

The Dirac Equation in Electronic Structure Theory

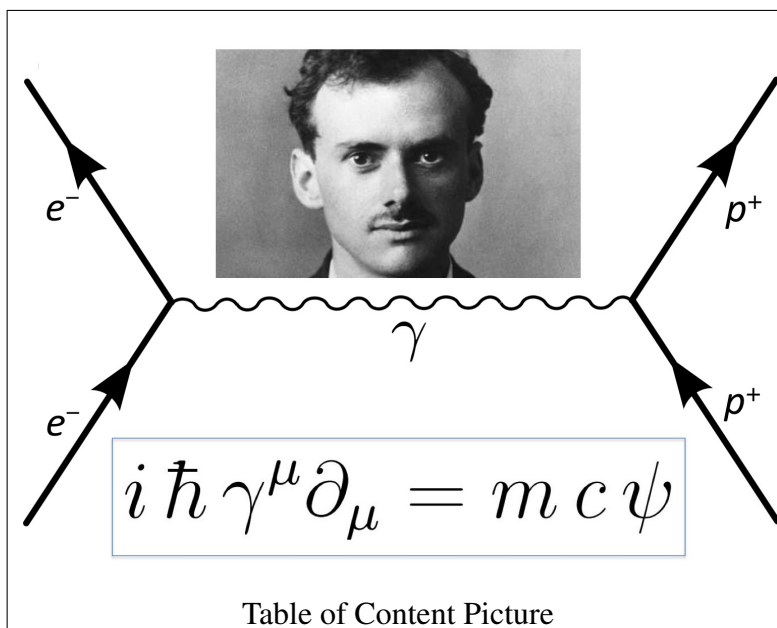
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Abstract

The Dirac equation applied in relativistic electronic structure theory is introduced and discussed in detail including transformations to two-component forms and corresponding picture change effects for properties. Recent progress in bound-state quantum electrodynamics and its importance to accurate electronic structure theory is analyzed. A short discussion on the current standard model and the search for new physics is also presented. Few applications are presented using prime examples for the importance of relativistic and quantum electrodynamic effects in heavy element chemistry. Special emphasis is put on the many theoretical and technical difficulties in treating the Dirac equation and quantum electrodynamic effects.

Keywords: Dirac equation; two-component forms; bound state quantum electrodynamics; standard model and beyond; relativistic and QED effects. ■



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1 INTRODUCTION

In 1928, P. A. M. Dirac proposed a relativistic formulation for the quantum theory of a spin 1/2 particle with rest mass m in external scalar $V(\vec{r}, t)$ and vector $\vec{A}(\vec{r}, t)$ fields,¹⁻³

$$i\hbar \frac{\partial}{\partial t} \Psi(\vec{r}, t) = \left\{ c\vec{\alpha} \left(\vec{p} + \frac{e}{c} \vec{A}(\vec{r}, t) \right) + \beta mc^2 + V(\vec{r}, t) \right\} \Psi(\vec{r}, t) \quad (1)$$

with the velocity of light set at $c = 299792458 \text{ ms}^{-1}$. In Dirac's formulation fermionic spin emerges naturally as a consequence of a relativistic treatment by enforcing Lorentz invariance, and in a complete covariant formulation it reads,^{*,†}

$$\{mc + \hbar\gamma_\mu \partial_\mu\} = 0 \quad (2)$$

with the external potential being introduced by minimal coupling (also called minimal substitution),

$$p_\mu \mapsto p_\mu - \frac{q}{c} A_\mu \quad (3)$$

with the charge $q = -e$ for an electron (e is the elementary charge), and $\partial_\mu = \frac{\partial}{\partial x_\mu}$. Here $\vec{\alpha}$, β and $\gamma_\mu = (\beta, -i\beta\gamma_1, -i\beta\gamma_2, -i\beta\gamma_3)$ are the Dirac (4×4) matrices in standard representation,

$$\vec{\alpha} = \begin{pmatrix} 0 & \vec{\sigma} \\ \vec{\sigma} & 0 \end{pmatrix}, \quad \beta = \gamma_0 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad \vec{\gamma} = -i\beta\vec{\alpha} = i \begin{pmatrix} 0 & -\vec{\sigma} \\ \vec{\sigma} & 0 \end{pmatrix} \quad (4)$$

and $\vec{\sigma}$ are the (2×2) Pauli spin matrices,

$$\sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_2 = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \quad (5)$$

The question naturally arises of why we should use the Dirac equation in electronic structure theory instead of the Schrödinger equation, as the latter has served the quantum chemical community extremely well for almost a century. We may rightly ask *what is wrong with the Schrödinger*

^{*}We keep this review short and without any proofs to not distract from the main important facts, and more complicated mathematical issues are moved into the footnotes. When appropriate, references are given for a more detailed treatment.

