theta$  and  $\phi$  can be carried out by choosing the axis of spherical coordinates in the reaction of  $\kappa$ , where

$$\kappa = \mathbf{k}_- - \mathbf{k}_+, \quad \kappa = |\mathbf{k}_- - \mathbf{k}_+|.$$

Then (see Fig. 3),

$$\begin{aligned} k_- \cos \theta_- + k_+ \cos \theta_+ &= \kappa \cos \theta + 2k_+ \cos \theta_+ \\ &= \kappa \cos \theta + 2k_+ [\cos \Theta \cos \theta \\ &\quad + \sin \Theta \sin \theta \cos(\Phi - \phi)]. \end{aligned}$$

Hence,

$$\begin{aligned} f(E_+, E_-) &= \frac{i}{(2\pi)^2} \int_{\Delta_-} \frac{d\mathbf{k}_-}{E_+^2 - E_-^2} \cdot N_+^* N_- \exp\{\frac{1}{2}i\pi(n_+ + n_-)\} \\ &\quad \times \lim_{r_g \rightarrow \infty} r_g^2 \int_{-1}^1 d \cos \theta e^{i\kappa r_g \cos \theta} (\kappa + 2k_+ \cos \Theta) \cos \theta, \quad (\text{A6}) \end{aligned}$$

where we have dropped the terms involving  $\gamma$  and will ignore the  $k_-$  dependence of  $n_-$ . This is easily justified in the limit  $r_g \rightarrow \infty$ . The integration over  $\theta$  can now be carried out. The result is

$$\begin{aligned} f(E_+, E_-) &= \frac{N_+^* N_- \exp\{\frac{1}{2}i\pi(n_+ + n_-)\}}{2\pi^2 \Gamma(1 - n_+) \Gamma(1 + n_-)} \\ &\quad \times \lim_{r_g \rightarrow \infty} \int_{\Delta_-} \frac{d\mathbf{k}_-}{E_+^2 - E_-^2} (\kappa + 2k_+ \cos \Theta) \\ &\quad \times \left( \frac{r_g}{\kappa} \cos \kappa r_g - \frac{1}{\kappa^2} \sin \kappa r_g \right). \quad (\text{A7}) \end{aligned}$$

We now introduce spherical polar coordinates in the  $\mathbf{k}_-$  integration. Carrying out the integration over the azimuthal angle yields

$$\begin{aligned} f(E_+, E_-) &= N_+^* N_- \{ \pi \Gamma(1 + n_-) \Gamma(1 - n_+) \}^{-1} \exp\{\frac{1}{2}i\pi(n_+ + n_-)\} \\ &\quad \times \lim_{r_g \rightarrow \infty} \int_{k_- = \Delta/2}^{k_- = \Delta/2} \{E_+^2 - E_-^2\}^{-1} k_-^2 dk_- \\ &\quad \times \int_{-1}^1 d \cos \beta \left[ \frac{r_g}{\kappa} \cos \kappa r_g - \frac{1}{\kappa^2} \sin \kappa r_g \right] \\ &\quad \times \left\{ \kappa + 2 \frac{k_+}{\kappa} (k_- \cos \beta - k_+) \right\}, \quad (\text{A8}) \end{aligned}$$

where  $\beta$  is the angle between  $\mathbf{k}_-$  and  $\mathbf{k}_+$ . If the variable  $\kappa$  is used instead of  $\beta$  for the angular integration,

$$\begin{aligned} f(E_+, E_-) &= \frac{N_+^* N_- \exp\{\frac{1}{2}i\pi(n_+ + n_-)\}}{\pi \Gamma(1 + n_-) \Gamma(1 - n_+)} \\ &\quad \times \lim_{r_g \rightarrow \infty} \int_{k_- = \Delta/2}^{k_- = \Delta/2} \frac{k_-}{k_+ E_+^2 - E_-^2} \\ &\quad \times \left\{ \begin{aligned} &(k_+ + k_-) \sin r_g (k_- - k_+) \\ &- (k_- - k_+) \sin r_g (k_+ + k_-) \end{aligned} \right\}, \quad (\text{A9}) \\ &= \lim_{r_g \rightarrow \infty} \left[ \int_{u_-}^{u_+} f(u) \frac{\sin u r_g}{u} du \right. \\ &\quad \left. - \int_{v_-}^{v_+} f(v) \frac{\sin v r_g}{v} dv \right], \end{aligned}$$

