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ON THE RELATIVISTIC ATOMIC HAMILTONIAN

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Abstract. — The relativistic hamiltonian for a multi-electron atom is studied to make explicit the assumptions and approximations which enter into it. This will allow corrections to the approximations to be made as needed within the framework provided by the assumptions.

The basic assumptions are the validity of quantum electrodynamics and the accuracy of using effective potentials to reduce quantum electrodynamics to hamiltonian form. The ambiguities of the potential obtained for the first order interaction are discussed. Another source of difficulty is in the precise definition of the position variables to be used for internal interactions and for interactions with external fields.

Using these results a hamiltonian could be constructed for nuclei with a particular spin. Instead the nuclear motion is approximated non-relativistically allowing a phenomenological description of the nuclear moments which is valid for all spins.

1. Introduction. — Atomic physics is starting to use a more complete relativistic formulation in its calculations. When working with the non-relativistic hamiltonian, there were relativistic corrections to the theory. The present relativistic hamiltonian may also need corrections. In order to understand when corrections might be needed and what they would be, the hamiltonian must be obtained by successive approximations from a theory which is as well founded as possible.

In the following it is useful to distinguish between two types of approximations. The first type is mathematically based on the theory being used. In this case the error can be estimated and the approximation improved through knowledge of the mathematics of the approximation. A truncated series is an example of this type. The second type of approximation is based on physical intuition and is usually used when there are unsolved mathematical problems in the way of a rigorous development. In this case the error cannot be estimated directly and the approximation usually cannot be improved. Hopefully, further work will change any such approximation into one of the first type.

If the nucleus is treated as a simple particle, all interactions within the atom are electromagnetic [1]. This allows the theory of quantum electrodynamics to be the starting point of the description of an atom. Unfortunately even two-particle bound states are very difficult to handle in a rigorous fashion [2]. The assumption will be made that for the case of interest quantum electrodynamics can be translated into hamiltonian form through the use of effective potentials, where an effective potential is defined as that potential which when added to the free particle hamiltonian gives the same scattering results as quantum electrodynamics [3]. Scattering is used because of the intractability of the quantum electrodynamic bound state problem. This may be regarded as the second type of approximation mentioned above where the correction is assumed to be not just small but zero. The justification comes from the accuracy of the calculations for hydrogen done by Grotch and Yennie [3] and from a treatment of the Bethe-Salpeter equation by Gross [4].

The effective potential, like quantum electrodynamics scattering calculations, is a power series in the fine structure constant, \( \alpha \). This series can be appro-
The Effective Potential. — The effective potential of interest is that one which describes the electromagnetic interaction of two pure Fermi particles, which have neither extent nor Pauli moments. This description is correct only for leptons, whose anomalous magnetic moments are ascribable to higher order quantum electromagnetic effects. The modifications for the interactions of the nucleus will be described in Section 4.

The interaction is to be approximated by the lowest order in $\alpha$ which is given by the exchange of a single virtual photon. Because of the close relation between the quantized and non-quantized forms of field theory, the effective potential can be directly written in momentum space [6]:

$$V_M(\epsilon, k) = 4 \pi q_1 q_2 \left( \frac{1}{k^2 - \epsilon^2} (1 - \alpha_1 \cdot \alpha_2) \right),$$

where $(\epsilon, k)$ is the four-momentum of the exchanged photon and $q_i$ and $\alpha_i$ are the charge and Dirac matrices for the $i$th lepton.

The requirement of gauge invariance or charge conservation imposes the condition

$$\epsilon - k \cdot \alpha = 0$$

on all physical states [6]. This allows the potential to be rewritten in the form

$$V_B(\epsilon, k) =$$

$$= 4 \pi q_1 q_2 \left( \frac{1}{k^2 - \epsilon^2} (\alpha_1 \cdot \alpha_2 - (\alpha_1 \cdot \hat{k})(\alpha_2 \cdot \hat{k})) \right).$$

(2.1)

(2.3)

This form can be derived directly by quantizing only the transverse electromagnetic field and then writing the interaction as the sum of the Coulomb interaction (first term) plus the exchange of one transverse photon (second term).

