A Time–Dependent Born–Oppenheimer Approximation with Exponentially Small Error Estimates

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1 May 2000

Abstract

We present the construction of an exponentially accurate time–dependent Born–Oppenheimer approximation for molecular quantum mechanics.

We study molecular systems whose electron masses are held fixed and whose nuclear masses are proportional to ϵ^{-4}, where ϵ is a small expansion parameter. By optimal truncation of an asymptotic expansion, we construct approximate solutions to the time–dependent Schrödinger equation that agree with exact normalized solutions up to errors whose norms are bounded by $C \exp\left(-\gamma/\epsilon^2\right)$, for some C and $\gamma > 0$.

*Partially Supported by National Science Foundation Grant DMS–9703751.

1 Introduction

In this paper we construct exponentially accurate approximate solutions to the time–dependent Schrödinger equation for a molecular system. The small parameter that governs the approximation is the usual Born–Oppenheimer expansion parameter ϵ, where ϵ^4 is the ratio of the electron mass divided by the mean nuclear mass. The approximate solutions we construct agree with exact solutions up to errors whose norms are bounded by $C \exp\left(-\gamma/\epsilon^2\right)$, for some C and $\gamma > 0$, under analyticity assumptions on the electron Hamiltonian.

The Hamiltonian for a molecular system with K nuclei and $N-K$ electrons moving in l dimensions has the form

$$H(\epsilon) \;=\; \sum_{j=1}^{K} -\frac{\epsilon^4}{2M_j}\,\Delta_{X_j} \;-\; \sum_{j=K+1}^{N} \frac{1}{2m_j}\,\Delta_{X_j} \;+\; \sum_{i<j} V_{ij}(X_i - X_j).$$

Here $X_j \in I\!R^l$ denotes the position of the $j^{\rm th}$ particle, the mass of the $j^{\rm th}$ nucleus is $\epsilon^{-4}M_j$ for $1 \le j \le K$, the mass of the $j^{\rm th}$ electron is m_j for $K+1 \le j \le N$, and the potential between particles i and j is V_{ij}. For convenience, we assume each $M_j = 1$. We set $d = Kl$ and let $X = (X_1,\,X_2,\,\dots,\,X_K) \in I\!R^d$ denote the nuclear configuration vector. We can then decompose $H(\epsilon)$ as

$$H(\epsilon) \;=\; -\frac{\epsilon^4}{2}\,\Delta_X \;+\; h(X).$$

The first term on the right hand side represents the nuclear kinetic energy, and the second is the "electron Hamiltonian" that depends parametrically on X. For each fixed X, $h(X)$ is a self-adjoint operator on the Hilbert space $\mathcal{H}_{\rm el} = L^2(I\!R^{(N-K)l})$.

The time–dependent Schrödinger equation we approximately solve in $L^2(I\!R^d,\,\mathcal{H}_{\rm el})$ as $\epsilon \to 0$ is

$$i\,\epsilon^2\,\frac{\partial\psi}{\partial t} \;=\; -\frac{\epsilon^4}{2}\,\Delta_X\,\psi \;+\; h(X)\,\psi$$

Asymptotic expansions in powers of ϵ of certain solutions to this equation are derived in [7, 8, 9]. We obtain our construction by truncating these expansions after an ϵ–dependent number of terms, in an effort to minimize the norm of the error. Similar strategies have been used to obtain exponentially accurate results for adiabatic approximations [25, 18, 19, 16] and semiclassical approximations [14, 15], both of which play roles in the Born–Oppenheimer approximation we are studying here.

Roughly speaking, the time–dependent Born–Oppenheimer approximation says the following for small ϵ: The electrons move very rapidly and adjust their state adiabatically as the more slowly moving nuclei change their positions. If the electrons start in a discrete energy level of $h(X)$, they will remain in that level. In the process, the electron states create an effective potential in which the motion of the heavy nuclei is well described by a semiclassical approximation. The asymptotic expansions show that this intuition is valid up to errors of order ϵ^k for any k.

In Born–Oppenheimer approximations, adiabatic and semiclassical limits are being taken simultaneously, and they are coupled. Analysis of errors for the adiabatic and semiclassical approximations shows that they are each accurate up to errors whose bounds have the form

$C\exp\left(-\gamma/\epsilon^2\right)$ [17, 14]. Non-adiabatic transitions are known in some systems to be of this order, and tunnelling in semiclassical approximations makes contributions of this order. Thus, one cannot expect to do better than approximations of this type because of two well-known physical phenomena that Born–Oppenheimer approximations do not take into account.

In some systems, tunnelling might dominate the error. In some, non-adiabatic electronic transitions may dominate. In others, the two effects can be of comparable magnitude.

One of the motivations for our work is to generate a "good" basis upon which to build a "surface hopping model" that would accurately describe non-adiabatic electronic transitions. Prior authors (see, *e.g.*, [26, 28, 29, 5]) have proposed such models based on the zeroth order time–dependent Born–Oppenheimer approximation. Using the zeroth order states as a basis of the surface hopping model, the non-adiabatic transitions appear at order ϵ^2. This is huge compared to the exponentially small physical phenomenon one would like to study, and we believe interference between transitions that occur at different times is responsible for the exponential smallness of the physically interesting quantity. Our view is that by choosing a much better set of states on which to base the model, one will obtain a much more useful approximation. Sir Michael Berry [3, 4, 21] has advocated such ideas for the somewhat simpler adiabatic approximation (which does not have the complications of the nuclear motion). These ideas have been used in [16, 18] to prove the accuracy of certain results for non–adiabatic transitions that are exponentially small.

Remarks:

1. There are some other exponentially accurate results in the general topic of Born–Oppenheimer approximations. The prior results come from study of the time–independent Schrödinger equation and depend on global properties of the system. Our results are time–dependent and make use of local information.