where

$$\begin{aligned} u &= k_- - k_+, \quad u_{\pm} = k_- - k_{\pm} \pm \Delta/2, \\ v &= k_- + k_+, \quad v_{\pm} = k_- + k_{\pm} \pm \Delta/2, \end{aligned}$$

$$f(0) = \{N_+^* N_- \exp[\frac{1}{2}i\pi(n_- + n_+)]\} \times \{\pi k_+ \Gamma(1 + n_-) \Gamma(1 - n_+)\}^{-1}.$$

Using

$$\int_{-\infty}^{\infty} (\sin u / u) du = \pi,$$

we find that

$$f(E_+, E_-) = |N_+|^2 e^{i\pi n_+} / |\Gamma(1 + n_+)|^2, \text{ or } 0, \quad (\text{A10})$$

according as  $\Delta_-$  includes  $\mathbf{k}_+$  or not. Hence the normalizing condition,

$$f(E_+, E_+) = 1, \quad (\text{A11})$$

yields

$$|N_+|^2 = 2\pi |n_+| / (e^{2\pi |n_+|} - 1). \quad (\text{A12})$$

Equations (A2), (A10), (A11), and (A12) may be compared with the nonrelativistic normalization condition,

$$\int_{\Delta_-} d\mathbf{k}_- \int \psi_+^* \psi_- d\tau = 1 \text{ or } 0, \quad (\text{A13})$$

according as  $\Delta_-$  contains  $\mathbf{k}_+$  or not. For the  $\psi$  in (A13) the solution of the nonrelativistic Schrödinger equation with a Coulomb potential, the normalizing constant may be shown to be the same as (A12). This has probably obscured the essential difference between (A2), (A10), (A11), and (A12) on the one hand, and (A13) combined with (A12) on the other. It is clear that if we carry out the transition to the nonrelativistic limit,  $c \rightarrow \infty$  or  $\lambda \rightarrow 0$ , (9) goes directly over into (A13).

## APPENDIX II

In this appendix we show how  $I_\mu$  can be evaluated in detail to order  $|n|^2$  even if  $\mu > 0$ . Consider, for example,  $I_1$ .

$$I_1 = (2\pi)^{-3} N_-^* N_+ \int d\tau \exp\{i(\mathbf{k}_+ - \mathbf{k}_- + \boldsymbol{\sigma}) \cdot \mathbf{r} + iqr\} \\ \times F^*(-n_-, 1; \rho_-) F(-n_+, 1; \rho_+) r^{-2}.$$

We now use the fact that

$$F(-n, 1; \rho) = 1 - n \text{Ei}(\rho) + O(|n|^2),$$

where  $\text{Ei}(\rho)$  is the exponential integral function. It is easily shown that correct to order  $|n|^2$ ,

$$I_1 = \int d\tau \exp\{i(\mathbf{k}_+ - \mathbf{k}_- + \boldsymbol{\sigma}) \cdot \mathbf{r} + iqr\} \\ \times [F^*(-n_-, 1; \rho_-) + F(-n_+, 1; \rho_+) \\ + |n_+ n_-| \text{Ei}(\rho_+) \text{Ei}^*(\rho_-) - 1] r^{-2}.$$

PHYSICAL REVIEW

VOLUME 96, NUMBER 4

NOVEMBER 15, 1954

Decay of  $\text{UX}_1$ ,  $\text{UX}_2$ , and  $\text{UZ}^\dagger$ 

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(Received March 31, 1954; revised manuscript received August 20, 1954)

