

# THE DOUGLAS-KROLL-HESS METHOD: CONVERGENCE AND BLOCK-DIAGONALIZATION OF DIRAC OPERATORS

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ABSTRACT. We show that the Douglas-Kroll block-diagonalization method for the Dirac operator with Coulomb potential is convergent in norm resolvent sense for coupling constant  $\gamma$  less than  $\gamma_c = 0.37758$  which corresponds to atomic number 51. Moreover, we give an explicit expression for the corresponding block-diagonalized Dirac operator.

## 1. INTRODUCTION

The one-particle Dirac operator is given by

$$(1) \quad D_\gamma := \boldsymbol{\alpha} \cdot \frac{1}{i} \nabla + \beta + \gamma V$$

acting in the Hilbert space  $\mathfrak{H} := L^2(\mathbb{R}^3) \otimes \mathbb{C}^4$ . Here  $\boldsymbol{\alpha}, \beta$  are the usual Dirac matrices. In this article we consider the Coulomb potential  $V = -1/|\cdot|$  with the coupling constant  $\gamma \in (-1, 1)$ . (We use units in which the rationalized Planck constant, the mass of the particle, and the velocity of light are equal to one.) We choose  $V$  as a Coulomb potential merely for convenience and definiteness. The method that we develop can be applied – in principle – to much more general situations. In fact it is not even necessary that the unperturbed operator is the free Dirac operator as the attentive reader will gather from the proofs.

It is well known that this operator is unbounded from below. Dirac postulated that negative energy states are already occupied by electrons, i.e., physical electrons are not allowed to exist in such states.

In most situations of chemical interest, pair-creation and excitations of the positronic degrees of freedom are far beyond the energy scale of the valence shell [30]. Therefore, it is a good approximation to neglect these quantum effects and “fill the Dirac sea”. Mathematically this means that one should consider the Dirac operator projected onto its positive spectral subspace, i.e.,  $P(\gamma)D_\gamma P(\gamma)$  in the Hilbert space  $P(\gamma)\mathfrak{H}$ , where  $P(\gamma) := \chi_{(0, \infty)}(D_\gamma)$  is the spectral projection of the Dirac operator  $D_\gamma$  to its positive spectral subspace.

For the free Dirac operator  $D_0$  the corresponding projection  $P(0)$  is well known; in momentum space it is the matrix valued multiplication operator

$$\mathcal{F}P_0\mathcal{F}^{-1} := \frac{E(\mathbf{p}) + \boldsymbol{\alpha} \cdot \mathbf{p} + \beta}{2E(\mathbf{p})}$$

where  $E(\mathbf{p}) := (1 + \mathbf{p}^2)^{1/2}$  and  $\mathcal{F}$  is the Fourier transform. In general, however, the corresponding projections are not explicitly known. Thus, it is important to develop an approximate procedure. One way of doing this was suggested in 1974 by M. Douglas and N.M. Kroll [7]. They start with the free case, i.e.,  $\gamma = 0$ , as the

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initial step. In the free case Foldy and Wouthuysen [9] showed that in momentum space the unitary transform

$$\mathcal{F}U_{\text{FW}}\mathcal{F}^{-1} := \frac{1 + \beta\boldsymbol{\alpha} \cdot \mathbf{p}}{\sqrt{2E(\mathbf{p})(E(\mathbf{p}) + 1)}}$$

maps the operator  $D_0$  into a block-diagonal form, i.e.,

$$\mathcal{F}U_{\text{FW}}D_0U_{\text{FW}}^{-1}\mathcal{F}^{-1} = \begin{pmatrix} E(\mathbf{p}) & 0 \\ 0 & -E(\mathbf{p}) \end{pmatrix}.$$

For non-vanishing  $\gamma$  the decoupling is done perturbatively. The method consists of a series of unitary transforms which are chosen to block-diagonalize the Hamiltonian up to a given order in the coupling constant  $\gamma$ . Every consecutive transform removes the lowest order in  $\gamma$  of the remaining off-diagonal part. This allows the decoupling up to any power.

Brummelhuis et al. [5] observed that the first order term corresponds to the Brown-Ravenhall operator. It was extensively studied in the physics literature by Sucher and Hardekopf [27, 11, 12]. Evans et al. [8], Tix [28, 29], and Balinsky and Evans [1, 2] later showed some basic mathematical properties, e.g., positivity and self-adjointness.

Jansen and Heß [17] corrected an error in the derivation of the second order term by Douglas and Kroll. Later, Samzow et al. [25] extended the method to multi-particle operators. Actually, Heß et al. turned the method together with the efficient handling of the matrix elements of the corresponding two-component Hamiltonian into the “most successful two-components computational tool of the relativistic quantum chemistry” [3]. From a mathematical point of view the Jansen-Heß operator has been studied by Brummelhuis et al. [5, 26], by Jakubaša-Amundsen [14, 16, 15], and by Jakubaša-Amundsen and Iantchenko [13].