Klein [20] and Martinez [22, 23, 24] show that resonances associated with predissociation processes have exponentially long lifetimes. Benchaou and Martinez [1, 2] also show that certain S–matrix elements associated with non–adiabatic transitions are exponentially small.

2. The papers cited in the previous remark obtain estimates that depend on the global structure of the electron energy levels. The results we obtain depend on a particular classical path. When the path stays away from the nuclear configurations where the gap between relevant electonic levels is minimized, one would expect the non–adiabatic errors from our approximation to be smaller, *i.e.*, both results would obtain errors of order $\exp(-\Gamma/\epsilon^2)$, but we would obtain a larger value of Γ.

We expect this because in our case, the Landau–Zener formula predicts that our Γ should come from the minimum gap between eigenvalues on the classical path, rather than the global minimum gap.

3. From a mathematical point of view, the optimal truncation procedure in this context was first stated for the adiabatic approximation for two component systems of ODE's by Berry [3, 4]. It was first proved to yield exponentially accurate results for Hilbert space valued ODE's by Nenciu [25]. In [14, 15] we used this idea for the semiclassical approximation, which is a complex valued PDE setting. The present paper can be viewed as extending these ideas to a Hilbert space valued PDE setting.

1.1 Hypotheses

We assume that the electron Hamiltonian $h(X)$ satisfies the following analyticity hypotheses:

H$_0$ **(i)** For any $X \in I\!\!R^d$, $h(X)$ is a self-adjoint operator on some dense domain $\mathcal{D} \subset \mathcal{H}_{\rm el}$, where $\mathcal{H}_{\rm el}$ is the electronic Hilbert space. We assume the domain $\mathcal{D}$ is independent of X and $h(X)$ is bounded from below uniformly in $I\!\!R^d$.

(ii) There exists a $\delta > 0$, such that for every $\psi \in \mathcal{D}$, the vector $h(X)\psi$ is analytic in $S_\delta = \{z \in \mathbb{C}^d \;:\; |\mathrm{Im}(z_j)| < \delta, \;\; j = 1, \dots, d\}$.

H$_1$ There exists an open set $\Xi \subset I\!\!R^d$, such that for all $X \in \Xi$, there exists an isolated, multiplicity one eigenvalue $E(X)$ of $h(X)$ associated with a normalized eigenvector $\Phi(X) \in \mathcal{H}_{\rm el}$. We assume without loss that the origin belongs to Ξ.

Remarks: 1. Hypothesis $\mathbf{H}_0$ implies that the family of operators $\{h(X)\}_{X \in S_\delta}$ is a holomorphic family of type A.

2. It follows from $\mathbf{H}_0$ and $\mathbf{H}_1$ that there exists $\delta' \in (0, \delta)$ and $\Xi' \subset \Xi$ such that the complex and vector valued functions $E(\cdot)$ and $\Phi(\cdot)$ admit analytic continuations on the set $\Sigma_{\delta'} = \{z \in \mathbb{C}^d \;:\; \mathrm{Re}(z) \in \Xi' \quad \text{and} \quad |\mathrm{Im}(z_j)| < \delta', \;\; j = 1, \dots, d\}$.

1.2 Summary of the Main Results

Our main results are stated precisely as Theorem 4.1 in Section 4. Two generalizations of this result are presented in Section 8.

Roughly speaking, Theorem 4.1 states the following:

Under hypotheses $\mathbf{H}_0$ and $\mathbf{H}_1$, we construct $\Psi_*(X, t, \epsilon)$ (that depends on a parameter g) for $t \in [0, T]$. For small values of g, there exist $C(g)$ and $\Gamma(g) > 0$, such that in the limit $\epsilon \to 0$,

$$\left\| \mathrm{e}^{-itH(\epsilon)/\epsilon^2} \Psi_*(X, 0, \epsilon) \;-\; \Psi_*(X, t, \epsilon) \right\|_{L^2(\mathbb{R}^d, \mathcal{H}_{\rm el})} \;\leq\; C(g)\; \mathrm{e}^{-\Gamma(g)/\epsilon^2}$$

In the state $\Psi_*(X, t, \epsilon)$, the electrons have a high probability of being in the electron state $\Phi(X)$. For any $b > 0$ and sufficiently small values of g, the nuclei are localized near a classical path $a(t)$ in the sense that there exist $c(g)$ and $\gamma(g) > 0$, such that in the limit $\epsilon \to 0$,

$$\left(\int_{|X - a(t)| > b} \|\Psi_*(X, t, \epsilon)\|_{\mathcal{H}_{\rm el}}^2 \, dx \right)^{1/2} \;\leq\; c(g)\; \mathrm{e}^{-\gamma(g)/\epsilon^2}.$$

The mechanics of the nuclear configuration $a(t)$ is determined by classical dynamics in the effective potential $E(X)$.

Two theorems in Section 8 generalize this result. The first allows the time interval to grow as ϵ tends to zero. The second allows more general initial conditions.

2 Coherent States and Classical Dynamics

In the construction of our approximation to the solution of the molecular Schrödinger equation, we need wave packets that describe the semiclassical dynamics of the heavy nuclei. In the present context, the semiclassical parameter is $\hbar = \epsilon^2$. We make use of a convenient set of coherent states (also called generalized squeezed states), that we express here in terms of the semiclassical parameter $\hbar$.

We recall the definition of the coherent states $\varphi_j(A,\, B,\, \hbar,\, a,\, \eta,\, X)$ that are described in detail in [13]. A more explicit, but more complicated definition is given in [12].