The usual three dimensional Fourier transforms are used to obtain the expressions for the interactions in configuration space. The significance of the variables will be discussed in the next section. The results are

$$V_M(\epsilon, R) = \frac{q_1 q_2}{R} (1 - \alpha_1 \cdot \alpha_2) \theta(\epsilon R)$$

(2.4)

and

$$V_B(\epsilon, R) = \frac{q_1 q_2}{R} \left( 1 - [\alpha_1 \cdot \alpha_2 - (\alpha_1 \cdot \hat{R})(\alpha_2 \cdot \hat{R})] \theta(\epsilon R) + [\alpha_1 \cdot \alpha_2 - 3(\alpha_1 \cdot \hat{R})(\alpha_2 \cdot \hat{R})] \tau(\epsilon R) \right),$$

(2.5)

where

$$\tau(\epsilon R) = \frac{1}{\epsilon^2 R^2} \left( \theta(\epsilon R) - 1 - \epsilon R \theta(\epsilon R) \right).$$

(2.6)

$\theta(\epsilon R)$ is determined by the contour in the complex $k$-plane with respect to the poles at $\pm \epsilon$. For scattering problems [7] a contour above (below) $-\epsilon$ and below (above) $+\epsilon$ transforms incoming (outgoing) plane waves into outgoing (incoming) plane-plus-spherical waves and

$$\theta(\epsilon R) = e^{-\epsilon R \theta(\epsilon R)}.$$
In the case where the states of the system are products of independent one particle states, possibly defined in the presence of an external field, and the entire interaction between the particles is treated as a perturbation, the matrix elements of \( \Delta V \) are identically zero. If part of the interparticle interaction is used to define the initial states and the rest of the potential is treated by a perturbation expansion, then the choice of the form of the potential may give quite different results to any given order of the expansion; although the limits are presumably the same. This latter situation is the case in hydrogen where the Coulomb interaction defines the bound states and in Hartree-Fock states. If the hyperfine structure of hydrogen is calculated to first order in perturbation theory using states defined by the Coulomb interaction, the result is of the correct order of magnitude when \( V_B \) is used but is \( \pi^2 \) times too large using \( V_M \). Also calculations of energy levels and quantum electrodynamic effects give good results using \( V_B \) [11].

The general effective interaction to first order in \( \pi \) may be written
\[
V = V_B + \pi \Delta V ,
\]
where \( \pi \) is an arbitrary number which may be chosen to give rapid convergence to a perturbation series. The calculations mentioned above [3] [11] imply that the optimum value is zero but these have been carried out for small \( \pi \) only. Whether a value of \( \pi \) near zero is optimum for large \( \pi \) as well remains to be answered.

### 3. The Definition of Variables

The relations between momentum and position in non-relativistic quantum mechanics are well understood. A given set of position and momentum variables are canonical if they obey the standard commutation relations:
\[
\left[ p_{ij}, x_{jk} \right] = -i \delta_{ij} \delta_{ab} .
\]

For a system of \( n + 1 \) particles one possible set of canonical variables is the positions and momenta of the separate particles. A more useful set consists of the position and momentum of the center-of-mass plus a set of internal positions and momenta. The internal variables are difficult to define independently if all particles are to be treated identically [12]; but in the case where one particle, e.g. the nucleus, is different from the others, a set of independent non-relativistic center-of-mass canonical variables can be defined:
\[
p = p_N + \sum p_j ,
\]
\[
x = \frac{1}{M} \left( m_N x_N + \sum m_j x_j \right) ,
\]
\[
p_i = p_i - \frac{m_i}{M} \left( p_N + \sum p_j \right) ,
\]
\[
r_i = x_i - x_N .
\]
where
\[
M = m_N + \sum m_j .
\]

Potentials which were derived in momentum space as functions of the momentum transfer have a configuration space dependence on \( r_i \) for the interaction between \( N \) and the \( i \)th particle and on \( r_i - r_j \) for the interaction between the \( i \)th and \( j \)th particles.

In the relativistic case many problems arise. The position canonical to the three-momentum of a particle [13] is not part of a four-vector, depends on the frame of reference, and depends on the spin of the particle in the frame of reference. For a multiparticle system it is again advantageous to use the center-of-mass plus internal variables but this problem has not been completely solved [14] [15]. To avoid this difficulty as long as possible we will define \( R \) and \( R_{ij} \) to be the position variables appearing in the interactions between the nucleus and the \( i \)th electron and between the \( i \)th and \( j \)th electron respectively. The exact forms of \( R \) and \( R_{ij} \) will depend on the form of the relativistic internal variables.