However, despite its success the question remains open as to whether proceeding to higher approximation really leads to better results as indicated numerically (Reiher and Wolf [23, 24]). This paper endeavors to fill this gap: we will show that the spectrum of the approximate operators will become arbitrarily close to the spectrum of the Dirac operator as the approximation degree increases.

The organization of the paper is as follows: in Section 2 we review the Douglas-Kroll method. The unitary transform  $U(\gamma)$ , which block-diagonalizes the Dirac operator, is constructed in Section 3. In Section 4 we define the Douglas-Kroll operator of order  $N$  and state our main results. In Section 5, we prove that the operator  $|D_0|^{1/2}U(\gamma)|D_0|^{-1/2}$  is analytic which implies that  $|D_0|^{-1/2}U(\gamma)D_\gamma U(\gamma)^{-1}|D_0|^{-1/2}$  is analytic as well. We use this in Section 6 to prove our main theorem. In Appendix A we state some useful equations. Finally, in Appendix B, we show that the expansion of the block-diagonalized Hamiltonian up to second order in powers of the coupling constant  $\gamma$  indeed yields the Jansen-Heß operator. In addition, however, the expansion gives the possibility of obtaining further corrections to the energy. In fact, we can even guarantee that these contributions yield – when summed – the correct eigenvalue.

## 2. THE DOUGLAS-KROLL METHOD

In order to be self-contained we shortly describe the Douglas-Kroll method (see [17] for details): the first step is to apply the (free) Foldy-Wouthuysen transform

to (1). We get

$$(2) \quad U_{\text{FW}} D_\gamma U_{\text{FW}}^{-1} =: \beta E(\frac{1}{i}\nabla) + \mathcal{E}_1 + \mathcal{O}_1.$$

Here we decompose the difference of the Foldy-Wouthuysen transformed operator and  $\beta E(\frac{1}{i}\nabla)$  into its diagonal part  $\mathcal{E}_1$  and off-diagonal part part, i.e.,

$$(3) \quad \mathcal{E}_1 = \gamma (\beta_+ U_{\text{FW}} V U_{\text{FW}}^{-1} \beta_+ + \beta_- U_{\text{FW}} V U_{\text{FW}}^{-1} \beta_-),$$

$$(4) \quad \mathcal{O}_1 = \gamma \beta_+ U_{\text{FW}} V U_{\text{FW}}^{-1} \beta_- + h.c..$$

Here  $\beta_\pm$  are the orthogonal projection onto the upper respectively lower two components of four spinors, i.e.,  $\beta_\pm = (1 \pm \beta)/2$ .

Note that both  $\mathcal{E}_1$  and  $\mathcal{O}_1$  are of order  $\gamma$ . The idea is then to remove the odd (off-diagonal) term to a higher order in  $\gamma$ . That can be carried out by a unitary transform

$$(5) \quad U_1 = (1 + W_1^2)^{1/2} + W_1,$$

with  $W_1^* = -W_1$ . The operator  $W_1$  is chosen such that — after applying  $U_1$  and expanding in  $\gamma$  — it removes the odd term proportional to  $\gamma$  in the Hamiltonian. This will be the case, if  $W_1$  satisfies the equation  $[\beta E(\frac{1}{i}\nabla), W_1] = \mathcal{O}_1$ . The resulting Hamiltonian can be written as

$$(6) \quad U_1 U_{\text{FW}} D_\gamma U_{\text{FW}}^{-1} U_1^{-1} \\ = \beta E(\frac{1}{i}\nabla) + \mathcal{E}_1 + \beta (W_1 E(\frac{1}{i}\nabla) W_1 + \frac{1}{2} \{W_1^2, E(\frac{1}{i}\nabla)\}) + [W_1, \mathcal{O}_1] + \rho_\gamma^2;$$

the term  $\rho_\gamma^2$  represents operators which are  $O(\gamma^3)$  on the diagonal and  $O(\gamma^2)$  on the off-diagonal part. Those off-diagonal terms of second order in  $\gamma$  can be removed using the method just described. This is the starting point of an iterative procedure which determines Hamiltonians that are formally diagonal up to any desired power in  $\gamma$ . We refer to the work of Wolf et al. [30] for details.