We adopt the standard multi-index notation. A multi-index $j = (j_1,\, j_2,\, \dots,\, j_d)$ is a d-tuple of non-negative integers. We define $|j| = \sum_{k=1}^{d} j_k$, $\quad X^j = X_1^{j_1} X_2^{j_2} \cdots X_d^{j_d}$, $j! = (j_1!)(j_2!)\cdots(j_d!)$, and $D^j = \frac{\partial^{|j|}}{(\partial X_1)^{j_1}(\partial X_2)^{j_2}\cdots(\partial X_d)^{j_d}}$.

Throughout the paper we assume $a \in I\!\!R^d$, $\eta \in I\!\!R^d$ and $\hbar > 0$. We also assume that A and B are $d \times d$ complex invertible matrices that satisfy

$$\begin{aligned} A^t\, B \,-\, B^t\, A &= 0, \\ A^*\, B \,+\, B^*\, A &= 2\, I. \end{aligned} \tag{2.1}$$

These conditions guarantee that both the real and imaginary parts of BA^{-1} are symmetric. Furthermore, $\mathrm{Re}\, BA^{-1}$ is strictly positive definite, and $(\mathrm{Re}\, BA^{-1})^{-1} = A\, A^*$.

Our definition of $\varphi_j(A,\, B,\, \hbar,\, a,\, \eta,\, X)$ is based on the following raising operators that are defined for $m = 1,\, 2,\, \dots,\, d$.

$$\mathcal{A}_m(A, B, \hbar, a, \eta)^* \;=\; \frac{1}{\sqrt{2\hbar}} \left[\sum_{n=1}^{d} \overline{B}_{n\,m}\,(X_n - a_n) \;-\; i\, \sum_{n=1}^{d} \overline{A}_{n\,m}\,(-i\hbar \frac{\partial}{\partial X_n} - \eta_n) \right].$$

The corresponding lowering operators $\mathcal{A}_m(A, B, \hbar, a, \eta)$ are their formal adjoints.

These operators satisfy commutation relations that lead to the properties of the $\varphi_j(A,\, B,\, \hbar,\, a,\, \eta,\, X)$ that we list below. The raising operators $\mathcal{A}_m(A, B, \hbar, a, \eta)^*$ for $m = 1,\, 2,\, \dots,\, d$ commute with one another, and the lowering operators $\mathcal{A}_m(A, B, \hbar, a, \eta)$ commute with one another. However,

$$\mathcal{A}_m(A, B, \hbar, a, \eta)\, \mathcal{A}_n(A, B, \hbar, a, \eta)^* \,-\, \mathcal{A}_n(A, B, \hbar, a, \eta)^*\, \mathcal{A}_m(A, B, \hbar, a, \eta) \;=\; \delta_{m,n}.$$

Definition For the multi-index $j = 0$, we define the normalized complex Gaussian wave packet (modulo the sign of a square root) by

$$\begin{aligned} \varphi_0(A,\, B,\, \hbar,\, a,\, \eta,\, X) \;&=\; \pi^{-d/4}\, \hbar^{-d/4}\, (\det(A))^{-1/2} \\ &\times\quad \exp\Big\{ -\langle\, (X-a),\, B\, A^{-1}\, (X-a)\, \rangle/(2\hbar) \,+\, i\,\langle\, \eta,\, (X-a)\, \rangle/\hbar \Big\}. \end{aligned}$$

Then, for any non-zero multi-index j, we define

$$\begin{aligned} \varphi_j(A,\, B,\, \hbar,\, a,\, \eta,\, \cdot\,) \;&=\; \frac{1}{\sqrt{j!}}\; (\,\mathcal{A}_1(A, B, \hbar, a, \eta)^*)^{j_1}\; (\,\mathcal{A}_2(A, B, \hbar, a, \eta)^*)^{j_2}\; \cdots \\ &\times\; (\,\mathcal{A}_d(A, B, \hbar, a, \eta)^*)^{j_d}\;\; \varphi_0(A\,, B,\, \hbar,\, a,\, \eta,\, \cdot\,). \end{aligned}$$

Properties 1. For $A = B = I$, $\hbar = 1$, and $a = \eta = 0$, the $\varphi_j(A,\,B,\,\hbar,\,a,\,\eta,\,\cdot)$ are just the standard Harmonic oscillator eigenstates with energies $|j| + d/2$.

2. For each admissible A, B, $\hbar$, a, and η, the set $\{\,\varphi_j(A,\,B,\,\hbar,\,a,\,\eta,\,\cdot)\,\}$ is an orthonormal basis for $L^2(I\!\!R^d)$.

3. In [12], the state $\varphi_j(A,\,B,\,\hbar,\,a,\,\eta,\,X)$ is defined as a normalization factor times

$$\mathcal{H}_j(A;\,\hbar^{-1/2}\,|A|^{-1}\,(X-a))\;\varphi_0(A,\,B,\,\hbar,\,a,\,\eta,\,X).$$

Here $\mathcal{H}_j(A;\,y)$ is a recursively defined $|j|^{\rm th}$ order polynomial in y that depends on A only through U_A, where $A = |A|\,U_A$ is the polar decomposition of A.

4. By scaling out the $|A|$ and $\hbar$ dependence and using Remark 3 above, one can show that $\mathcal{H}_j(A;\,y)\,\mathrm{e}^{-y^2/2}$ is an (unnormalized) eigenstate of the usual Harmonic oscillator with energy $|j| + d/2$.

5. When the dimension d is 1, the position and momentum uncertainties of the $\varphi_j(A,\,B,\,\hbar,\,a,\,\eta,\,\cdot)$ are $\sqrt{(j+1/2)\hbar}\ |A|$ and $\sqrt{(j+1/2)\hbar}\ |B|$, respectively. In higher dimensions, they are bounded by $\sqrt{(|j|+d/2)\hbar}\ \|A\|$ and $\sqrt{(|j|+d/2)\hbar}\ \|B\|$, respectively.