[†]Common (3-)vectors $\vec{v} \in V(\mathbb{R}^3)$ are used here with a vector sign, and 4-vectors (Lorentz vectors) $x_\mu = (ict, x_1, x_2, x_3) \in \mathcal{M}$ ($\mu = 0, 1, 2, 3$) in Minkowski space \mathcal{M} are used with Greek subscript symbols. The Einstein sum convention for repeated indices in products of 4-vectors/tensors are used. We note that in special relativity we do not need the more commonly used formulation of co- and contra-variant vectors in physics (which requires the introduction of a dual space), which is more important in the theory of general relativity.

equation, or, why do we have to turn to the more complicated Dirac equation known to be not without difficulties?[‡] From a purely theoretical view point the answer is simple: In the Schrödinger picture the time-evolution is given by the unitary transformation

$$\Psi(t) = U(t - t_0) \Psi(t_0) = e^{-i\hat{H}(t-t_0)/\hbar} \Psi(t_0) \quad (6)$$

(related to Stone's theorem[§]) leading to the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \Psi(t) = \hat{H} \Psi(t) \quad (7)$$

If we accept the first derivative in time as being fundamental as eq.(7) describes the time evolution of a quantum system with H being the Hamiltonian, we strictly require first derivatives in space in \hat{H} to enforce (relativistic) Lorentz invariance. The Schrödinger equation is, however, *not* Lorentz invariant as space derivatives appear in second instead of first order,

$$i\hbar \frac{\partial}{\partial t} \Psi(\vec{r}, t) = \left[-\frac{\hbar^2}{2\mu} \Delta + V(\vec{r}, t) \right] \Psi(\vec{r}, t) \quad (8)$$

This may not be seen as an important drawback if nonrelativistic particles are involved,[¶] but the Schrödinger equation is also “spin-free”, e.g. the nonrelativistic Hamiltonian \hat{H} does not contain terms accounting for fine-structure effects for a spin 1/2 particle which becomes large in heavy elements, and experimentally observed fine (and hyperfine) splittings in atoms and molecules cannot be reproduced. If the spin of a particle is introduced into a (nonrelativistic) Galilei-invariant formalism, the resulting Lévy-Leblond equation leads to an incorrect spin-orbit coupling term.⁴ If spin interaction terms like the spin-orbit operator are introduced ad-hoc into the Schrödinger equation, the specific form of this operator still requires theoretical justification, and this can only be done through the Dirac equation (see section 3 on two-component forms). We also know that for heavy elements the Schrödinger equation yields results for inner shell ionization potentials that

[‡]Unlike the semi-bound nonrelativistic Schrödinger operator \hat{H} , the relativistic Dirac operator \hat{D} is unbound requiring extra boundary conditions in a variational treatment. Moreover, it requires the introduction of a finite nuclear charge to keep \hat{D} self-adjoint beyond nuclear charge 137.

[§]Stone's theorem is a basic theorem of functional analysis that establishes a one-to-one correspondence between self-adjoint operators on a Hilbert space \mathcal{H} to one-parameter families of unitary operators that are strongly continuous. This defines the derivative of the mapping $t \mapsto Ut$, which is only supposed to be continuous.

[¶]We define a particle to behave nonrelativistically if its interaction energy E_{int} with an external field (such as the Coulomb field of a nucleus) is much less than the particle's rest mass energy, i.e. $E_{int} \ll mc^2$. For an electron the rest mass energy is $m_e c^2 = 510.998910(13)$ keV which can be compared for example to the K-shell ionization potential of Pb with 88 keV.

Z	Element	nl	E_b [eV]	$\beta = v/c$	nl	E_b [eV]	$\beta = v/c$
3	Li	1s			2s		
20	Ca	1s			4s		
54	Xe	1s			5p		
80	Hg	1s			6s		
92	U	1s			5f		
112	Cp	1s			7s		
118	UUo	1s			7p _{3/2}		

Table 1: K- and valence-shell binding energies and corresponding velocities β (in units of c) for some selected elements.

seriously deviate from experimental values. This may, however, not impress chemists and we may still ask why relativistic effects should be of any importance to valence electrons, as the interaction energy of an electron with a positively charged (but screened) nucleus is only in the eV range and much less compared to the electron's rest mass energy.