### 3. EXACT BLOCK-DIAGONALIZATION OF THE DIRAC HAMILTONIAN

The Dirac operator can be written in terms of its positive and negative spectral projections as

$$(7) \quad D_\gamma = P(\gamma) D_\gamma P(\gamma) + P^\perp(\gamma) D_\gamma P^\perp(\gamma),$$

here  $P^\perp(\gamma) := 1 - P(\gamma)$ . We note that to block-diagonalize  $D_\gamma$  is equivalent to find a unitary transform  $\tilde{U}(\gamma)$  such that

$$(8) \quad \tilde{U}(\gamma) P(\gamma) \tilde{U}^{-1}(\gamma) = \beta_+.$$

Since  $U_{\text{FW}} P(0) U_{\text{FW}}^* = \beta_+$ , it is enough to find a transform  $U(\gamma)$  such that

$$(9) \quad U(\gamma) P(\gamma) U^{-1}(\gamma) = P(0).$$

Assuming the existence of such a transform  $U(\gamma)$ , we can say that  $D_\gamma$  is unitarily equivalent to the operator

$$(10) \quad \beta_+ U_{\text{FW}} U(\gamma) D_\gamma U^{-1}(\gamma) U_{\text{FW}}^{-1} \beta_+ + \beta_- U_{\text{FW}} U(\gamma) D_\gamma U^{-1}(\gamma) U_{\text{FW}}^{-1} \beta_-$$

which is block-diagonal. Here the terms in the sum correspond to the electronic and positronic states respectively. Both terms can be viewed as acting on  $L^2(\mathbb{R}^3) \otimes \mathbb{C}^2$  in a canonical way.

In the following we will construct such a unitary map  $U(\gamma)$  and show its desired properties. We begin by recalling a property of the operator

$$(11) \quad T_\eta := |x|^{-1/2}(D_0 - i\eta)^{-1}|x|^{-1/2}$$

with  $\eta \in \mathbb{R}$ : Nenciu [22] conjectured and Kato [19], Formula 2.5, proved

$$(12) \quad \|T_\eta\| \leq 1.$$

In the next lemmata we will show some basic properties of  $P(\gamma)$ .

**Lemma 1.** *The projection  $P(\gamma)$  has an analytic continuation into the disc  $\mathcal{D} := \{\gamma \in \mathbb{C} \mid |\gamma| < 1\}$ .*

*Proof.* Using the resolvent expansion given in [22], Lemma 2.2., we have for real  $\gamma$  in the above disc

$$(13) \quad \frac{1}{D_\gamma - i\eta} = \frac{1}{D_0 - i\eta} + \gamma \frac{1}{D_0 - i\eta} |V|^{1/2} \frac{1}{1 - \gamma T_\eta} |V|^{1/2} \frac{1}{D_0 - i\eta}.$$

We note that the right hand side is meaningful and norm-analytic in  $\mathcal{D}$ , since  $\|\gamma T_\eta\| < 1$  because of (12). This follows immediately by expanding  $(1 - \gamma T_\eta)^{-1}$ .

To prove the analyticity of  $P(\gamma)$  we use

$$(14) \quad P(\gamma) = \frac{1}{2} + \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{1}{D_\gamma - i\eta} d\eta$$

where the integral is meant as Cauchy principal value in the strong sense (see Kato [18], Lemma VI-5.6). Therefore,

$$(15) \quad \begin{aligned} P(\gamma) - P(0) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} (D_\gamma - i\eta)^{-1} - (D_0 - i\eta)^{-1} d\eta \\ &= \int_{-\infty}^{\infty} \sum_{n=0}^{\infty} \gamma^n (D_0 - i\eta)^{-1} |V|^{1/2} (T_\eta)^n |V|^{1/2} (D_0 - i\eta)^{-1} d\eta. \end{aligned}$$

The integrand as function of  $(n, \eta)$  is integrable in both the counting measure in  $n$  and the Lebesgue measure in  $\eta$ . In fact,

$$(16) \quad \begin{aligned} &\|\gamma^n (D_0 - i\eta)^{-1} |V|^{1/2} (T_\eta)^n |V|^{1/2} (D_0 - i\eta)^{-1}\| \\ &= \gamma^n \sup_{f, g \in \mathfrak{H}} \frac{|(f, (D_0 - i\eta)^{-1} |V|^{1/2} (T_\eta)^n |V|^{1/2} (D_0 - i\eta)^{-1} g)|}{\|f\| \|g\|} \\ &\leq \frac{\pi}{2} \gamma^n \sup_{f, g \in \mathfrak{H}} \frac{\| |D_0|^{1/2} (D_0 + i\eta)^{-1} f \| \| |D_0|^{1/2} (D_0 - i\eta)^{-1} g \|}{\|f\| \|g\|} \end{aligned}$$

where we used (12) followed by Kato's inequality  $|\cdot|^{-1} \leq (\pi/2)\sqrt{-\Delta}$ . Furthermore, setting

$$(17) \quad G_\gamma(\eta) := |D_\gamma|^{1/2} (D_\gamma - i\eta)^{-1},$$

we have

$$(18) \quad \int_{-\infty}^{\infty} \|G_\gamma(\eta)\varphi\|^2 d\eta = \pi\|\varphi\|^2$$

for  $\varphi \in \mathfrak{H}$  and  $\gamma \in (-1, 1)$ . Therefore, using the Schwarz inequality

$$(19) \quad \int_{-\infty}^{\infty} d\eta \|G_{\gamma'}(\eta)f\| \|G_\gamma(\eta)g\| \leq \pi\|f\| \|g\|$$

for  $|\gamma'| < 1$  and  $f, g \in \mathfrak{H}$ . Thus, the right hand side of (16) is integrable, and the lemma is proved.  $\square$