6. When we approximately solve the Schrödinger equation, the choice of the sign of the square root in the definition of $\varphi_0(A,\,B,\,\hbar,\,a,\,\eta,\,\cdot)$ is determined by continuity in t after an arbitrary initial choice.

The following simple but very useful lemma is proven in [15].

Lemma 2.1 *Let $P_{|j|\le n}$ denote the projection onto the span of the $\varphi_j(A,\,B,\,\hbar,\,a,\,\eta,\,\cdot)$ with $|j| \le n$.*

$$(X-a)^m\;P_{|j|\le n} \;=\; P_{|j|\le n+|m|}\;(X-a)^m\;P_{|j|\le n}, \tag{2.2}$$

and

$$\left\|\,(X-a)^m\,P_{|j|\le n}\right\| \;\le\; \left(\sqrt{2\hbar}\;d\,\|\,A\,\|\right)^{|m|}\;\left(\frac{(n+|m|)!}{n!}\right)^{1/2}. \tag{2.3}$$

In the Born-Oppenheimer approximation, the semiclassical dynamics of the nuclei is generated by an effective potential given by a chosen isolated electronic eigenvalue $E(X)$ of the electronic hamiltonian $h(X)$, $X \in I\!\!R^d$. For a given effective potential $E(X)$ we describe the semiclassical dynamics of the nuclei by means of the time dependent basis constructed as follows:

By assumption $\mathbf{H}_1$, the potential $E\,:\,\Xi \subset I\!\!R^d \to I\!\!R$ is smooth and bounded below. Associated to $E(X)$, we have the following classical equations of motion:

$$\begin{aligned}
\dot{a}(t) &= \eta(t),\\
\dot{\eta}(t) &= -\nabla E(a(t)),\\
\dot{A}(t) &= i\,B(t),\\
\dot{B}(t) &= i\,E^{(2)}(a(t))\,A(t),\\
\dot{S}(t) &= \frac{\eta(t)^2}{2} - E(a(t)),
\end{aligned} \tag{2.4}$$

where $E^{(2)}$ denotes the Hessian matrix for E. We always assume the initial conditions $A(0)$, $B(0)$, $a(0)$, $\eta(0)$, and $S(0) = 0$ satisfy (2.1).

The matrices $A(t)$ and $B(t)$ are related to the linearization of the classical flow through the following identities:

$$\begin{aligned} A(t) &= \frac{\partial a(t)}{\partial a(0)}\, A(0) \,+\, i\, \frac{\partial a(t)}{\partial \eta(0)}\, B(0), \\ B(t) &= \frac{\partial \eta(t)}{\partial \eta(0)}\, B(0) \,-\, i \frac{\partial \eta(t)}{\partial a(0)}\, A(0). \end{aligned}$$

Because E is smooth and bounded below, there exist global solutions to the first two equations of the system (2.4) for any initial condition if $\Xi = I\!R^d$. From this, it follows immediately that the remaining three equations of the system (2.4) have global solutions. If $\Xi \neq I\!R^d$, for any initial conditions, there exists a $0 < T \leq \infty$ so that solutions to the system (2.4) exist for any time $t \in [0,\, T]$. T is finite if and only if the solution $a(t)$ corresponding to the chosen initial condition leaves the set Ξ in finite time.

Furthermore, it is not difficult [11, 12] to prove that conditions (2.1) are preserved by the flow.

The usefulness of our wave packets stems from the following important property [13]. If we decompose the potential as

$$E(X) \,=\, W_a(X) \,+\, Z_a(X) \,\equiv\, W_a(X) \,+\, (E(X) - W_a(X)),$$

where $W_a(X)$ denotes the second order Taylor approximation (with the obvious abuse of notation)

$$W_a(X) \,\equiv\, E(a) \,+\, E^{(1)}(a)\,(X-a) \,+\, E^{(2)}(a)\,(X-a)^2/2$$

then for all multi-indices j,

$$\begin{aligned} & i\hbar\, \frac{\partial}{\partial t} \left[\mathrm{e}^{iS(t)/\hbar}\ \varphi_j(A(t), B(t), \hbar, a(t), \eta(t), X) \right] \\ = & \left(-\, \frac{\hbar^2}{2}\, \Delta \,+\, W_{a(t)}(X) \right) \left[\mathrm{e}^{iS(t)/\hbar}\ \varphi_j(A(t), B(t), \hbar, a(t), \eta(t), X) \right], \end{aligned}$$

if $A(t)$, $B(t)$, $a(t)$, $\eta(t)$, and $S(t)$ satisfy (2.4). In other words, our semiclassical wave packets φ_j exactly take into account the kinetic energy and quadratic part $W_{a(t)}(X)$ of the potential when propagated by means of the classical flow and its linearization around the classical trajectory selected by the initial conditions.

In the rest of the paper, whenever we write $\varphi_j(A(t), B(t), \hbar, a(t), \eta(t), X)$, we tacitly assume that $A(t), B(t), a(t), \eta(t)$, and $S(t)$ are solutions to (2.4) with initial conditions satisfying (2.1).

3 The Born–Oppenheimer Expansion in Powers of ϵ

In this section we derive an explicit formal expansion in ϵ for the solution to the molecular Schrödinger equation by means of a multiple scales analysis. This asymptotic analysis is similar to that performed, *e.g.*, in [10]. We discuss this in detail because we need more detailed information on the structure of successive terms in the expansion.