Consider the relativistic energy-momentum relation for a freely moving electron with rest mass m_e ,

$$E^2 = c^2 p^2 + m_e^2 c^4 \quad (9)$$

Introducing the relativistic momentum $\vec{p} = \gamma m \vec{v}$ we get

$$E^2 = c^2 p^2 + m_e^2 c^4 = c^2 (\gamma^2 m_e^2 v^2) + m_e^2 c^4 = m_e^2 c^4 \left(\frac{1}{1 - \beta^2} \right) \quad (10)$$

with the Lorentz factor defined as

$$\gamma = \frac{1}{\sqrt{1 - \beta^2}} = \frac{1}{\sqrt{1 - v^2/c^2}} \quad (11)$$

The Taylor expansion of eq.(10) leads to the well known classical kinetic energy term (after subtracting the rest mass energy). Eq.(10) allows us to convert typical shell binding energies E_{nl} for electrons into velocities a free electron would have with the same energy. The results for some few selected elements and energy levels are shown in Table 1.

Dirac stated in 1929² that relativistic effects are not important to chemistry as valence electrons move rather slowly compared to the velocity of light, and similar statements were made not too long ago.^{||} It came therefore as a surprise when relativistic effects for valence electrons were announced to be important in heavy elements, to such an extend that chemical properties

^{||}Dirac's statement in 1929 reads² *The general theory of quantum mechanics is now almost complete, the imper-*

can change significantly.⁷⁻¹⁴ In fact, strong relativistic effects in the valence shell are more common than were thought originally¹⁵ despite the fact that the binding energy of a valence electron $E_{int} \ll m_e c^2$. While we expect large relativistic effects for tightly bound K- or L-shell electrons in heavy elements (here binding energies are in the keV region), for valence electrons this simple picture predicts negligible relativistic effects. Why this is not is due quantum mechanics at work, and will be analyzed in more detail in section 2.5. For now a few illustrative examples of such strong relativistic valence shell effects will demonstrate that this is indeed the case.

Gold is known to show very large relativistic effects in the 6s valence space.^{14,15} Bulk gold is yellow because of relativistic effects, the oxidation state +III in gold compounds is substantially stabilized by relativistic effects, and the unusual zig-zag structures found in the solid state of gold(I) halides are due to relativistic effects. Mercury is a liquid because of relativistic effects, and the mercury battery ... The lead battery ... Superheavy elements. Explain why

More on history of Dirac equation and difficulties treating the many electron system

2 THE DIRAC EQUATION

2.1 Deriving the Dirac Equation

The Dirac equation can be derived from the time evolution in eq.(6) by a) enforcing Lorentz invariance, b) fulfilling the relativistic energy-momentum relation,** i.e. eq.(9), and c) by minimal coupling (3) introducing both vector and scalar potentials. This automatically yields a (non-reducible)

fections that still remain being in connection with the exact fitting of the theory with relativity ideas. These gives rise to difficulties only when high-speed particles are involved, and are therefore of no importance in the consideration of atomic and molecular structure and ordinary chemical reactions, in which it is, indeed, usually sufficiently accurate if one neglects relativity variation of mass with velocity and assumes only Coulomb forces between the various electrons and atomic nuclei. Even in 1988 Glashow wrote⁵ Modern elementary-particle physics is founded upon the two pillars of quantum mechanics and relativity. I have made little mention of relativity so far because, while the atom is very much a quantum system, it is not very relativistic at all. Relativity becomes important only when velocities become comparable to the speed of light. Electrons in atoms move rather slowly, at a mere one percent of light speed. Thus it is that a satisfactory description of the atom can be obtained without Einstein's revolutionary theory. However, as early as in 1932 Vallarta and Rosen stated that⁶ While this [relativistic] correction may be expected to be negligible for most atoms, it becomes appreciable as the atomic number increases because for very heavy atoms the electron velocities in the vicinity of the nucleus become high. For such atoms the electron density close to the nucleus may be appreciably changed by the relativity correction.

^{**}Direct quantization of the relativistic energy-momentum relation leads to a square-root operator which is not Lorentz invariant. However, in its quadratic form it leads to the Klein-Gordon equation for spin 0 particles.