**Lemma 2.** *For  $\gamma \in (-\sqrt{3}/2, \sqrt{3}/2)$  we have the following estimate*

$$(20) \quad \|P(\gamma) - P(0)\| \leq \frac{\gamma^2 \pi}{4} \nu_\gamma^{-1/2}.$$

Here  $\nu_\gamma$  is the constant defined in Appendix A. In particular, we have  $\|P(\gamma) - P_0\| < 1$  for  $|\gamma| < 0.68418$ .

*Proof.* Using the resolvent equation in (14), we write  $P(\gamma) = P_0 + \gamma P_1 + Q_\gamma$  where  $P_0 = P(0)$ ,  $P_1 := \frac{1}{2\pi} \int_{-\infty}^{\infty} d\eta (D_0 - i\eta)^{-1} V (D_0 - i\eta)^{-1}$ , and  $Q_\gamma := \frac{\gamma^2}{2\pi} \int_{-\infty}^{\infty} d\eta (D_0 - i\eta)^{-1} V (D_0 - i\eta)^{-1} V (D_\gamma - i\eta)^{-1}$ . Noting that  $(P_0 - P(\gamma))^2 = 1 - P_0 P(\gamma) P_0 - P_0^\perp P(\gamma)^\perp P_0^\perp$  and also that  $P_0 P_1 P_0 = 0 = P_0^\perp P_1 P_0^\perp$  we have

$$(21) \quad \begin{aligned} \|(P_0 - P(\gamma))\psi\|^2 &= (P_0^\perp \psi, Q_\gamma P_0^\perp \psi) - (P_0 \psi, Q_\gamma P_0 \psi) \\ &\leq \|Q_\gamma\| (\|P_0^\perp \psi\|^2 + \|P_0 \psi\|^2) = \|Q_\gamma\| \|\psi\|^2 \end{aligned}$$

To compute the norm of  $Q_\gamma$  we estimate for  $f, g \in \mathfrak{H}$

$$(22) \quad \begin{aligned} |(f, (D_0 - i\eta)^{-1} V (D_0 - i\eta)^{-1} V (D_\gamma - i\eta)^{-1} g)| \\ &\leq \|T_{-\eta} |V|^{1/2} (D_0 + i\eta)^{-1} f\| \| |V|^{1/2} (D_\gamma - i\eta)^{-1} g \|, \\ &\leq \frac{\pi}{2} \| |D_0|^{1/2} (D_0 + i\eta)^{-1} f \| \| |D_0|^{1/2} (D_\gamma - i\eta)^{-1} g \|, \\ &\leq \nu_\gamma^{-1/2} \frac{\pi}{2} \|G_0(\eta) f\| \|G_\gamma(\eta) g\|, \end{aligned}$$

where we have used (12), Kato's inequality, and (43). The lemma follows now from (19).  $\square$

**Theorem 1.** *For  $|\gamma| < 0.68418$*

$$(23) \quad U(\gamma) = (P_0 P(\gamma) + P_0^\perp P^\perp(\gamma)) (1 - (P_0 - P(\gamma))^2)^{-1/2},$$

*is analytic in  $\gamma$  and unitary. Moreover, it fulfills  $U(\gamma) P(\gamma) U^*(\gamma) = P_0$ .*

*Proof.* The operator is well defined due to Lemma 2. The unitarity as well as the Property (9) can be easily verified using that  $(P_0 - P(\gamma))^2$  is equal to  $1 - (P_0 P(\gamma) P_0 + P_0^\perp P(\gamma)^\perp P_0^\perp)$  and commutes with  $P_0$  and  $P(\gamma)$ . Finally, the analyticity follows from Lemma 1 and the fact that  $(1 - A)^{-1/2}$  has a convergent expansion in powers of  $A$  as long as  $\|A\| < 1$ .  $\square$

First, we note that the above strategy of diagonalization can be found in [18] but has not been applied to Dirac operators in this context. The block diagonalization of such operators under the condition that the potential is bounded by 1 has been carried out by Langer and Tretter [20] by different means.