We start with the molecular Schrödinger equation for d nuclear configuration dimensions,

$$i\,\epsilon^2\,\frac{\partial\Psi}{\partial t} \;=\; -\,\frac{\epsilon^4}{2}\,\Delta_X\,\Psi \,+\, h(X)\,\Psi. \tag{3.1}$$

We consider the isolated, multiplicity one, smooth eigenvalue $E(X)$ of $h(X)$ of hypothesis $\mathbf{H}_1$. For the moment we assume $E(X)$ is well defined on all of $I\!\!R^d$ rather than just on a subset $\Xi \subset I\!\!R^d$. Later we introduce a cut-off function to take care of the general case. We consider the solution $a(t)$, $\eta(t)$, $A(t)$, $B(t)$, and $S(t)$ to the system (2.4) of ODE's. Then, we choose the phase of the eigenfuction $\widetilde{\Phi}(X,t)$ so that

$$\langle\, \widetilde{\Phi}(X,t),\ (\,i\,\frac{\partial}{\partial t} \,+\, i\,\eta(t)\,\nabla_X\,)\,\widetilde{\Phi}(X,t)\,\rangle_{\mathcal{H}_{el}} \;=\; 0. \tag{3.2}$$

This can always be done. See, *e.g.*, [10].

The multiple scales analysis consists of separating the two length scales that are important in the nuclear variable X. The electron wave function is sensitive on an $O(1)$ scale in this variable, so X, or equivalently, $X-a(t)$ is relevant. The quantum mechanical fluctuations of nuclear wave function occur on an $O(\epsilon)$ length scale, so $(X-a(t))/\epsilon$ is also relevant. We replace the variable X by both $w = X-a(t)$ and $y = w/\epsilon$, and consider them as independent variables. This leads to the new problem of studying

$$\begin{aligned} i\,\epsilon^2\,\frac{\partial\widehat{\Psi}}{\partial t} \;=\; \Bigg[&-\,\frac{\epsilon^4}{2}\,\Delta_w \,-\, \epsilon^3\,\nabla_w\cdot\nabla_y \,-\, \frac{\epsilon^2}{2}\,\Delta_y \,+\, i\,\epsilon^2\,\eta(t)\cdot\nabla_w \,+\, i\,\epsilon\,\eta(t)\cdot\nabla_y \\ &+\;\; [\,h(a(t)+w) \,-\, E(a(t)+w)\,] \,+\, E(a(t)+\epsilon y)\Bigg]\,\widehat{\Psi}. \end{aligned} \tag{3.3}$$

We easily check that if $\widehat{\Psi}(w,y,t)$ solves (3.3) then $\widehat{\Psi}(X-a(t),(X-a(t))/\epsilon,t)$ solves (3.1).

We define $\Phi(w,t) \,=\, \widetilde{\Phi}(X,t)$. Then (3.2) becomes

$$\langle\, \Phi(w,t),\ i\,\frac{\partial}{\partial t}\,\Phi(w,t)\,\rangle_{\mathcal{H}_{el}} \;=\; 0. \tag{3.4}$$

We seek solutions to (3.3) of the form

$$\widehat{\Psi}(w,y,t) \;=\; e^{i\,S(t)/\epsilon^2}\; e^{i\,\eta(t)\cdot y/\epsilon}\,\phi(w,y,t).$$

This requires $\phi(w,y,t)$ to satisfy

$$\begin{aligned} i\,\epsilon^2\,\frac{\partial\phi}{\partial t} \;=\; \Bigg[&-\,\frac{\epsilon^4}{2}\,\Delta_w \,-\, \epsilon^3\,\nabla_w\cdot\nabla_y \,+\, \left(-\,\frac{\epsilon^2}{2}\,\Delta_y \,+\, \frac{\epsilon^2}{2}\,E^{(2)}(a(t))\,y^2\right) \\ &+\;\; [\,h(a(t)+w) \,-\, E(a(t)+w)\,] \\ &+\;\; \left(E(a(t)+\epsilon y) \,-\, E(a(t)) - \epsilon E^{(1)}(a(t))\cdot y - \epsilon^2\,E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\Bigg]\,\phi, \end{aligned} \tag{3.5}$$

where here and below we make use of the shorthand notation

$$E^{(m)}(x)\,\frac{y^m}{m!} \;=\; \sum_{\{k\,:\,|k|=m\}} \frac{(D^kE)(x)\,y^k}{k!},$$

in the usual multi-index notation. We next assume that $\phi(w,y,t)$ has an expansion of the form

$$\phi(w,y,t) \;=\; \phi_0(w,y,t) \,+\, \epsilon\,\phi_1(w,y,t) \,+\, \epsilon^2\,\phi_2(w,y,t) \,+\, \cdots$$

We further decompose each ϕ_n as

$$\phi_n(w,y,t) \;=\; g_n(w,y,t)\,\Phi(w,t) \,+\, \phi_n^{\perp}(w,y,t),$$

by projecting into the $\Phi(w,t)$ direction and into the orthogonal directions in $\mathcal{H}_{el}$.

We substitute this expansion into (3.5) and equate terms of the corresponding powers of ϵ.

Order 0. The zeroth order terms require

$$[\,h(a(t)+w) \,-\, E(a(t)+w)\,]\,\phi_0(w,y,t) \;=\; 0.$$

This forces

$$\phi_0^{\perp}(w,y,t) \;=\; 0.$$

Order 1. The first order terms require

$$[\,h(a(t)+w) \,-\, E(a(t)+w)\,]\,\phi_1(w,y,t) \;=\; 0.$$

This forces

$$\phi_1^{\perp}(w,y,t) \;=\; 0.$$

Order 2. The second order terms require

$$i\,\frac{\partial\phi_0}{\partial t} \;=\; \left(-\,\frac{1}{2}\,\Delta_y \,+\, E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\,\phi_0 \,+\, [\,h(a(t)+w) \,-\, E(a(t)+w)\,]\,\phi_2.$$

We separately examine the components of this equation in the Φ direction and in the orthogonal directions. By (3.4), this yields the two conditions

$$i\,\frac{\partial g_0}{\partial t} \;=\; \left(-\,\frac{1}{2}\,\Delta_y \,+\, E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\,g_0, \tag{3.6}$$

and

$$[\,h(a(t)+w) \,-\, E(a(t)+w)\,]\,\phi_2 \;=\; i\,g_0\,\frac{\partial\Phi}{\partial t}. \tag{3.7}$$

We arbitrarily choose g_0 to be the following w–independent particular solution of (3.6):

$$g_0(w,y,t) \;=\; \epsilon^{-d/2}\,\sum_{|j|\le J} c_{0,j}\;\varphi_j(A(t),B(t),1,0,0,y), \tag{3.8}$$

where $c_{0,j} = c_j$ is determined by the initial conditions.