The exact solution of the two-particle case [14] produces quite complex general expressions for the center-of-mass and internal variables, which become the non-relativistic expressions in the center-of-mass rest frame. This suggests that the approximation of the relativistic internal variables by the non-relativistic ones is made better by restricting the system to the center-of-mass rest frame. With this restriction,
\[
p_N = - \sum p_i ,
\]
and assuming the validity of the approximation, we set
\[
R_i = r_i ,
\]
\[
R_{ij} = r_i - r_j ,
\]
and
\[
p_i = -i \frac{\partial}{\partial r_i} .
\]

The method used to derive the potential has resulted in a dependence on \( \varepsilon \), the energy transfer, which can be taken to be the change in energy of either interacting particle [8]. This implies that enough of the problem has been solved to assign a value to \( \varepsilon \). This definition can be applied only to states where the particles are independent enough to be assigned separate energies.

Estimating \( \varepsilon \) to have a maximum near the hydrogenic ionization energy and \( R \) to be of the order of magnitude of the Bohr radius, \( \varepsilon R \) is of the order of \( \varepsilon z \). This suggests that a good first approximation, particularly for \( z \) small, is to set \( \varepsilon \) to zero. The difference between this hamiltonian and the hamiltonian dependent on \( \varepsilon \) could be treated as a perturbation whose dependence on \( \varepsilon \) could be approximated. In turn, by values obtained from the approximate hamiltonian.
Difficulties arise on evaluating non-diagonal matrix elements of the perturbation which do not conserve the total energy as defined by the approximate hamiltonian. In such cases there are two possible values of $e$ depending on which of the two particles is used to define the «energy transfer». Fortunately this ambiguity does not cause difficulty until the second order of perturbation theory.

4. The Nucleus. — The interactions discussed in section 2 were for pure Fermi particles which do not have either anomalous magnetic moments or extent. To describe a spin-$\frac{1}{2}$ nucleon interacting with the electromagnetic field a Pauli term and form factors must be used [16]. In this fashion the interactions can easily be generalized to include a spin $-\frac{1}{2}$ nucleus.

The nuclear motion can be approximated non-relativistically. The order of magnitude of a relativistic correction to this approximation may be estimated by writing:

$$p_i^2 = \frac{1}{8m_i^3} \left( \frac{p_i^2}{2m_i} \right)^2.$$  

The quantity in parenthesis is roughly the total nuclear mass effect. A generous estimate of which is 1 000 mk [17], giving $10^{-11}$ mk for an estimate of this correction. Since other correction terms and better evaluations of this term seem unlikely to change the estimate of the total correction by more than a few orders of magnitude, the corrections to the non-relativistic approximation of nuclear motion can be ignored in all cases as well below present experimental accuracy.

The treatment of the nucleus may now be generalized by replacing the charge and magnetic moment of the nucleus by distributions centered on the position of the nucleus. These distributions may be defined empirically:

$$q(p) = <\psi_n | q(p) | \psi_n >,$$ 

$$\mu(p) = <\psi_n | \mu(p) | \psi_n >,$$

where the barred quantities are operators acting on internal states of the nucleus. Although not indicated explicitly, the distributions depend on the initial and final nuclear states. These general distributions are included in the non-relativistic nuclear hamiltonian by the replacements:

$$q \varphi(R) \rightarrow \int q(p) \varphi(R-p) d^3p,$$ 

$$\mu \cdot B(R) \rightarrow \int \mu(p) \cdot B(R-p) d^3p.$$ 

For a spin-$\frac{1}{2}$ nucleus the distributions can be obtained by starting in momentum space with the complete interaction including the relativistic form factors, going to configuration space by a Fourier transform, and reducing the nuclear part of the hamiltonian to the non-relativistic limit. The charge and magnetic moment distributions are shown in this way to be the Fourier transforms of the relativistic form factors $F_i$ and $G_M$ [17]. This derivation also shows that the distributions can be expected to depend on the energy transfer. For the proton the relative error of ignoring this dependence is about $(e/750$ MeV)$^2$ [18].