Next, we remark that the transform  $U(\gamma)$  is not unique: Already Kato commented [18], II-4.2, that there are other ways to derive such transforms which allow to relax the condition on  $\gamma$ . However, they have the disadvantage that the expressions are recursively determined instead of being given in a closed algebraic form. Nevertheless, all of these transforms coincide up to order  $\gamma^2$  (inclusive). This feature was also used in [30], where they explore the fact that the ansatz in (5) is not unique. For another expansion method see [16].

## 4. DEFINITION OF THE DOUGLAS-KROLL OPERATOR

The fully block-diagonalized Dirac operator is

$$(24) \quad H_\gamma := \beta_+ U_{\text{FW}} U(\gamma) D_\gamma U^{-1}(\gamma) U_{\text{FW}}^{-1} \beta_+ + \beta_- U_{\text{FW}} U(\gamma) D_\gamma U^{-1}(\gamma) U_{\text{FW}}^{-1} \beta_-.$$

In order to make contact with the Douglas-Kroll method we consider the expansion in  $\gamma$  of  $U(\gamma) D_\gamma U^{-1}(\gamma)$  obtaining the formal operator

$$(25) \quad \sum_{j=0}^N \gamma^j \sum_{k=0}^j u_k D_0 u_{j-k}^* + \sum_{j=0}^{N-1} \gamma^{j+1} \sum_{k=0}^j u_k V u_{j-k}^*.$$

Here  $u_j$ , for  $j \in \mathbb{N}$ , are the coefficients of the expansion of  $U(\gamma)$ , i.e.,

$$(26) \quad U(\gamma) = \sum_{j=0}^{\infty} \gamma^j u_j.$$

Let us define the operator

$$(27) \quad R_\gamma^N := U(\gamma) D_\gamma U(\gamma)^{-1} - \sum_{j=0}^N \gamma^j \sum_{k=0}^j u_k D_0 u_{j-k}^* - \sum_{j=0}^{N-1} \gamma^{j+1} \sum_{k=0}^j u_k V u_{j-k}^*$$

acting in  $C_0^\infty(\mathbb{R}^3)^4$ . We can now formulate one of our main results.

**Theorem 2.** *If  $\gamma$  or  $1/N$  are small enough, the operator  $U(\gamma) D_\gamma U(\gamma)^{-1} - R_\gamma^N$  admits a distinguished self-adjoint extension, the pseudo-Friedrichs extension  $d_\gamma^N$  for which  $D(d_\gamma^N) \subset D(|D_0|^{1/2}) \equiv H^{1/2}(\mathbb{R}^3)^4$ . Moreover, for  $|\gamma| < \gamma_c$  with  $\gamma_c = 0.37758$ , the operator  $d_\gamma^N$  converges in norm resolvent sense to  $U(\gamma) D_\gamma U(\gamma)^{-1}$ .*

We are not yet in a position to give the proof of this result. We postpone it to Section 6 and define instead the  $N$ -th order Douglas-Kroll operator as

$$(28) \quad H_\gamma^N := \beta_+ U_{\text{FW}} d_\gamma^N U_{\text{FW}}^{-1} \beta_+ + \beta_- U_{\text{FW}} d_\gamma^N U_{\text{FW}}^{-1} \beta_-.$$

One can explicitly verify that the first terms correspond precisely to the ones obtained in [17] (see Appendix B). The important fact following from Theorem 2 is, that the eigenvalues of  $d_\gamma^N$  converge to the eigenvalues of  $D_\gamma$  as the degree  $N$  of the approximation tends to infinity:

**Corollary 1.** *For  $\gamma < \gamma_c$  hold: If  $\lambda \in \mathbb{R} \setminus \sigma(D_\gamma)$ , then  $\lambda \notin \sigma(d_\gamma^N)$  for all large enough  $N$ . If  $\lambda \in \sigma(D_\gamma)$ , then there exists  $\lambda_N \in \sigma(d_\gamma^N)$  for which  $\lim_{N \rightarrow \infty} \lambda_N = \lambda$ .*

For a proof of this result see e.g. [6] Chapter 2.

5. ANALYTICITY OF  $|D_0|^{1/2} U(\gamma) |D_0|^{-1/2}$ 

We first need some technical results:

**Lemma 3.** *The operator  $|D_0|^{1/2} (P(\gamma) - P_0) |D_0|^{-1/2}$  has an analytic continuation into the disc  $\mathcal{D} := \{\gamma \in \mathbb{C} \mid |\gamma| < 1/2\}$ .*

