# Elastic Scattering of Yukawa Particles. I

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Elastic scattering of the heavy particle obeying Bose statistics is examined by the relativistically invariant equations recently developed by Proca and by Dirac. Perturbation calculus of the "variation of constants" type is applied. For the evaluation of certain sums occurring in the scattering cross sections, destruction operators are constructed similar to those first developed by Weisskopf for the Dirac equation. While according to Mott's calculation of the scattering of electrons a polarization effect does not occur until the second approximation, such an effect appears in our case even in the first approximation. The second approximation will be treated in a later paper.

### Ι

**TITHIN** the last year a heavy particle possessing spin unity and obeying Bose statistics has been introduced by various authors<sup>1</sup> with considerable success for the explanation of nuclear forces. Originally this particle was thought to be of the Pauli-Weisskopf type possessing spin zero,<sup>2</sup> but the resulting nuclear forces proved to be of the wrong sign.<sup>3</sup> Thus for the first time these elementary particles find application whose wave equations belong to the family recently developed by Dirac,<sup>4</sup> for the case of  $s > \frac{1}{2}$ . While the comparative simplicity of the wave equations of such particles seems to demolish the unique position of the wave equation of particles with  $s=\frac{1}{2}$ , the closer analysis of these new equations reveals that certain simplifying facts-e.g. the possibility of the construction of only one stress-energy tensor<sup>5</sup>---are common only to particles of spin 0,  $\frac{1}{2}$  and 1. Thus one is perhaps justified in conjecturing that only these latter particles will find permanent places in the theory. In this connection it is of interest to note that the wave equations for particles with spin 1 have been found and

discussed independently although from a different point of view, by Proca.6

The various effects occurring when such particles interact with matter, for instance with protons, are bremstrahlung, inelastic scattering through exchange, and purely elastic Rutherfordian scattering. The first two effects have been discussed by the authors mentioned under reference 1. The third effect will be treated in this paper. Although we regard it as improbable that this elastic scattering will ever be separated from the other two effects, it seems to us necessary to understand quantitatively all effects through which a heavy particle can interact with matter. Furthermore, since spin terms are of considerable influence upon the result, it may perhaps be possible in the future to decide between the two alternatives of s=0 or s=1 by performing a scattering experiment.

The corresponding computation for electrons was carried out in considerable detail by Mott<sup>7</sup> and was later very much simplified by Sauter.8 In this latter paper a Born approximation was used and the calculations were carried out leaving the scalar force field through which the impinging particle and the nucleus interact, completely general. By not specializing this general field to Coulomb interaction, until the final result, it was possible to avoid all convergence difficulties. In this way the first Born approximation gives-

<sup>&</sup>lt;sup>1</sup>Yukawa, Proc. Phys. Math. Soc. Japan 17, 48 (1935); Yukawa and Sakata, Proc. Phys. Math. Soc. Japan 19, 1084 (1937); Yukawa, Sakata and Taketani, Proc. Phys. Math. Soc. Japan 20, 319 (1938); Kemmer, Proc. Phys. A166, 127 (1938); Fröhlich, Heitler and Kemmer, Proc. Roy. Soc. A166, 154 (1938); Bhabha, Proc. Roy. Soc. A166, 501 (1938); Heitler, Proc. Roy. Soc., A166, 529 (1938). <sup>2</sup> Pauli and Weisskopf, Helv. Phys. Acta 7, 709 (1934).

<sup>&</sup>lt;sup>8</sup> Third and fourth paper of reference 1

<sup>&</sup>lt;sup>4</sup> Dirac, Proc. Roy. Soc. A155, 447 (1936).

<sup>&</sup>lt;sup>5</sup> Fierz, Helv. Phys. Acta 1938, in press.

<sup>&</sup>lt;sup>6</sup> Proca, J. de phys. et rad. 7, 347 (1936).

<sup>.</sup> Mott, Proc. Roy. Soc. A124, 425 (1929); 135, 7 N. F 429 (1932). <sup>8</sup> F. Sauter, Ann. d. Physik 18, 61 (1933).

except for relativistic corrections—the Rutherford term, while polarization terms appear but in the second approximation.

In the following sections the same treatment is applied to the Dirac-Proca equations for particles of spin unity. In Section II the unperturbed wave functions are discussed and various preparatory computations are carried out. In order to exploit fully the orthogonality properties of the eigenfunctions, we shall find it necessary to construct operators analogous to the destruction or annihilation operators which Weisskopf<sup>9</sup> and others have set up for the Dirac equations. In Section III perturbation theory of the variationof-constants type is developed and expressions for the cross section of scattered particles are obtained in first and second approximation. In Section IV the first approximation is evaluated and discussed. A report on the evaluation of the second approximation will be given in a later paper.

Π

(a) In Yukawa's notation the Dirac-Proca equations are

$$\begin{pmatrix} \frac{1}{c} \frac{\partial}{\partial t} + \frac{i}{\hbar c} V \end{pmatrix} \mathbf{F} - \operatorname{curl} \mathbf{G} - K \mathbf{U} = 0,$$

$$\begin{pmatrix} \frac{1}{c} \frac{\partial}{\partial t} + \frac{i}{\hbar c} V \end{pmatrix} \mathbf{U} + \operatorname{grad} U_0 + K \mathbf{F} = 0,$$

$$\operatorname{div} \mathbf{F} + K U_0 = 0,$$

$$(2)$$

С

$$\operatorname{url} \mathbf{U} - K\mathbf{F} = 0.$$

V(x, y, z) is the potential energy through which the (stationary) nucleus interacts with the arriving particle.  $K = Mc/\hbar$  is its reciprocal Compton wave-length. The wave functions **F**, **G**, **U** are complex vectors, while  $U_0$  is a complex scalar. The wave equations (1), (2) are relativistically invariant, but as we shall not make use of this property it does not seem necessary to introduce a four-dimensional index notation. Suffice it to say that actually **F** and **G** form an antisymmetric tensor, and **U** and  $U_0$  a four-vector.