This development may be carried further using spherical harmonics and radial functions. Some aspects of a purely empirical approach are discussed in Appendix 2.

5. Conclusion. — The starting point for the relativistic atomic hamiltonian was the hypothesis that the interactions are described by quantum electrodynamics. It was further assumed that in the atomic case quantum electrodynamics can be put in the form of a relativistic hamiltonian with an effective potential. The effective potential was evaluated only to the first order in the fine structure constant. The nuclear motion was found to be very well approximated by the non-relativistic limit. With these conditions, in the center-of-mass rest frame, and in the absence of external fields the relativistic hamiltonian for an atom is

$$H = H_N + H_e.$$  

The nuclear part which includes all interactions between the nucleus and the electrons is

$$H_N = m_N + \frac{1}{2m_N}(p^2_N - \sum qA_i)^2 + \sum (q\varphi) +$$ 

$$+ \sum (m \cdot V_i A_i).$$

where $q$ and $m$ are the nuclear charge and magnetic moment distributions which are to be used as in (4.3). The use of the center-of-mass rest frame requires (3.7) and the other quantities are

$$\phi_i = - \frac{e}{R_i},$$

$$A_i = - \frac{e}{R_i} \left\{ [\alpha_i - (\alpha_i \cdot \hat{R}_i) \hat{R}_i] 0(e_i R_i) -$$ 

$$- [\alpha_i - 3(\alpha_i \cdot \hat{R}_i) \hat{R}_i] \tau(e_i R_i) \right\},$$

and

$$V_i = \frac{\partial}{\partial \hat{R}_i}.$$ 

The electron part of the hamiltonian,

$$H_e = \sum H_i + \sum V \psi_j,$$

consists of the relativistic free electron hamiltonians:

$$H_i = z_i \cdot p_i + \beta_i m.$$
and the inter-electron interaction,

\[ V_{ij} = \frac{e^2}{R_{ij}} \left\{ \left[ 1 - \{ \alpha_i, \alpha_j - (\alpha_i, \tilde{R}_{ij}) (\alpha_j, \tilde{R}_{ij}) \} \right] \theta(\epsilon_{ij}, R_{ij}) + \right. \]

\[ + \left. \left[ (\alpha_i, \alpha_j - 3(\alpha_i, \tilde{R}_{ij}) (\alpha_j, \tilde{R}_{ij}) \right] \tau(\epsilon_{ij}, R_{ij}) \right\} \] \quad (5.8)

In these expressions the indices refer to the electrons and \( \theta \) and \( \tau \) are as defined in (2.8) and (2.9).

In order to use the hamiltonian the relationships among the variables must be known. Approximating these non-relativistically leads to (3.8), (3.9), and (3.10). The dependence on the energy transfer, \( \epsilon \), may be approximated using first order perturbation theory based on states which are solutions of the hamiltonian obtained by setting the energy transfer to zero. The nuclear distributions are approximated by a point nucleus.

Through the formalism of quantum electrodynamics, charge conservation introduces a degree of freedom into the effective potential. The forms used above work well for calculations using perturbation theory when \( \epsilon \) is small. It is possible that for the more relativistic electrons of the heavier atoms a perturbation expansion will converge faster if the interactions are modified by the addition of

\[ a_i \Delta \varphi_i = a_i \left\{ \left( -\frac{e}{R_i} \right) \left[ \theta(\epsilon_i, R_i) - 1 \right] \right\} \] \quad (5.9)

\[ a_i \Delta A_i = a_i \left\{ \left( -\frac{e}{R_i} \right) (\alpha_i, \tilde{R}_i) \tilde{R}_i \theta(\epsilon_i, R_i) + \right. \]

\[ + \left. \left[ \alpha_i - 3(\alpha_i, \tilde{R}_i) \tilde{R}_i \right] \tau(\epsilon_i, R_i) \right\} \] and

\[ a_{ij} \Delta V_{ij} = a_{ij} \frac{e^2}{R_{ij}} \left\{ \left[ \theta(\epsilon_i, R_i) - 1 \right] - \right. \]

\[ - (\alpha_i, \tilde{R}_{ij}) (\alpha_j, \tilde{R}_{ij}) \theta(\epsilon_{ij}, R_{ij}) \]

\[ - \left. \left[ (\alpha_i, \alpha_j - 3(\alpha_i, \tilde{R}_{ij}) (\alpha_j, \tilde{R}_{ij}) \right] \tau(\epsilon_{ij}, R_{ij}) \right\} \] \quad (5.8)

where \( a_i \) and \( a_{ij} \) are numerical constants whose values are chosen to enhance the rate of convergence of the perturbation series.