(4×4) matrix first-order differential equation (1) introducing 4-component spinors, and thus in a natural way the spin of a fermionic particle (such as the electron) and the existence of fermionic anti-particles. However, we will derive the Dirac equation here “through the back-door”, which may not be the correct way, but is less tedious. For this we need a special property of the Pauli matrices. Given two vectors \vec{a} and \vec{b} we have

$$(\vec{a}\vec{\sigma})(\vec{b}\vec{\sigma}) = (\vec{a}\vec{b})I_{2 \times 2} + i\vec{\sigma}(\vec{a} \times \vec{b}) \quad (12)$$

where $I_{2 \times 2}$ is the (2×2) unit matrix. For the special case of $\vec{a} = \vec{b}$ (and we take the momentum vector here) we have

$$(\vec{p}\vec{\sigma})(\vec{p}\vec{\sigma}) = p^2 \quad (13)$$

Now we can do a “magic trick”. Starting with the quantized version of the relativistic energy-momentum relation for an electron of rest mass m_e we have

$$\left(\frac{E^2}{c^2} - p^2\right)\phi_1 = \left(-\hbar^2 \frac{\partial^2}{\partial t^2}\right)\phi_1 = \left[\frac{i\hbar}{c} \frac{\partial}{\partial t} + (\vec{\sigma}\vec{p})\right] \left[\frac{i\hbar}{c} \frac{\partial}{\partial t} - (\vec{\sigma}\vec{p})\right] \phi_1 = (m_e c)^2 \phi_1 \quad (14)$$

If we choose the second expression in square brackets as

$$\left[\frac{i\hbar}{c} \frac{\partial}{\partial t} - (\vec{\sigma}\vec{p})\right] \phi_1 = m_e c \phi_2 \quad (15)$$

This leads to two coupled differential equations,

$$\left[\frac{i\hbar}{c} \frac{\partial}{\partial t} + (\vec{\sigma}\vec{p})\right] \phi_2 = m_e c \phi_1 \quad \text{and} \quad \left[\frac{i\hbar}{c} \frac{\partial}{\partial t} - (\vec{\sigma}\vec{p})\right] \phi_1 = m_e c \phi_2 \quad (16)$$

Introducing the linear combinations $\phi_L = \phi_1 + \phi_2$ and $\phi_S = \phi_1 - \phi_2$ we obtain the two coupled differential equations,

$$\frac{i\hbar}{c} \frac{\partial}{\partial t} \phi_L - (\vec{\sigma}\vec{p}) \phi_S = m_e c \phi_L \quad \text{and} \quad -\frac{i\hbar}{c} \frac{\partial}{\partial t} \phi_S + (\vec{\sigma}\vec{p}) \phi_L = m_e c \phi_S \quad (17)$$

leading after minimal substitution (3) to the Dirac equation (1). ϕ_L and ϕ_S are the large and small component respectively, which will be discussed in more detail in section 2.2.1. The Dirac operator is unbound and the spectrum for a negatively charged spin 1/2 particle of mass m (e.g. an electron or a muon) is shown in Figure 1. For details on the functional analytical properties of the Dirac equation see refs.^{16,17}.

For a free spin 1/2 particle the relativistic energy-momentum reveals an energy spectrum of $E \in \{-\infty, -mc^2\} \cup \{+mc^2, \infty\}$. The correct interpretation of the negative energy continuum

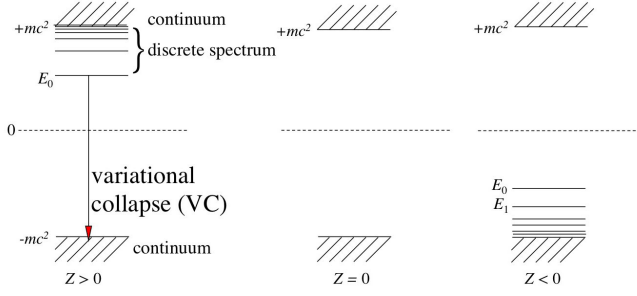


Figure 1: Schematic spectrum of the Dirac operator for nuclear charges $Z > 0$, $Z = 0$ and $Z < 0$.

$\{-\infty, -mc^2\}$ in the spectrum of the Dirac operator has caused many misconceptions to flourish. An electron in a bound or positive energy continuum state could fall into the negative energy continuum (in principle down to $E \rightarrow -\infty$) releasing a photon (variational collapse). We experienced a similar situation in pre-quantum theory where Bohr had to confine electrons in discrete orbits in order not to fall into the positively charged nucleus. Now, here we can do exactly the same (formally) confining electrons to the bound states above the negative energy continuum. The original interpretation of Dirac that the negative energy continuum is completely filled with energy and thus the Pauli principle prevents electrons from entering such states is only of historical importance.^{††} Further, if we use the same philosophy for the Klein-Gordon equation for spin-0 particles (bosons), there is no Pauli principle at work preventing *all* bosons to go to $E \rightarrow -\infty$. A correct interpretation of these negative energy states comes from Feynman and Stueckelberg in quantum field theory which is discussed in the QED chapter 4. If QED effects are ignored and the Dirac equation is treated “as it is”, we simply have to make sure that a variational collapse is prevented (see section 2.4). Formally this can be done by introducing projection operators Λ_+ to the Hilbert sub-space of the states above the negative energy continuum,

$$D_+ = \Lambda_+ D \Lambda_+ \quad (18)$$

This will be discussed in more detail in section 2.4.