In order to get rid of Eqs. (2) which play the role of accessory conditions, we eliminate **G** and

 $U_0$  with the result:

$$K\left(\frac{1}{c}\frac{\partial}{\partial t} + \frac{i}{\hbar c}V\right)\mathbf{F} - \text{grad div }\mathbf{U} + \Delta\mathbf{U} - K^{2}\mathbf{U} = 0, \quad (3)$$

$$K\left(\frac{1}{c}\frac{\partial}{\partial t}+\frac{i}{\hbar c}V\right)\mathbf{U}-\text{grad div }\mathbf{F} + K^{2}\mathbf{F}=0.$$
(4)

In this form each equation contains a first derivative with respect to the time which is just the form required by transformation theory. We are now ready to apply the method of the variation of constants, regarding V as perturbation parameter.

An alternative approach is to eliminate  $\mathbf{F}$  in Eq. (3) by means of Eq. (4), then to assume a definite stationary time dependence with an energy E. Then equations of the following type result:

 $\{\Delta + (E/\hbar c)^2 - K^2\}$  **U** = linear vector functions of **U** and of **F** whose coefficients involve up to second derivatives of *V*. (5)

Similarly for **F**. These Poisson-type equations may then be solved by successive approximations in the familiar way by means of Green functions.<sup>10</sup> Because of the complicated structure of the right side of the above equation it was found to be simpler to work with (3) and (4).

(b) We now have to study the unperturbed system in some detail. The unperturbed wave vectors i.e., the solutions of (3) and (4) for V=0 we shall write  $_{0}\mathbf{U}$  and  $_{0}\mathbf{F}$ . We then enter these equations with the following plane wave expressions

$${}_{0}\mathbf{U} = (2\pi)^{-\frac{3}{2}}\mathbf{u} \exp\left[i(\mathbf{k}\cdot\mathbf{x}) - iEt/h\right],$$
  
$${}_{0}\mathbf{F} = (2\pi)^{-\frac{3}{2}}\mathbf{f} \exp\left[i(\mathbf{k}\cdot\mathbf{x}) - iEt/h\right].$$
(6)

**u** and **f** are as yet unknown vectorial wave amplitudes which may yet depend upon **k**, the wave vector, and upon E, the energy. The secular determinant for E can be calculated:

$$[(E/\hbar c)^2 - K^2 - k^2]^3 = 0.$$
(7)

It has two triple roots  $E/\hbar c = \pm (K^2 + k^2)^{\frac{1}{2}}$  which

906

<sup>&</sup>lt;sup>9</sup> Weisskopf, Zeits. f. Physik 89, 27 (1934).

<sup>&</sup>lt;sup>10</sup> This is the original method of Born and Wentzel. See Sommerfeld, *Wave Mechanics*, p. 192, also *Handbuch der Physik*, Vol. 24<sub>1</sub>, p. 712–721.

correspond to the states of positive and negative energy. We shall distinguish between these in the following manner: Let

$$\epsilon = + (K^2 + k^2)^{\frac{1}{2}} \tag{8}$$

be the positive value of the energy divided by  $\hbar c$ . Let  $\eta$  be a quantum number which is capable of only the values +1 and -1. We shall then write the argument of the exponential function in (6)

$$i(\mathbf{k} \cdot \mathbf{x}) - ic\eta \epsilon t \quad \text{with} \quad \eta = \pm 1.$$
 (9)

The fact that each root of (7) is threefold, can be described, as has been pointed out by various authors, by ascribing a longitudinal and two transverse normal vibrations to both  $_0$ **U** and  $_0$ **F**. In accordance with this fact it is convenient to introduce a system of mutually perpendicular unit vectors  $\mathbf{e}(\boldsymbol{p}, \mathbf{k})$  of the following kind:<sup>11</sup>

$$\mathbf{e}(0,\mathbf{k}) = \mathbf{k}/|k|, \qquad (10)$$

$$(\mathbf{e}^*(\boldsymbol{p}, \mathbf{k}) \cdot \mathbf{e}(\boldsymbol{p}', \mathbf{k})) = \delta_{p p'}.$$
 (11)

The unit vectors  $\mathbf{e}(\mathbf{I})$ ,  $\mathbf{e}(-\mathbf{I})$  are evidently complex, in particular

$$\mathbf{e}^*(\mathbf{I}) = \mathbf{e}(-\mathbf{I}).$$

They correspond to right or left circular polarization. One can also use a system of real unit vectors, namely

$$\mathbf{e}(1) = 2^{-\frac{1}{2}} (\mathbf{e}(\mathbf{I}) + \mathbf{e}(-\mathbf{I})),$$
  
$$\mathbf{e}(-1) = 2^{-\frac{1}{2}} i^{-1} (\mathbf{e}(\mathbf{I}) - \mathbf{e}(-\mathbf{I})),$$
  
(12)

which correspond to linear polarization. We shall for the present use the complex system.