We let the Hilbert space $\mathcal{H}_{\rm el}^{\perp}$ be the subspace of $\mathcal{H}_{\rm el}$ orthogonal to $\Phi(w,t)$. The restriction of $[\,h(a(t)+w) \,-\, E(a(t)+w)\,]$ to $\mathcal{H}_{\rm el}^{\perp}$ is invertible, and we denote the inverse by $r(w,t) = [\,h(a(t)+w) \,-\, E(a(t)+w)\,]_r^{-1}$. With this notation, equation (3.7) forces

$$\begin{aligned}\phi_2^{\perp}(w,y,t) &= i\,g_0(w,y,t)\;r(w,t)\;\frac{\partial\Phi}{\partial t}(w,t)\\ &= \epsilon^{-d/2}\sum_{|j|\le J} d_{2,j}(w,t)\;\varphi_j(A(t),B(t),1,0,0,y),\end{aligned} \tag{3.9}$$

where

$$d_{2,j}(w,t) \;=\; c_{0,j}\;r(w,t)\;\frac{\partial\Phi}{\partial t}(w,t) \tag{3.10}$$

is $\mathcal{H}_{\rm el}$–valued.

Order 3. The third order terms require

$$\begin{aligned} i\,\frac{\partial\phi_1}{\partial t} &= \left(-\,\frac{1}{2}\,\Delta_y \,+\, E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\,\phi_1\\ &\quad -\nabla_w\cdot\nabla_y\,\phi_0 \,+\, E^{(3)}(a(t))\frac{y^3}{3!}\,\phi_0 \,+\, [\,h(a(t)+w) \,-\, E(a(t)+w)\,]\,\phi_3.\end{aligned}$$

We separately examine the components of this equation in the Φ direction and in the orthogonal directions. By (3.4), this yields the two conditions

$$\begin{aligned} i\,\frac{\partial g_1}{\partial t} \,-\, \left(-\,\frac{1}{2}\,\Delta_y \,+\, E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\,g_1 &\\ = \,-\,(\nabla_y g_0)\cdot\langle\,\Phi,\;\nabla_w\Phi\,\rangle \,+\, E^{(3)}(a(t))\,\frac{y^3}{3!}\,g_0,&\end{aligned} \tag{3.11}$$

and

$$[\,h(a(t)+w) \,-\, E(a(t)+w)\,]\;\phi_3 \;=\; i\,g_1\,\frac{\partial\Phi}{\partial t} \,+\, (\nabla_y g_0)\,\cdot\,(P_{\perp}\,\nabla_w\Phi), \tag{3.12}$$

where $P_{\perp}(w,t)$ is the projection in $\mathcal{H}_{el}$ onto $\mathcal{H}_{\rm el}^{\perp}$.

The solution to (3.11) with $g_1(w,y,0) = 0$ can be written as

$$g_1(w,y,t) \;=\; \epsilon^{-d/2}\sum_{|j|\le J+3} c_{1,j}(w,t)\;\varphi_j(A(t),B(t),1,0,0,y),$$

for some coefficients $c_{1,j}(w,t)$.

Equation (3.12) determines

$$\begin{aligned}\phi_3^{\perp}(w,y,t) &= r(w,t)\,\left(i\,g_1(w,y,t)\,\frac{\partial\Phi}{\partial t}(w,t) + (\nabla_y g_0)(w,y,t)\cdot(P_{\perp}(w,t)\nabla_w\Phi(w,t))\right)\\ &= \epsilon^{-d/2}\sum_{|j|\le J+3} d_{3,j}(w,t)\;\varphi_j(A(t),B(t),1,0,0,y),\end{aligned}$$

where

$$d_{3,j}(w,t) \;=\; i\,\Big(r(w,t)\,\dot{\Phi}(w,t)\Big)\;c_{1,j}(w,t) \\ +\sum_{|q|\le J} r(w,t)\;(P_\perp\,\nabla_w\Phi)(w,t)\cdot\langle\varphi_j,\,\nabla_y\varphi_q\rangle\;c_{0,q}(w,t).$$

Here and below $\dot{} \equiv \dfrac{\partial}{\partial t}$.