*Proof.* We follow the same strategy as in Lemma 1. The analyticity follows from (15), the fact that  $|V|^{1/2} T_\eta^n = (|V|(D_0 - i\eta)^{-1})^n |V|^{1/2}$ , and the following estimate

$$(29) \quad \begin{aligned} & \|\gamma^n |D_0|^{1/2} (D_0 - i\eta)^{-1} (|V|(D_0 - i\eta)^{-1})^n |V| |D_0|^{-1/2} (D_0 - i\eta)^{-1}\| \\ & \leq 2(2\gamma)^n \sup_{f, g \in \mathfrak{S}} \|G_0(\eta) f\| \|G_0(\eta) g\| \end{aligned}$$

where we used Hardy's inequality  $|\cdot|^{-2} \leq 4|D_0|^2$ .  $\square$

**Lemma 4.** For  $\gamma \in (-\sqrt{3}/2, \sqrt{3}/2)$  we have the following estimate

$$(30) \quad \||D_0|^{1/2}(P(\gamma) - P_0)|D_0|^{-1/2}\| \leq \gamma\nu_\gamma^{-1} \sqrt{1 + \frac{\pi}{2}\gamma}.$$

In particular, we have  $\||D_0|^{1/2}(P(\gamma) - P_0)|D_0|^{-1/2}\| < 1$  for  $|\gamma| < \gamma_c$ .

*Proof.* For  $f, g \in H^{1/2}(\mathbb{R}^3)^4$  we have

$$(31) \quad \begin{aligned} & |(f, |D_0|^{1/2}(P(\gamma) - P_0)|D_0|^{-1/2}g)| \\ &= \frac{\gamma}{2\pi} \left| \int_{-\infty}^{\infty} (f, |D_0|^{1/2}(D_0 - i\eta)^{-1}V(D_\gamma - i\eta)^{-1}|D_0|^{-1/2}g)d\eta \right| \\ &\leq \frac{\gamma}{2\pi} \int_{-\infty}^{\infty} \||D_0|^{1/2}(D_0 + i\eta)^{-1}f\| \| |V|(D_\gamma - i\eta)^{-1}|D_0|^{-1/2}g \| d\eta. \end{aligned}$$

Now, using Hardy's inequality and (43) we estimate

$$(32) \quad \begin{aligned} \||V|(D_\gamma - i\eta)^{-1}|D_0|^{-1/2}g\| &\leq 2\||D_0|(D_\gamma - i\eta)^{-1}|D_0|^{-1/2}g\| \\ &\leq 2\nu_\gamma^{-1} \||D_\gamma|(D_\gamma - i\eta)^{-1}|D_0|^{-1/2}g\|. \end{aligned}$$

Therefore, (19) implies

$$(33) \quad |(f, |D_0|^{1/2}(P(\gamma) - P_0)|D_0|^{-1/2}g)| \leq \gamma\nu_\gamma^{-1} \|f\| \||D_\gamma|^{1/2}|D_0|^{-1/2}g\|$$

which leads to the desired result after using Kato's inequality on the right hand side of the latter expression.  $\square$

**Lemma 5.** The operator  $|D_0|^{1/2}U(\gamma)|D_0|^{-1/2}$  is bounded and analytic in  $\gamma$  for  $|\gamma| < \gamma_c$ .

*Proof.* Using properties of orthogonal projections we have the operator identity  $P_0P(\gamma) + P_0^\perp P(\gamma)^\perp = 1 - (P_0^\perp - P_0)(P(\gamma) - P_0)$ . Inserting the latter equation in (23) we write

$$(34) \quad U(\gamma) = (1 - (P_0^\perp - P_0)(P(\gamma) - P_0))(1 - (P_0 - P(\gamma))^2)^{-1/2}.$$

Let us note first that

$$(35) \quad \begin{aligned} |D_0|^{1/2}(1 - (P_0^\perp - P_0)(P(\gamma) - P_0))|D_0|^{-1/2} \\ = 1 - (P_0^\perp - P_0)|D_0|^{1/2}(P(\gamma) - P_0)|D_0|^{-1/2} \end{aligned}$$

is bounded by Lemma 4. Secondly we use (44) and look at

$$(36) \quad \begin{aligned} |D_0|^{1/2}(1 - (P_0 - P(\gamma))^2)^{-1/2}|D_0|^{-1/2} \\ = \frac{1}{\pi} \int_{-1}^1 \frac{1}{\sqrt{1-y^2}} |D_0|^{1/2} \frac{1}{(1-y(P_0 - P(\gamma)))} |D_0|^{-1/2} dy \\ = \frac{1}{\pi} \int_{-1}^1 \frac{1}{\sqrt{1-y^2}} \frac{1}{(1-y|D_0|^{1/2}(P_0 - P(\gamma))|D_0|^{-1/2})} dy. \end{aligned}$$

By Lemma 4 we can estimate the operator inside the integral above as

$$(37) \quad \begin{aligned} (1 - y|D_0|^{1/2}(P_0 - P(\gamma))|D_0|^{-1/2})^{-1} \\ \leq [1 - |y| \||D_0|^{1/2}(P_0 - P(\gamma))|D_0|^{-1/2}\|]^{-1}. \end{aligned}$$