Utilizing the notation defined by Eqs. (8) to (11) we may now refine our Ansatz (6), to:

$${}_{0}\mathbf{U}(\eta, p, \mathbf{k}; \mathbf{x}, t) = (2\pi)^{-\frac{3}{2}}\mathbf{u}(\eta, p, \mathbf{k})$$

$$\times \exp\left[i(\mathbf{k} \cdot \mathbf{x}) - ic\eta\epsilon t\right],$$

$${}_{0}\mathbf{F}(\eta, p, \mathbf{k}; \mathbf{x}, t) = (2\pi)^{-\frac{3}{2}}f(\eta, p, \mathbf{k})$$

$$\times \exp\left[i(\mathbf{k} \cdot \mathbf{x}) - ic\eta\epsilon t\right],$$
(13)

with  $\eta = \pm 1$ ;  $p = \pm I$  or 0. The amplitude vectors **u** and **f** have yet to be determined. Entering (3) and (4) for the case of V=0 and using (8) we

ve have:

$$-i\eta\epsilon K\mathbf{f}(\eta p\mathbf{k}) + \mathbf{k}(\mathbf{k} \cdot \mathbf{u}(\eta p\mathbf{k})) -\epsilon^{2}\mathbf{u}(\eta p\mathbf{k}) = 0,$$
(14)  
$$-i\eta\epsilon K\mathbf{u}(\eta p\mathbf{k}) + \mathbf{k}(\mathbf{k} \cdot \mathbf{f}(\eta p\mathbf{k})) + K^{2}\mathbf{f}(\eta p\mathbf{k}) = 0.$$

(Round brackets and dot signify the scalar product.) This system decomposes into three pairs of equations as one sees upon putting

$$\mathbf{u}(\eta, p, \mathbf{k}) = u(\eta, p, k)\mathbf{e}(p, \mathbf{k}),$$
  
$$\mathbf{f}(\eta, p, \mathbf{k}) = f(\eta, p, k)\mathbf{e}(p, \mathbf{k})$$
(15)

and considering (10) and (11). We will write the solutions

$$u(\eta pk) = K \epsilon \Delta(\eta pk),$$
  

$$f(\eta pk) = i\eta (K^2 + p^2 k^2) \Delta(\eta pk)$$
(16)

where  $\Delta(\eta pk)$  is an as yet arbitrary multiplicative constant.

(c) We now have to study the orthogonality properties of the unperturbed solutions (13), (15), (16) and obtain a convenient normalization which will determine  $\Delta(\eta pk)$ . For this purpose it is necessary to derive the continuity equation. Multiply Eq. (3) scalarly with **U**\*, then Eq. (4) with  $-\mathbf{F}^*$  and add. Then subtract from this expression its complex conjugate. The result can be written

$$c^{-1}\dot{\boldsymbol{\rho}} + \text{div } \mathbf{j} = 0, \quad \boldsymbol{\rho} = i^{-1} \{ (\mathbf{U}^* \cdot \mathbf{F}) - (\mathbf{F}^* \cdot \mathbf{U}) \}. \quad (17)$$

The expression for j we shall not write down, as we are not going to make any use of it. It is important to be aware of the fact that this is really the continuity equation and not perhaps the time component of the equation expressing the conservation of energy and momentum.

Equation (17) furnishes in the usual fashion an orthogonality condition. When substituting in the expression for  $\rho$  the undisturbed eigenfunctions, viz.  $_{0}\mathbf{U}^{*}(\eta, \rho, \mathbf{k}; \mathbf{x}, t)$  and  $_{0}\mathbf{F}^{*}(\eta, \rho, \mathbf{k}; \mathbf{x}, t)$ for  $\mathbf{U}^{*}$  and  $\mathbf{F}^{*}$ , and  $_{0}\mathbf{U}(\eta', \rho', \mathbf{k}'; \mathbf{x}, t)$  and  $_{0}\mathbf{F}(\eta', \rho', \mathbf{k}'; \mathbf{x}, t)$  for  $\mathbf{U}$  and  $\mathbf{F}$ , integration over  $\mathbf{x}$ space gives the  $\delta$  function  $\delta(\mathbf{k}'-\mathbf{k})$ . For the remaining expression, i.e., the  $\rho$  symbol formed with the vector amplitudes (15), we introduce because of its frequent reappearance the sym-

<sup>&</sup>lt;sup>11</sup> See H. A. Kramers, Quantentheorie des Elektrons und der Strahlung, Vol. 2, p. 259.

bol  $\bar{\rho}$ :

$$(\eta' p' \mathbf{k}' | \bar{\rho} | \eta p \mathbf{k}) = i^{-1} (u_{\alpha}^{*} (\eta' p' \mathbf{k}') f_{\alpha} (\eta p \mathbf{k}) - f_{\alpha}^{*} (\eta' p' \mathbf{k}') u_{\alpha} (\eta p \mathbf{k})). \quad (18)$$