The decay of  $\text{Th}^{234}(\text{UX}_1)$  and  $\text{Pa}^{234}(\text{UX}_2 \text{ and } \text{UZ})$  has been investigated by means of a coincidence scintillation spectrometer. Three gamma rays at 29, 63, and 92 kev were found in  $\text{UX}_1$ . Four gamma rays at 250, 750, 1000, and 1810 kev were found in  $\text{UX}_2$ , and four gamma rays at 250, 760, 910, and 1680 kev were observed in  $\text{UZ}$ . The coincidence relations between the gamma rays, and also between the gamma rays and the beta radiation, have been investigated. A decay scheme is proposed and spin and parity assignments have been made for some of the levels of  $\text{Pa}^{234}$  and  $\text{U}^{234}$ .

THE beta-emitting substance in natural uranium was isolated by Crookes<sup>1</sup> as early as 1900 and called UX. Later on Fajans and Göhring<sup>2</sup> showed that this substance is a radioactive equilibrium of two isotopes,  $\text{UX}_1$  and  $\text{UX}_2$ , in modern nomenclature  $\text{Th}^{234}$  and  $\text{Pa}^{234}$ , respectively. Hahn<sup>3</sup> showed that  $\text{Pa}^{234}$  decays with two different half-lives and that both activities emit beta and gamma rays. The long-lived activity was given the symbol UZ. Actually,  $\text{UX}_2$  and UZ were the first example of an isomeric pair.

The radiation from  $\text{UX}_1$ ,  $\text{UX}_2$ , and UZ has been studied extensively for a long time. At first absorption technique was used, but more recently the beta radiation has been investigated by means of magnetic spectrometers, and is therefore fairly well known. Almost nothing is known about the gamma radiation, however, and it has been difficult to set up a consistent decay scheme. It seemed therefore worthwhile to make a complete reinvestigation of the radiation from  $\text{UX}_1$ ,  $\text{UX}_2$  and UZ, using a coincidence scintillation spectrometer.

The apparatus used in this work has been described elsewhere.<sup>4</sup> It consists of two scintillation spectrometers.

Each one can be used as a beta- or gamma-ray spectrometer, depending on the crystal material. The pulses from the first spectrometer are led to a single channel analyzer, which selects pulses corresponding to a certain energy-range. The pulses from the second spectrometer are displayed on an oscilloscope screen and are recorded by photographic methods. The oscilloscope sweep is triggered by coincidences between the selected pulses from the first spectrometer and pulses from the second spectrometer. Hence, a recorded pulse distribution shows the spectrum of the radiation which is in coincidence with the radiation component selected by the analyzer. One can, for example, select beta rays within a certain energy range and study the corresponding coincidence gamma-ray spectrum. It is also possible to select a certain gamma ray and record the corresponding beta component. One can finally study gamma-gamma coincidences by selecting a certain gamma ray and recording the corresponding coincidence gamma spectrum. Hence, this apparatus makes possible a complete investigation of the coincidence relations in a radioactive decay.

The radioactive material used for making sources was isolated from natural uranium by an ether-water extraction method. It was precipitated as the fluoride with lanthanum as the carrier. After a short while it contains  $\text{UX}_1$ ,  $\text{UX}_2$ , and UZ in equilibrium. The work on  $\text{UX}_1$  and  $\text{UX}_2$  has been done with sources made up of this material. The activity of UZ is so low that it does not interfere with the measurements on  $\text{UX}_1$  and  $\text{UX}_2$ . The radiation of  $\text{UX}_1$  is of low energy, that of

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<sup>2</sup> K. Fajans and O. Göhring, Naturwiss. **1**, 399 (1913).

<sup>3</sup> O. Hahn, Naturwiss. **9**, 84 (1921).

<sup>4</sup> S. A. E. Johansson, Ames Laboratory Report ISC-431 (unpublished).