Order n. The n^{th} order terms require

$$i\,\frac{\partial\phi_{n-2}}{\partial t} \;=\; \left(-\,\frac{1}{2}\,\Delta_y\,+\,E^{(2)}(a(t))\,\frac{y^2}{2!}\right)\,\phi_{n-2}\,-\,\frac{1}{2}\,\Delta_w\,\phi_{n-4}\,-\,\nabla_w\cdot\nabla_y\,\phi_{n-3} \\ +\;\sum_{m=3}^{n}E^{(m)}(a(t))\,\frac{y^m}{m!}\,\phi_{n-m}\,+\,[\,h(a(t)+w)\,-\,E(a(t)+w)\,]\;\phi_n.$$

The components of this equation in the $\Phi(w,t)$ direction require

$$i\,\frac{\partial g_{n-2}}{\partial t}\,-\,\left(-\,\frac{1}{2}\,\Delta_y\,+\,\frac{1}{2!}\,E^{(2)}(a(t))\,y^2\right)\,g_{n-2} \\ =\;-\,\frac{1}{2}\,\Delta_w\,g_{n-4}\,-\,\langle\,\Phi,\,\nabla_w\Phi\,\rangle\cdot(\nabla_w g_{n-4})\,-\,\frac{1}{2}\,\langle\,\Phi,\,\Delta_w\Phi\,\rangle\,g_{n-4} \\ -\,\nabla_w\cdot\nabla_y g_{n-3}\,-\,\langle\,\Phi,\,\nabla_w\Phi\,\rangle\cdot(\nabla_y g_{n-3})\,+\,\sum_{m=3}^{n}E^{(m)}(a(t))\,\frac{y^m}{m!}\,g_{n-m} \\ -\,\frac{1}{2}\,\langle\,\Phi,\,\Delta_w\phi_{n-4}^{\perp}\,\rangle\,-\,\langle\,\Phi,\,\nabla_w\cdot\nabla_y\phi_{n-3}^{\perp}\,\rangle\,+\,i\,\langle\,\frac{\partial\Phi}{\partial t},\,\phi_{n-2}^{\perp}\,\rangle. \tag{3.13}$$

Note that the last term has been transformed from $-\,i\,\langle\,\Phi,\,\dfrac{\partial\phi_{n-2}^{\perp}}{\partial t}\,\rangle$ to $i\,\langle\,\dfrac{\partial\Phi}{\partial t},\,\phi_{n-2}^{\perp}\,\rangle$. The equivalence of these expressions follows from differentiation of $\langle\,\Phi,\,\phi_{n-2}^{\perp}\,\rangle\;=\;0$ with respect to t.

The components orthogonal to $\Phi(w,t)$ require

$$[\,h(a(t)+w)\,-\,E(a(t)+w)\,]\;\phi_n \\ =\;P_\perp\,\left(i\,\frac{\partial\phi_{n-2}^{\perp}}{\partial t}\right)\,+\,\left(\frac{1}{2}\,\Delta_y\,-\,\frac{1}{2!}\,E^{(2)}(a(t))\,y^2\right)\,\phi_{n-2}^{\perp}\,-\,\sum_{m=3}^{n}E^{(m)}(a(t))\,\frac{y^m}{m!}\,\phi_{n-m}^{\perp} \\ +\;\frac{1}{2}\,P_\perp\,\Delta_w\phi_{n-4}^{\perp}\,+\,(P_\perp\,\nabla_w\Phi)\cdot(\nabla_w g_{n-4})\,+\,\frac{1}{2}\,(P_\perp\,\Delta_w\Phi)\,g_{n-4} \\ +\;P_\perp\,\nabla_w\cdot\nabla_y\phi_{n-3}^{\perp}\,+\,(P_\perp\,\nabla_w\Phi)\cdot(\nabla_y g_{n-3})\,+\,i\,\frac{\partial\Phi}{\partial t}\,g_{n-2}. \tag{3.14}$$

Equation (3.14) determines $\phi_n^{\perp}(w,y,t)$ by an application of $[h(a(t)+w)-E(a(t)+w)]_r^{-1}$. It is easily checked that the solution to (3.13) with $g_{n-2}(w,y,0)\;=\;0$ has the form

$$g_{n-2}(w,y,t)\;=\;\epsilon^{-d/2}\sum_{|j|\le J+3n-6}c_{n-2,j}(w,t)\;\varphi_j(A(t),B(t),1,0,0,y), \tag{3.15}$$

for some coefficients $c_{n-2,j}(w,t)$, and that the y dependence of the vector $\phi_n^\perp$ has the same form, with other coefficients depending on (w,t), *i.e.*,

$$\phi_n^\perp(w,y,t) \;=\; \epsilon^{-d/2} \sum_{|j|\le J+3n-6} d_{n,j}(w,t)\ \varphi_j(A(t),B(t),1,0,0,y), \tag{3.16}$$

where the $d_{n,j}(w,t)$ take their values in the electronic Hilbert space.

Equations (3.13) and (3.14) determine $c_{n-2,j}$ and $d_{n,j}$. When recursively solving these equations, we must determine $d_{n,j}$ before $c_{n,j}$ because the right hand side of (3.13) (with $n-2$ replaced by n) contains $\phi_n^\perp$.

The solution to (3.14) in terms of the $d_{n,j}$, is

$$d_{n,j}(w,t) \;=\; \sum_{i=1}^{8}\ \Delta_i(w,t), \tag{3.17}$$

where

$$\begin{aligned}
\Delta_1(w,t) &= i\,r(w,t)\,P_\perp(w,t)\ \dot{d}_{n-2,j}(w,t)\\
\Delta_2(w,t) &= -\sum_{3\le|m|\le n}\frac{(D^mE)(a(t))}{m!}\sum_{|q|\le J+3(n-|m|-2)}\langle\varphi_j,\ y^m\varphi_q\rangle\ r(w,t)\ d_{n-|m|,q}(w,t)\\
\Delta_3(w,t) &= \frac{1}{2}\,r(w,t)\,P_\perp(w,t)\ (\Delta_w d_{n-4,j})(w,t)\\
\Delta_4(w,t) &= r(w,t)\,P_\perp(w,t)\ (\nabla_w\Phi)\cdot(\nabla_w c_{n-4,j})(w,t)\\
\Delta_5(w,t) &= \frac{1}{2}\,r(w,t)\,P_\perp(w,t)\ (\Delta_w\Phi)\ c_{n-4,j}(w,t)\\
\Delta_6(w,t) &= \sum_{|q|\le J+3(n-5)} r(w,t)\,P_\perp(w,t)\ \langle\varphi_j,\ \nabla_y\varphi_q\rangle\ (\nabla_w d_{n-3,q})(w,t)\\
\Delta_7(w,t) &= \sum_{|q|\le J+3(n-3)} r(w,t)\,P_\perp(w,t)\ (\nabla_w\Phi)\ \langle\varphi_j,\ \nabla_y\varphi_q\rangle\ c_{n-3,q}(w,t)\\
\Delta_8(w,t) &= i\,r(w,t)\,P_\perp(w,t)\ \dot{\Phi}(w,t)\ c_{n-2,j}(w,t).
\end{aligned}$$