The Greek indices signify (cartesian) vector (or tensor) components; doubly occurring Greek indices are to be summed from 1 to 3. For  $\mathbf{k} = \mathbf{k}'$  the above quantity vanishes because of the orthogonality of the  $\mathbf{e}(p, \mathbf{k})$  vectors (Eq. (11)) except if p = p'. In this case, one has, using (16):

$$\begin{aligned} (\eta', p, \mathbf{k} | \bar{\rho} | \eta, p, \mathbf{k}) &= \epsilon K (K^2 + p^2 K^2) \\ &\times (\eta' + \eta) \Delta^*(\eta', p, k) \Delta(\eta p k), \end{aligned}$$

which vanishes unless  $\eta' = \eta$ . We can use this fact for the determination of  $\Delta(\eta pk)$ , by normalizing  $(\eta p \mathbf{k} | \bar{p} | \eta p \mathbf{k})$  to have the value  $\eta$ . This leads, except for an arbitrary phase factor, to the following expression:

$$\Delta(\eta p k) = \left[ 2\epsilon K (K^2 + p^2 k^2) \right]^{-\frac{1}{2}}.$$
 (19)

We summarize the above consideration by the orthogonality relation:

$$\langle \eta' p' \mathbf{k}' | \bar{\rho} | \eta p \mathbf{k} \rangle = \eta \delta_{\eta \eta'} \delta_{p p'}.$$
<sup>(20)</sup>

The fact that the charge is not positive definite is completely analogous to the case of s=0 i.e., of the scalar Gordon-Klein-Schrödinger equation. For this latter equation this was first pointed out by Pauli and Weisskopf.<sup>2</sup>

The converse orthogonality relations are obtained in the following manner: Multiply (20) by  $\eta' f_{\beta}(\eta' p' \mathbf{k})$  and sum over  $\eta'$  and p'.  $\beta$  indicates any cartesian component. The following relations result:

$$\sum_{\eta,p} \eta f_{\alpha}^{*}(\eta p \mathbf{k}) f_{\beta}(\eta p \mathbf{k}) = 0,$$
  
$$\sum_{\eta,p} \eta u_{\alpha}^{*}(\eta p \mathbf{k}) u_{\beta}(\eta p \mathbf{k}) = 0,$$
 (21)  
$$\sum_{\eta,p} \eta u_{\alpha}^{*}(\eta p \mathbf{k}) f_{\beta}(\eta p \mathbf{k}) = i \delta_{\alpha\beta}.$$

(d) The completeness relation may be easily derived. A general solution of the (unperturbed) wave equations (3) and (4) is:

$$\mathbf{U}(\mathbf{x}, t) = \sum_{\eta, p} \int d\mathbf{k} A(\eta, p, \mathbf{k}) \,_{0} \mathbf{U}(\eta, p, \mathbf{k}; \mathbf{x}, t),$$

$$\mathbf{F}(\mathbf{x}, t) = \sum_{\eta, p} \int d\mathbf{k} A(\eta, p, \mathbf{k}) \,_{0} \mathbf{F}(\eta, p, \mathbf{k}; \mathbf{x}, t).$$
(22)

 $d\mathbf{k}$  stands for  $dk_1dk_2dk_3$ , **x** for (x, y, z). We compute the Integral

$$\int d\mathbf{x} i^{-1} \{ (\mathbf{U}^* \cdot \mathbf{F}) - (\mathbf{F}^* \cdot \mathbf{U}) \}$$

Introducing into it (22) and applying (1) **k** orthogonality, (2) p orthogonality, and (3)  $\eta$  orthogonality according to (20) we obtain

$$\int d\mathbf{x} i^{-1} \{ (\mathbf{U}^* \cdot \mathbf{F}) - (\mathbf{F}^* \cdot \mathbf{U}) \}$$
$$= \int d\mathbf{k} \sum_{\eta, p} \eta |A(\eta, p, \mathbf{k})|^2. \quad (23)$$

(e) In later sections we shall make considerable use of so-called "destruction operators" or "annihilation operators" analogous to those introduced by Weisskopf<sup>12</sup> into the theory of the Dirac electron. There are two kinds of destruction operators. The first kind has the eigenvalue +1 for a state with a definite polarization quantum number p and zero for states with any other polarization quantum number. The second kind has the eigenvalue +1 for a state of positive energy (i.e., with  $\eta = +1$ ) and zero for the state with negative energy.

We shall first deal with the former kind. Omitting for the moment the quantum number  $\eta$  which is of no importance, we have because of (15):

$$\frac{i}{|k|} [\mathbf{k} \times \mathbf{u}(p)] = p \mathbf{u}(p);$$
  
eigenvalues +1, 0, -1,  
$$-\frac{1}{k^2} [\mathbf{k} \times [\mathbf{k} \times \mathbf{u}(p)]] = p^2 \mathbf{u}(p);$$
  
eigenvalues +1, 0, +1,

We might have written f(p) as well; it is immaterial upon which amplitude vector we are operating. With these two operators and the identity operator one can construct three operators for which the relation holds:

$$D_{\alpha\beta}(p_0, \mathbf{k})u_{\beta}(p, k) = \delta_{p, p_0}u_{\alpha}(p, \mathbf{k}).$$
(24)  
These are:

$$D_{\alpha\beta}(\pm \mathbf{I}, \mathbf{k}) = \pm \frac{1}{2i|k|} \epsilon_{\alpha\beta\gamma} k_{\gamma} + \frac{1}{2} \delta_{\alpha\beta} - \frac{1}{2k^2} k_{\alpha} k_{\beta},$$

$$D_{\alpha\beta}(0, \mathbf{k}) = \frac{1}{k^2} \mathbf{k}_{\alpha} \mathbf{k}_{\beta}.$$

$$(25)$$

$$\frac{D_{\alpha\beta}(0, \mathbf{k}) = \frac{1}{k^2} \mathbf{k}_{\alpha} \mathbf{k}_{\beta}.$$

$$(25)$$

908

 $\epsilon_{\alpha\beta\gamma}$  is the familiar antisymmetric tensor (Levi-Civita symbol).