Therefore,

$$(38) \quad \begin{aligned} & \| |D_0|^{1/2} (1 - (P_0 - P(\gamma))^2)^{-1/2} |D_0|^{-1/2} \| \\ & \leq [1 - \| |D_0|^{1/2} (P_0 - P(\gamma)) |D_0|^{-1/2} \|]^{-1} \end{aligned}$$

which together with Lemmata 3 and 4 completes the proof.  $\square$

## 6. CONVERGENCE OF THE DOUGLAS-KROLL HAMILTONIAN

**Lemma 6.** *For  $\gamma < \gamma_c$ , the operator  $|D_0|^{-1/2} U(\gamma) D_\gamma U(\gamma)^{-1} |D_0|^{-1/2}$  is bounded and analytic in  $\mathfrak{H}$ .*

*Proof.* We write

$$(39) \quad \begin{aligned} |D_0|^{-1/2} U(\gamma) D_\gamma U(\gamma)^{-1} |D_0|^{-1/2} &= (|D_0|^{-1/2} U(\gamma) |D_0|^{1/2}) \\ &\quad \times (|D_0|^{-1/2} D_\gamma |D_0|^{-1/2}) (|D_0|^{1/2} U(\gamma)^{-1} |D_0|^{-1/2}) \end{aligned}$$

and observe that each term is bounded and analytic by Lemma 5 in one case or by Kato's inequality in the other.  $\square$

An immediate consequence of Lemma 6 is that, for  $\gamma < \gamma_c$

$$(40) \quad \left\| |D_0|^{-1/2} R_\gamma^N |D_0|^{-1/2} \right\| \rightarrow 0,$$

as  $N \rightarrow \infty$ .

*Proof of Theorem 2.* Note first that for  $u \in C_0^\infty(\mathbb{R}^3)^4$  the following estimate holds

$$(41) \quad \begin{aligned} & \left| (u, |D_\gamma|^{-\frac{1}{2}} U^{-1}(\gamma) R_\gamma^N U(\gamma) |D_\gamma|^{-\frac{1}{2}} u) \right| \\ & \leq \left\| |D_0|^{-\frac{1}{2}} R_\gamma^N |D_0|^{-\frac{1}{2}} \right\| \left\| |D_0|^{\frac{1}{2}} U(\gamma) |D_0|^{-\frac{1}{2}} \right\|^2 \left\| |D_\gamma|^{-\frac{1}{2}} |D_0| |D_\gamma|^{-\frac{1}{2}} \right\| \|u\|^2. \end{aligned}$$

The left hand side is obviously bounded for  $\gamma < \gamma_c$ . We proceed now to prove the self-adjointness of  $d_\gamma^N$ . We will now take the Pseudo-Friedrichs extension (see [18] Theorem VI-3.11) of this operator. The operator  $U(\gamma)$  leaves the Sobolev space  $H^{1/2}(\mathbb{R}^3)^4$  invariant by Lemma 5. Thus it is enough to prove

$$(42) \quad |(u, U^{-1}(\gamma) R_\gamma^N U(\gamma) u)| \leq b_{\gamma, N}(u, |D_\gamma| u) \quad u \in C_0^\infty(\mathbb{R}^3)^4.$$

with  $b_{\gamma, N} < 1$ . This, however, is clearly true because (41), if  $\gamma$  is small or  $N$  is big.

Finally, the resolvent convergence of  $d_\gamma^N$  to  $U(\gamma) D_\gamma U^{-1}(\gamma)$  follows from Corollary VI-3.12 in [18] and (40).  $\square$

## APPENDIX A. SOME USEFUL EQUATIONS

We introduce the constant  $\nu_\gamma^2 := (1/2)\{1 + C_\gamma^2 - [(1 - C_\gamma^2)^2 + 4\gamma^2 C_\gamma^2]^{1/2}\}$  with  $C_\gamma := (1/3)[(4\gamma^2 + 9)^{1/2} - 4\gamma]$ . Then

**Lemma 7** (Morozov [21]). *For  $\gamma \in (-\sqrt{3}/2, \sqrt{3}/2)$  we have*

$$(43) \quad |D_\gamma|^2 \geq \nu_\gamma^2 |D_0|^2.$$



We note that this result generalizes [4], Inequality (2) to the massive case.

The following formula is useful for an integral representation of the unitary operator  $U(\gamma)$ . For  $|a| < 1$  we have

$$(44) \quad (1 - a^2)^{-1/2} = \frac{1}{\pi} \int_{-1}^1 \frac{1}{\sqrt{1-y^2}} \frac{1}{(1-ya)} dy.$$

In order to verify the above see, e.g., [10] Formula 3.197.4.