Explain the problem with probability density.

^{††}To cite J. Schwinger 1973: *The picture of an infinite sea of negative energy electrons is now best regarded as an historical curiosity, and forgotten. Unfortunately, this episode, and discussions of vacuum fluctuations, seem to have left people with the impression that the vacuum, the physical state of nothingness, is actually the scene of wild activity.*

2.2 Solutions for two-particle systems with opposite charge

2.2.1 The Electron-Nucleus Interaction

The stationary Dirac equation for an electron in an external (classical) Coulomb field is,

$$\left\{ c\vec{\alpha}\vec{p} + \beta mc^2 - Zr^{-1} - (E + mc^2) \right\} \Psi(\vec{r}) = 0 \quad (19)$$

and we shifted the spectrum by $-mc^2$ in order to coincide with the nonrelativistic spectrum from the Schrödinger equation. This can formally be done by replacing the β -matrix, i.e. $\beta' \rightarrow \beta - mc^2$. This leads to two coupled radial differential equations,

In the expression (20) we skipped the center of mass separation, which is rather more difficult for the Dirac equation.^{18–21} Consider a relativistic effective Hamiltonian treating the nucleus of mass M nonrelativistically^{‡‡} except for the rest mass energy,

$$Mc^2 + \frac{P^2}{2M} + \left\{ c\vec{\alpha}\vec{p} + \beta mc^2 - \frac{Z}{|\vec{R} - \vec{r}|} - (E + mc^2) \right\} \Psi(\vec{R}, \vec{r}) = 0 \quad (20)$$

Explain charge conjugation briefly here and positronic states.

Hydrogen atom with hyperfine structure and QED effects

2.2.2 The Coulomb singularity at the origin and finite extension of the nucleus

Nuclear charges greater than 137 and the Coulomb singularity.

2.2.3 The Electron-Positron Interaction

The two-body Dirac equation describing two oppositely charged particles of mass m_1 and m_2 in a center-of-mass-system interacting via a classical Coulomb interactions is,

Positronium

2.2.4 Other two-particle systems

Muonium, quarkonium, etc., problem with confinement, MIT bag model

^{‡‡}There is no need for the nucleus to be treated as a fermionic (half-integer nuclear spin) or bosonic (integer nuclear spin) particle (or even down to the quark level), as for example hyperfine effects can be treated effectively in a two-component form.

2.3 The classical Breit interaction

Introduce Breit in a classical way

2.4 The multi-electron Dirac equation

Multi-electron Dirac equation, variational principle, properties

2.4.1 Treating the multi-electron Dirac equation

Mainly Trond's expertise

2.5 Why are relativistic effects large in valence shell of heavy elements?

Clean up misconceptions

3 TWO-COMPONENT FORMS

3.1 Elimination of the small component

Foly-Wouthuysen, block diagonalizations

3.2 Unitary transformations

General discussion of unitary transformations

3.3 Matrix formulations

Mainly Trond's expertise

3.4 Applications

Examples compare 4- with 2-component

4 QUANTUM ELECTRODYNAMICS

4.1 Introduction to Feynman diagrams

Feynman rules

4.2 Vacuum polarization and self-energy

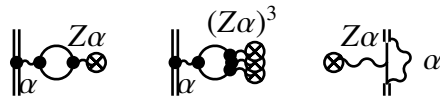


Figure 2: From the left to the right: vacuum polarisation of order $\alpha(Z\alpha)$ (Uehling) and $\alpha(Z\alpha)^3$ (Wichmann-Kroll), and electron self-energy of order $\alpha(Z\alpha)$. Note that according to Furry's theorem diagrams with a loop containing odd number of vertices vanish.²²

4.3 Breit interaction from QED

Frequency dependent Breit, how to treat frequency in CI

4.4 Higher order terms

Few electron systems only

4.5 Applications

Examples

5 BEYOND QUANTUM ELECTRODYNAMICS

5.1 The standard model and beyond

Introduce the standard model, Problems with standard model (dark matter, dark energy, fundamental constants, ...)

5.2 Electroweak interactions

Symmetries, CPT theorem, P, CP violation etc., chirality, anapole moment

5.2.1 The electron dipole moment

Beyond the standard model

5.3 CONCLUSIONS

conclusions

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