To construct the second kind of operators we shall go back to Eqs. (14). Adding to the first of these  $-i\epsilon K \mathbf{f}(\eta)$  and dividing by  $-2i\epsilon K$  we have:

$$(i/2K\epsilon)(\epsilon^{2}\delta_{\alpha\beta}-k_{\alpha}k_{\beta})u_{\beta}(\eta)+\frac{1}{2}f_{\alpha}(\eta)$$
$$=\frac{1}{2}(\eta+1)f_{\alpha}(\eta). \quad (26_{1})$$

The right-hand side has the factor 1 if  $\eta = +1$ and zero if  $\eta = -1$ . Similarly we get from the second equation (14):

$$-(i/2K\epsilon)(K^{2}\delta_{\alpha\beta}+k_{\alpha}k_{\beta})f_{\beta}(\eta)+\frac{1}{2}u_{\alpha}(\eta)$$
$$=\frac{1}{2}(\eta+1)u_{\alpha}(\eta). \quad (26_{2})$$

We introduce the abbreviations:

$$Y_{\alpha\beta}(\mathbf{k}) = (i/2K\epsilon) (\epsilon^2 \delta_{\alpha\beta} - k_\alpha k_\beta),$$
  

$$Z_{\alpha\beta}(\mathbf{k}) = (1/2iK\epsilon) (K^2 \delta_{\alpha\beta} + k_\alpha k_\beta).$$
(27)

III

(a) We shall now apply perturbation theory to (3) and (4). We develop the eigenvectors of the perturbed problem in terms of the unperturbed eigenvectors analogous to (22). Introducing these expressions into (3) and (4) we obtain:

$$\sum_{\eta,p} \int d\mathbf{k} \{ c^{-1} A(\eta p \mathbf{k}; t) + i(\hbar c)^{-1} A(\eta p \mathbf{k}; t) V(\mathbf{x}) \}_0 \mathbf{F}(\eta p \mathbf{k}; \mathbf{x}t) = 0,$$
  
$$\sum \int d\mathbf{k} \{ c^{-1} A(\eta p \mathbf{k}; t) V(\mathbf{x}) \}_0 \mathbf{F}(\eta p \mathbf{k}; \mathbf{x}t) = 0,$$

$$\sum_{\eta,p} d\mathbf{k} \{ \}_{0} \mathbf{U}(\eta p \mathbf{k}; \mathbf{k}) = 0.$$
  
The first of these integrals is now multiplied

scalarly with  $i^{-1}{}_{0}\mathbf{U}^{*}(\eta_{1}, p_{1}, \mathbf{k}_{1}; \mathbf{x}, t)$  the second with  $i_{0}\mathbf{F}^{*}(\eta_{1}, p_{1}, \mathbf{k}_{1}; \mathbf{x}, t)$ . Then the two are added and integrated over the **x** space. Applying to the term containing  $\dot{A}$  the orthogonality conditions after the manner of Section 2(c) one finds

$$i\hbar A(\eta_1 p_1 \mathbf{k}_1; t) = \sum_{\eta, p} \int d\mathbf{k} A(\eta p \mathbf{k}; t)$$
$$\times (\eta_1 p_1 \mathbf{k}_1 | V| \eta p \mathbf{k}) e^{ic(\eta_1 \epsilon_1 - \eta \epsilon) t}. \quad (28)$$

The matrix element of the perturbation potential V can easily be brought into connection with the usual matrix element of the Schrödinger equa-

$$(\eta_1 p_1 \mathbf{k}_1 \mid V \mid \eta p \mathbf{k}) = (\eta_1 p_1 \mathbf{k}_1 \mid \bar{\rho} \mid \eta p \mathbf{k}) \cdot (\mathbf{k}_1 \mid V \mid \mathbf{k}), \quad (29)$$

where

$$(\mathbf{k}_{1} \mid V \mid \mathbf{k}) = (2\pi)^{-3} \int d\mathbf{x} V(\mathbf{x}) \\ \times \exp\left[i(\mathbf{k} - \mathbf{k}_{1} \cdot \mathbf{x})\right]. \quad (30)$$

Before integrating (28) in the usual fashion, a definite initial condition has to be agreed upon. We shall assume that at t=0

- (1) only particles of momentum  $\hbar \mathbf{k}_0$ , and
- (2) only particles of positive energy, i.e., with η<sub>0</sub>=+1 are present.

(3) For 
$$p = +I$$
 the amplitude is to be  $A_{+I}^{(0)}$ ,

for  $p = 0 A_0^{(0)}$ , and for  $p = -I A_{-I}^{(0)}$ .