## APPENDIX B. DERIVATION OF THE HAMILTONIAN

In this section we explicitly derive the block-diagonalized Hamiltonians up to the order  $N = 2$ . In particular, we will look at (28) and compare it with (6). Expanding the projection  $P(\gamma)$  yields for  $\gamma < 1$

$$(45) \quad P(\gamma) = \sum_{n=0}^{\infty} \gamma^n P_n$$

with

$$(46) \quad P_n = \frac{1}{2\pi} \int_{-\infty}^{\infty} \left( \frac{-1}{D_0 - i\eta} V \right)^n \frac{1}{D_0 - i\eta} d\eta.$$

Next, we compute the first coefficients of the expansion of  $U(\gamma)$  as given by (23) and (26) and get

$$(47) \quad \begin{aligned} u_0 &= 1, \\ u_1 &= (P_0 - P_0^\perp) P_1, \\ u_2 &= (P_0 - P_0^\perp) P_2 + \frac{1}{2} P_1^2, \end{aligned}$$

here  $P_i$  with  $i \in \mathbb{N}$  are given in (46). The following properties for the  $P_i$  stem from the fact that  $P(\gamma)$  is idempotent, and will be used later:

$$(48) \quad P_1 = P_0 P_1 + P_1 P_0,$$

$$(49) \quad P_2 = P_2 P_1 + P_2 P_0 + P_1^2.$$

For  $N = 0$  we have  $d_\gamma^0 = D_0$ . Therefore,

$$(50) \quad H_\gamma^0 = U_{\text{FW}} P_0 D_0 P_0 U_{\text{FW}}^{-1} + U_{\text{FW}} P_0^\perp D_0 P_0^\perp U_{\text{FW}}^{-1} = \beta \sqrt{-\Delta + m},$$

which corresponds to the first term in (6). In the above we used

$$(51) \quad \beta_+ U_{\text{FW}} = U_{\text{FW}} P_0.$$

For  $N = 1$  we have

$$(52) \quad d_\gamma^1 = d_\gamma^0 + \gamma D_0 u_1^* + \gamma u_1 D_0 + \gamma V,$$

the terms  $u_1 D_0$  and its adjoint do not contribute to  $H_\gamma^1$ . In fact, using that (51) in (28), and the fact that since (48) holds,  $P_0 P_1 P_0 = P_0^\perp P_1 P_0^\perp = 0$  we can eliminate those terms. Therefore,

$$(53) \quad H_\gamma^1 = U_{\text{FW}} P_0 (D_0 + \gamma V) P_0 U_{\text{FW}}^{-1} + U_{\text{FW}} P_0^\perp (D_0 + \gamma V) P_0^\perp U_{\text{FW}}^{-1},$$

which corresponds to the first two terms in (6). The electronic part is the Brown-Ravenhall operator. Note that  $\mathcal{E}$  in (6) fulfills  $U_{\text{FW}}^{-1} \mathcal{E} U_{\text{FW}} = P_0 \gamma V P_0 + P_0^\perp \gamma V P_0^\perp$ .

Finally, we consider the case  $N = 2$ , we have

$$(54) \quad d_\gamma^2 = d_\gamma^1 + \gamma^2 (D_0 u_2^* + u_2 D_0 + u_1 D_0 u_1^* + u_1 V + V u_1^*)$$

We note that

$$(55) \quad \begin{aligned} P_0 u_2 P_0 &= P_0 (P_0 P_2 P_0 + \frac{1}{2} P_1^2) P_0 = P_0 (-\frac{1}{2} P_1^2) P_0, \\ P_0^\perp u_2 P_0^\perp &= P_0^\perp (-\frac{1}{2} P_1^2) P_0^\perp \end{aligned}$$

where we used (49) projected by  $P_0$  and  $P_0^\perp$ , respectively. Therefore,

$$(56) \quad \begin{aligned} H_\gamma^2 &= H_\gamma^1 + U_{\text{FW}} P_0 \left( P_1 D_0 P_1 - \left\{ \frac{1}{2} P_1^2, D_0 \right\} + P_1 V + V P_1 \right) P_0 U_{\text{FW}}^{-1} \\ &+ U_{\text{FW}} P_0^\perp \left( P_1 D_0 P_1 - \left\{ \frac{1}{2} P_1^2, D_0 \right\} - P_1 V - V P_1 \right) P_0^\perp U_{\text{FW}}^{-1}, \end{aligned}$$

which are the terms that appear in (6), since

$$\begin{aligned} U_{\text{FW}}^{-1} W_1 U_{\text{FW}} &= (P_0 - P_0^\perp) P_1, \\ U_{\text{FW}}^{-1} \mathcal{O}_1 U_{\text{FW}} &= P_0 \gamma V P_0^\perp + P_0^\perp \gamma V P_0. \end{aligned}$$

Here the electronic part of  $H_\gamma^2$  is the Jansen-Heß operator.

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