Appendix 1. External Interactions. — In quantum electrodynamics external fields are treated as algebraic fields, not operators, which are included in the description of «free» particles, i.e. those which do not interact with each other [6]. This results in modifying the hamiltonian in the usual way:

\[ H' = H'_N + H'_e. \] \quad (A1.1)

\[ H'_N = m_N + \frac{1}{2m_N} \left\{ p_N - \sum (qA_i) + (q \Lambda^{ext}(\chi_N)) \right\}^2 + \]

\[ + \sum (q\varphi) + (q \varphi^{ext}(\chi_N)) + \sum (m \cdot V_i \times A_i) - \]

\[ - (m \cdot V_N \times \Lambda^{ext}(\chi_N)) \], \quad (A1.2)

\[ H'_e = \sum H_i + \sum_{i<j} V_{ij} + \sum V_{ext}^{i}, \] \quad (A1.3)

\[ V_{ext}^{i} = - e \left\{ \varphi^{ext}(\chi_i) - \alpha_i \cdot \Lambda^{ext}(\chi_i) \right\}. \] \quad (A1.4)

The principal difficulty with the external interactions is with the external variables used while the rest of the hamiltonian uses internal variables. Even in the center-of-mass rest frame of a two particle system, the relativistic expression for the position of each particle does not reduce to a simple form but includes spin dependent terms [14].

The lowest order approximation would be to use the non-relativistic formulae for the absolute positions:

\[ \chi_N = X - \frac{1}{M} \sum m_j F_j, \] \quad (A1.5)

\[ \chi_i = X + r_i - \frac{1}{M} \sum m_j r_j. \] \quad (A1.6)

Since the eigenfunctions of the relativistic position operators have a spread of about one Compton wavelength, this approximation should be fairly accurate if the external fields vary little within one electron Compton wavelength.

An approximate relativistic correction to an electron position can be obtained by using the two-particle form of Osborn [14] and treating the rest of the atom as the second particle:

\[ - \frac{1}{4m_i M} (p_i r_i + r_i p_i^2 + 2 p_i \times s_i). \] \quad (A1.7)

With this correction the external field is restricted to vary little within the order of a nuclear Compton wavelength. However since our internal variables are imperfectly defined, they may introduce a much larger error.

Appendix 2. Some Empirical Aspects of Charge and Magnetic Moment Distributions. — The charge and magnetic moment distributions to be discussed will be the general ones defined by (4.2) which interact with an electromagnetic field as in (4.3). Treating the distributions as functions of position and using the modified spherical harmonics defined by Racali [19], they can be written:

\[ q(p) = \sum (2k + 1) C^k(p) q^k(p), \] \quad (A2.1)

\[ m^r_s(p) = \sum (-1)^{k-s}(2k + 1) \left( \frac{2k + 1}{3} \right)^{\frac{k}{2}} \times \]

\[ \times \left\{ C^\dagger(p) m^{k+1rk} \right\}^1_s, \] \quad (A2.2)
where
\[ q^k_q(p) = \frac{1}{4\pi} \int C_q^k q(p) \, d\Omega_p, \quad (A2.3) \]
\[ m'^{k+1}_q(p) = \frac{1}{4\pi} \int \{ C^k m'^{k+1}_q \} \, d\Omega_p. \]

(A2.4)

The general form of the magnetic moment distribution can be somewhat simplified since the field it interacts with in (4.3) is the curl of another field [20]. In terms of this interaction two dipole distributions, \( m \) and \( m' \), cannot be distinguished if
\[ \mathbf{V} \times (\mathbf{m} - \mathbf{m'}) = 0. \]

These two distributions also produce the same magnetic field.