Thus the wave vectors of the incident particles are:

$$\mathbf{U}_{\text{prim}} = \sum_{p} A_{p}^{(0)} {}_{0}\mathbf{U}(1, p, \mathbf{k}_{0}; \mathbf{x}, t),$$
  
$$\mathbf{F}_{\text{prim}} = \sum_{p} A_{p}^{(0)} {}_{0}\mathbf{F}(1, p, \mathbf{k}_{0}; \mathbf{x}, t).$$
 (31)

For t=0 the value of A is therefore

$$A(\eta, p, \mathbf{k}; 0) = \delta_{\eta, 1} \delta(\mathbf{k} - \mathbf{k}_0) A_p^{(0)}.$$
 (32)

To obtain the first approximation from (28) this condition is introduced into it on the right-hand term and the time integration performed. The result is:

$$\hbar c A^{(1)}(\eta, p, \mathbf{k}; t) = - \eta \frac{e^{i c (\eta \epsilon_k - \epsilon_0) t} - 1}{\eta \epsilon_k - \epsilon_0} (\mathbf{k} | V | \mathbf{k}_0)$$

$$\cdot \sum_{p_0} A_{p_0}^{(0)}(\eta, p, \mathbf{k} | \bar{\rho} | 1, p_0, \mathbf{k}_0)$$
(33)

in which  $\epsilon_0$  is written instead of  $\epsilon(\mathbf{k}_0)$ . In the same standard manner the second approximation is obtained: The above first approximation is substituted into the right-hand side of (28). Integration gives:

$$(\hbar c)^{2} A^{(2)}(\eta, p, \mathbf{k}; t) = \eta \sum_{\eta', p'} \eta' \int d\mathbf{k}' \\ \times (\mathbf{k} \mid V \mid \mathbf{k}') (\mathbf{k}' \mid V \mid \mathbf{k}_{0}) \\ \cdot \sum_{p_{0}} A_{p_{0}^{(0)}}(\eta, p, \mathbf{k} \mid \bar{\rho} \mid \eta', p', \mathbf{k}') \\ \times (\eta', p', \mathbf{k}' \mid \bar{\rho} \mid 1, p_{0}, \mathbf{k}_{0}) \cdot \Phi$$

$$(34_{1})$$

with

$$\Phi(\eta, \eta'; \epsilon, \epsilon', \epsilon_0; t) = \frac{e^{i\epsilon(\eta\epsilon-\epsilon_0)t}-1}{(\eta\epsilon-\epsilon_0)(\eta'\epsilon'-\epsilon_0)} - \frac{e^{i\epsilon(\eta\epsilon-\eta'\epsilon')t}-1}{(\eta\epsilon-\eta'\epsilon')(\eta'\epsilon'-\epsilon_0)}.$$
 (342)

The thus-obtained values for  $A(\eta, p, \mathbf{k}; t)$  should be put into (22) in order to furnish the eigenvectors of the perturbed problem in first or second approximation.

(b) It is, however, not necessary for our purpose to compute the eigenfunctions. All we are interested in is the number of particles  $N(\mathbf{k}, t)$  scattered into the solid angle element  $d\Omega$  between the time t=0 when the perturbation is switched on and the time t=t. This quantity is connected with the total number of scattered particles

$$\begin{split} \int d\Omega N(\mathbf{k}, t) &= \int d\mathbf{x} i^{-1} \{ (\mathbf{U}^* \cdot \mathbf{F}) - (\mathbf{F}^* \cdot \mathbf{U}) \} \\ &= \int d\mathbf{k} \sum_{\eta, p} \eta |A(\eta p \mathbf{k}, t)|^2 \end{split}$$

according to (23). Since

$$d\mathbf{k} = d\Omega k^2 dk$$

one can drop the  $d\Omega$  integration both sides. Finally the intensity measured at a distance R from the scatterer is given by:

$$J = \frac{1}{R^2} \frac{\partial N(\mathbf{k}, t)}{\partial t},$$

$$= \frac{1}{R^2} \frac{\partial}{\partial t} \int_0^\infty k^2 dk \sum_{\eta, p} \eta |A(\eta p \mathbf{k}, t)|^2.$$
(35)

Substituting for A the sum of the expressions given by (33) and (34) one obtains J up to the second approximation.

IV

(a) In this section J will be evaluated in first approximation. Introducing (33) into (35) we have:

$$J^{(1)} = \frac{1}{R^2} \frac{\partial}{\partial t} \int_0^\infty k^2 dk \sum_{\eta, p} \sigma(\eta p \mathbf{k}; \mathbf{k}_0) \cdot 4 \frac{\sin^2(c/2)(\eta \epsilon_k - \epsilon_0)t}{(\eta \epsilon_k - \epsilon_0)^2},$$

$$\sigma(\eta p \mathbf{k}; \mathbf{k}_0) = (\hbar c)^{-2} \eta | (\mathbf{k} | V | \mathbf{k}_0) |^2$$
$$\times | \sum_{p_0} A_{p_0}^{(0)}(\eta p \mathbf{k} | \bar{\rho} | 1 p_0 \mathbf{k}_0)$$

2

Only the term with  $\eta = +1$  gives a resonance point within the range of the *k* integration. Since  $\sigma(\dots)$  is a slowly varying function of *k* it can be taken outside of the integral with  $|\mathbf{k}_0|$ substituted for  $|\mathbf{k}|$ . The result is:

$$J^{(1)} = \frac{1}{R^2} \frac{2\pi}{\hbar^2 c^2} \epsilon_0 k_0 | (\mathbf{k} | V | \mathbf{k}_0) |^2 \sum_p | \sum_{p_0} \\ \times A_{p_0}^{(0)} (1 p \mathbf{k} | \bar{\rho} | 1 p_0 \mathbf{k}_0) |^2.$$
(36)

Although  $|\mathbf{k}|^2 = |\mathbf{k}_0|^2$ , their directions differ of course. The term  $|(\mathbf{k}|V|\mathbf{k}_0)|^2$  furnishes, for Coulomb interaction the well-known Rutherfordian angle function  $\csc^4(\vartheta/2)$ .

(b) The other factor which we will call P contains the relativistic and polarization effects due to the Dirac-Proca equations. Written in a more detailed fashion it is according to (18):

$$P = i^{-2} \sum_{p} \sum_{p_{0}} \sum_{p_{0}'} A_{p_{0}}^{(0)} * A_{p_{0}'}^{(0)}$$

$$\{ u_{\alpha} * (1p_{0}\mathbf{k}_{0}) f_{\alpha}(1p\mathbf{k}) - f_{\alpha} * (1p_{0}\mathbf{k}_{0}) u_{\alpha}(1p\mathbf{k}) \}$$

$$\cdot \{ u_{\beta} * (1p\mathbf{k}) f_{\beta}(1p_{0}'\mathbf{k}_{0}) - f_{\beta} * (1p\mathbf{k}) u_{\beta}(1p_{0}'\mathbf{k}_{0}) \}. \quad (37)$$

The summation over p is easy to perform by means of the destruction operators of Section 2(e). For instance, the product of the first term of the first parenthesis with the first of the second parenthesis contains

$$\sum_{p} f_{\alpha}(1p\mathbf{k}) u_{\beta}^{*}(1p\mathbf{k}) = \sum_{p} \sum_{\eta} \eta \frac{\eta+1}{2} f_{\alpha}(\eta p\mathbf{k}) u_{\beta}^{*}(\eta p\mathbf{k}).$$

Application of  $(26_1)$  from right to left and use of (27) make it possible to write this as:

$$\sum_{p,\eta} \eta \left[ Y_{\alpha\gamma}(\mathbf{k}) u_{\gamma}(\eta p \mathbf{k}) + \frac{1}{2} f_{\alpha}(\eta p \mathbf{k}) \right] u_{\beta}^{*}(\eta p \mathbf{k}) = \frac{1}{2} i \delta_{\alpha\beta}.$$

The latter is in accord with the orthogonality conditions (21). Similarly from the second term of the first parenthesis and from the first term of the second parenthesis of (37) there arises:

$$\sum_{p} u_{\alpha}(1p\mathbf{k})u_{\beta}^{*}(1p\mathbf{k}) = iZ_{\alpha\beta}(\mathbf{k}).$$

910

In this fashion (37) becomes:

$$P = \frac{1}{2} \sum_{p_0} |A_{p_0}^{(0)}|^2 + i^{-1} \sum_{p_0, p_0'} A_{p_0}^{(0)*} A_{p_0'}^{(0)} [\cdots], \quad (38)$$
$$[\cdots] = Y_{\alpha\beta}(\mathbf{k}) u_{\alpha}^* (1p_0 \mathbf{k}_0) u_{\beta} (1p_0' \mathbf{k}_0) - Z_{\alpha\beta}(\mathbf{k}) f_{\alpha}^* (1p_0 \mathbf{k}_0) f_{\beta} (1p_0' \mathbf{k}_0)$$

after account is taken of (18) and (20). Since the second term contains the arbitrary amplitude constants  $A_{p_0}^{(0)}$ , one has to spread out each term of the double sum into a sum of p and  $\eta$  in order to be able to apply the orthogonality conditions. Let us deal separately with terms where  $p_0 = p_0'$ and with those where  $p_0 = p_0'$ .

(a)  $p_0 = p_0'$ . The square bracket, the factor of  $|A_{p_0}^{(0)}|^2$  has now to be treated with the operators D of Section 2(e). According to (24) and (26) we have:

$$u_{\alpha}^{*}(1p_{0}\mathbf{k}_{0})u_{\beta}(1p_{0}\mathbf{k}_{0}) = D_{\beta\gamma}(p_{0}\mathbf{k}_{0})\sum_{p,\eta}\eta u_{\alpha}^{*}(\eta p \mathbf{k}_{0})$$
$$\times \{Z_{\gamma\delta}(\mathbf{k}_{0})f_{\delta}(\eta p \mathbf{k}_{0}) + \frac{1}{2}u_{\gamma}(\eta p \mathbf{k}_{0})\}.$$

Treating the second term of  $[\cdots]$  similarly, we finally obtain using (21):

$$[\cdots] = i D_{\alpha\beta}(p_0 \mathbf{k}_0) \{ Z_{\beta\gamma}(\mathbf{k}_0) Y_{\gamma\alpha}(\mathbf{k}) + Y_{\beta\gamma}(\mathbf{k}_0) Z_{\gamma\alpha}(\mathbf{k}) \}.$$
(39)

From (25) and (27) we get as contribution to P in the case

$$p_{0} = p_{0}' = 0:$$

$$|A_{0}^{(0)}|^{2} [1 + (1/4K^{2}\epsilon^{2})(k^{4} - (\mathbf{k} \cdot \mathbf{k}_{0})^{2})],$$

$$p_{0} = p_{0}' = +1.$$
(40)

$$|A_{\pm \mathbf{I}}^{(0)}|^{2} [1 + (1/8K^{2}\epsilon^{2})(k^{4} - (\mathbf{k} \cdot \mathbf{k}_{0})^{2})].$$

( $\beta$ )  $p_0 = I$ ,  $p_0' = -I$ . By use of (27), (15), (16) and (19) the square bracket of (38) becomes:

$$i(k^2/4K^2\epsilon^2)(\mathbf{k}\cdot\mathbf{e}(-\mathbf{I},\mathbf{k}_0))^2.$$