Let
\[ m^{(k+1)k}_q = 0 \]
\[ m^{(k+1)k}_q = m^{(k+1)k}_q \]
\[ m'^{k+1}_q = m'^{k+1}_q + \left( \frac{k(2k-1)}{(k+1)(2k+3)} \right)^{1/2} \times \]
\[ \times \left\{ m'^{k+1}_q - (2k+1) \rho^{k-1} \int_0^\infty \frac{1}{\rho^{2k}} m'^{k+1}_q \, d\rho \right\}. \]

This allows the sum of \( \kappa \) in (A2.2) to include only \( k \) and \( k - 1 \).

The radial functions can be further reduced by going to the operator form. The use of reduced matrix elements eliminates the dependence of the radial functions on the magnetic quantum number, \( q \), and also imposes limits on the rank, \( k \), which depend on the initial and final nuclear states in (4.2). The use of parity further reduces the number of radial functions. For example, if the initial and final nuclear states are identical and \( S \) and parity is used then to be non-zero \( q^k \) and \( m^{(k+1)k} \) must have \( k \leq 2 S \) and \( k \) even and \( m'^{k+1} \) must have \( k \leq 2 S \) and \( k \) odd.

In using these expansions the interactions (4.3) become integrals over \( \rho \). Since the nuclear size is much smaller than the size of the atom, a Taylor expansion in \( \rho \) of the field may be expected to yield a series of rapidly diminishing terms. This Taylor expansion may be included symbolically in the distribution through the use of \( \delta \)-functions:

\[ \delta^{(k+n)}(\rho) = \left( \frac{d}{d\rho} \right)^{k+n} \delta(\rho) \quad (A2.7) \]
and
\[ Q^{(n)} = 4\pi \int \rho^{k+n} q^k(p) \rho^2 \, d\rho, \quad (A2.8) \]
\[ M^{(k+1)k}_n = 4\pi \int \rho^{k+1} m^{(k+1)k}_n \rho^2 \, d\rho. \]

This allows the nucleus to be treated as a point.

If we can make the apparently reasonable physical assumption that at least one of the two charge or dipole distributions, nuclear or electron, does not have any singularities in any of its derivatives at \( \rho = 0 \), then these formulae can be further simplified by using a three dimensional Taylor expansion instead of a radial one for \( \rho > 0 \). This corresponds to the standard multipole expansion [21]. In this case all terms of odd \( n \) are zero. However, many of the functions used to describe the distribution have singularities at the origin. For example any function containing an exponential depending on an odd power of \( \rho \) has a singularity such as the relativistic and non-relativistic [7] hydrogenic solutions, the Fourier transform of the relativistic proton form factors [18], and the standard Fermi nuclear charge distribution [22]. In any case the terms with \( n = 0 \) can be expected to be larger than the others. For these terms, the standard moments can be defined [21]:

\[ Q^k = \int \rho^k C^k_q q(p) \, d^3\rho = Q^k_{q(\rho = 0)}, \quad (A2.8) \]
\[ M^{(k+1)k}_n = \left( \frac{1}{k(2k-1)} \right)^{1/2} \int \rho^{k-1} \left\{ C^{(k+1)k}_m(p) \right\}^k \, d^3\rho \]

References

[1] This implies that the weak interactions may be ignored. Any effects of interest caused by the weak interactions could presumably be handled by a perturbation series.
GELL-MANN (M.) and LOW (F.), Phys. Rev., 1951, 84, 350.
[4] GROSS (F.), reported by Grotch and Yennie [3].
[5] This is the manner in which Grotch and Yennie [3] performed their calculations.
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A few other references may be found in Rose (M. E.), *Relativistic Electron Theory*, 1961, Wiley, New York.


[11] Some calculations may be found in [2] and [3].

A few others are:

Fulton (T.) and Karplus (R.), *Phys. Rev.*, 1954, 93, 1109.


[18] This calculation is based on the « dipole fit » to the nucleon form factors given by Gasiorowicz [16].


The notation for the coupling of tensor operators is that of Innes (F. R.), *Phys. Rev.*, 1953, 91, 31.

[20] This need not be assumed. The vigorous derivation of the spin+ case gives this form for the non-relativistic magnetic interaction of the nucleus.