As this expression no longer possesses cylindrical symmetry, it becomes convenient to work with

the following rectangular components:

$$\mathbf{k}_{0} = (k, 0, 0),$$

$$\mathbf{e}(\mathbf{I}, \mathbf{k}_{0}) = (0, 2^{-\frac{1}{2}}, i2^{-\frac{1}{2}}),$$

$$\mathbf{e}(-\mathbf{I}, \mathbf{k}_{0}) = (0, 2^{-\frac{1}{2}}, -i2^{-\frac{1}{2}}),$$

$$\mathbf{k} = k(\cos \vartheta, \sin \vartheta \cos \varphi, \sin \vartheta \sin \varphi).$$

Then the contribution to P becomes:

$$A_{\rm I}{}^{(0)}*A_{-\rm I}{}^{(0)}\cdot\frac{k^4}{8K^2\epsilon^2}\sin^2\vartheta e^{-2i\varphi}.$$
 (41)

( $\gamma$ ) Because of the Hermitian character of each term of (38) the contribution of  $p_0 = -I$ ,  $p_0' = +I$  is

$$A_{-I}^{(0)} * A_{I}^{(0)} \cdot \frac{k^4}{8K^2\epsilon^2} \sin^2 \vartheta e^{+2i\varphi}.$$
(42)

( $\delta$ ) The contributions of terms one of whose  $p_0$  numbers is zero vanish. Collecting terms from (40) to (42) the following expression for P is obtained:

$$P = \sum_{p_0} |A_{p_0}^{(0)}|^2 + \frac{k^4}{4K^2\epsilon^2} \sin^2 \vartheta$$

$$\times \{|A_0^{(0)}|^2 + \frac{1}{2} |A_1^{(0)}e^{i\varphi} + A_{-1}^{(0)}e^{-i\varphi}|^2\}. \quad (43)$$

(c) From this formula it is seen, that as long as the incident waves are solely longitudinal, or if transverse, strictly right or left circular, the inclusion of relativistic and spin terms gives only a  $\vartheta$  effect which will be superimposed upon the Rutherfordian term  $|(\mathbf{k}|V|\mathbf{k}_0)|^2$ . But if both right and left circularly polarized waves are incident, i.e., if part of the incident wave is linearly polarized, an azimuth effect arises. This is seen when we put

$$A_{I} = A_{L}e^{-i\varphi_{0}}; \quad A_{-I} = A_{L}e^{i\varphi_{0}}.$$

Then P becomes, if we disregard longitudinal terms for the moment:

$$P = 2A_L^2 \{1 + (K^4/4K^2\epsilon^2) \sin^2\vartheta \cos^2(\varphi - \varphi_0)\}.$$

The appearance of a polarization even in the first approximation seems surprising in com-

parison with the electron for which such effects according to Mott and Sauter do not appear until the second Born approximation. However, a certain similarity which the Dirac-Proca equations bear to the Maxwell equations and the vectorial character of the wave functions (as compared to the spinor character for  $s=\frac{1}{2}$ ) may render our result more plausible.

## The effect is of the fourth order in v/c for

$$k^4/K^2\epsilon^2 = \beta^4/(1-\beta^2)$$

and is most pronounced at right angles to the direction of the incident particles.

The evaluation of the second approximation (34) and a closer discussion of the matrix element  $(\mathbf{k} | V | \mathbf{k}_0)$  will be given in a later paper.

#### PHYSICAL REVIEW

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# The Vapor Pressure of Isotopes

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It is shown here that if all degrees of freedom in a vapor are nearly classical and if there is no association, the vapor pressure of the lighter isotope is always higher. If only the external degrees of freedom are nearly classical and the internal ones are in the lowest state, the coupling of purely harmonic vibrations has either no influence or tends probably to a further increase of the excess vapor pressure of the light isotope. Anharmonicity and the change in the van der Waals forces probably account for those cases in which the heavier isotope has the higher vapor pressure.

# I. INTRODUCTION

THE difference in the physical and chemical equilibrium conditions for isotopes is a pure quantum phenomenon.<sup>1</sup> Classical theory would give no effects of the difference in mass. This latter only affects the statistical distribution of momenta.<sup>2</sup> In classical theory, however, the integration over the momenta is independent of the integration over the coordinates and influences the different states (e.g., vapor and liquid) in the same manner. Therefore it does not modify the equilibrium between two states. This suggests a systematic investigation of the isotope effect in different temperature regions.

### II. LOW TEMPERATURES

At low temperatures the differences are most conspicuous. The adequate method which is in

general use starts from the condition at absolute zero. The greatest effect in this region is due to the difference in zero point energy of the motion of the molecule as a whole in the field of its neighbors. Since the operator of the kinetic energy is positive definite the lowest quantum level of the lighter isotope will always be higher than that of the heavier isotope. As a consequence the lighter molecule will have a smaller heat of evaporation and therefore a greater vapor pressure.<sup>3</sup> The difference of entropy of translation and possibly rotation in the vapor state works in the opposite direction but has less importance in the low temperature region. All this has frequently been the subject of discussions.

## III. HIGH TEMPERATURES

As mentioned before, there are no differences between isotopes at temperatures so high that

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<sup>&</sup>lt;sup>1</sup> We exclude cases in which gravity or centrifugal forces are of importance.

<sup>&</sup>lt;sup>2</sup> The very small differences in potential energies for different isotopes shall be neglected except at the end of the paper.

<sup>&</sup>lt;sup>8</sup> A possible exception might occur if one has association in the vapor phase connected with a very high intermolecular vibrational frequency.