Similarly, the solution to (3.13) in terms of the $c_{n,j}$ is obtained by integration with respect to t of $i\,\dot{c}_{n,j}(w,t)$, where

$$i\,\dot{c}_{n,j}(w,t) \;=\; \sum_{i=1}^{9}\ \Gamma_i(w,t), \tag{3.18}$$

where

$$\begin{aligned}
\Gamma_1(w,t) &= -\,\frac{1}{2}\ (\Delta_w c_{n-2,j})(w,t)\\
\Gamma_2(w,t) &= -\ \langle\Phi,\ \nabla_w\Phi\rangle\cdot(\nabla_w c_{n-2,j})(w,t)\\
\Gamma_3(w,t) &= -\,\frac{1}{2}\ \langle\Phi,\ \Delta_w\Phi\rangle\ c_{n-2,j}(w,t)
\end{aligned}$$

$$\Gamma_4(w,t) \;=\; -\sum_{|q|\leq J+3(n-1)} \langle \varphi_j,\, \nabla_y \varphi_q\rangle \cdot (\nabla_w c_{n-1,q})(w,t)$$

$$\Gamma_5(w,t) \;=\; -\sum_{|q|\leq J+3(n-1)} \langle \Phi,\, \nabla_w \Phi\rangle \cdot \langle \varphi_j,\, \nabla_y \varphi_q\rangle \; c_{n-1,q}(w,t)$$

$$\Gamma_6(w,t) \;=\; \sum_{3\leq|m|\leq n+2} \; \sum_{|q|\leq J+3(n+2-m)} \frac{(D^m E)(a(t))}{m!} \, \langle \varphi_j,\, y^m \varphi_q\rangle \; c_{n+2-m,q}(w,t)$$

$$\Gamma_7(w,t) \;=\; \frac{1}{2}\, \langle \Phi,\, (\Delta_w d_{n-2,j})(w,t)\rangle$$

$$\Gamma_8(w,t) \;=\; -\sum_{|q|\leq J+3(n-3)} \langle \varphi_j,\, \nabla_y \varphi_q\rangle \cdot \langle \Phi,\, (\nabla_w d_{n-2,q})(w,t)\rangle$$

$$\Gamma_9(w,t) \;=\; i\, \langle \dot{\Phi},\, d_{n,j}(w,t)\rangle.$$

4 The Main Result

We introduce a C^∞ real valued cut-off function $F : I\!\!R^d \to I\!\!R$ that equals 1 in a neighborhood of the origin and equals zero away from the origin. More precisely, we choose $0 < b_0 < b_1 < \infty$, such that

$$\text{supp } (\partial_{w_i} F)(w) \;\subseteq\; \{w \in I\!\!R^d \;:\; b_0 < |w| < b_1\},$$

for any $i \in \{1, \dots, d\}$, and such that for any $t \in \Omega$, all quantities appearing in the above expansion are well defined for $w \in I\!\!R^d$ with $|w| < b_1$. Here Ω is a particular simply connected open complex neighborhood of the real interval $[0,\, T]$ that we construct in Section 5 under hypotheses $\mathbf{H}_0$ and $\mathbf{H}_1$.

We define our approximate solution to (3.1) at order N by the following expression:

$$\hat{\Psi}_N(w,y,t) \tag{4.1}$$
$$= \; F(w)\, \mathrm{e}^{iS(t)/\epsilon^2}\, \mathrm{e}^{i\eta(t)\cdot y/\epsilon} \left(\sum_{n=0}^{N} \epsilon^n \, g_n(w,y,t)\, \Phi(w,t) \; + \; \sum_{n=2}^{N+2} \epsilon^n \, \phi_n^\perp(w,y,t) \right).$$

We prove in Section 7.2 that this quantity agrees with an exact solution up to an error whose norm is bounded by ϵ^N for $t \in [0,\, T]$.

We emphasize that once the molecular hamiltonian $h(X)$ and its spectral data $E(X)$, $\Phi(X)$ are given, the only arbitrary input of the above derived expansion consists of the set of coefficients $c_{0,j}$, $|j| \leq J$. We note that at time $t = 0$, we have $c_{n,j}(0,w) \equiv 0$ for all $n \geq 1$. Thus, at $t = 0$, the approximation reduces to

$$\hat{\Psi}_N(w,y,0)$$
$$= \; F(w)\, \mathrm{e}^{iS(0)/\epsilon^2}\, \mathrm{e}^{i\eta(0)\cdot y/\epsilon} \left(g_0(0,y,0)\, \Phi(w,0) \; + \; \sum_{n=2}^{N+2} \epsilon^n \, \phi_n^\perp(w,y,0) \right).$$

This expression is completely determined by $g_0(0,y,0)$, the nuclear part of the wave function parallel to the chosen electronic level at time 0.

As is usual in the study of adiabatic problems, in order to get accurate information on the evolution of an initial wave function associated with a specific electronic level, one needs to include a higher order component perpendicular to that electronic level. This higher order part is completely determined by the parallel part. Here it is given (up to phase and cut-off functions) at time 0 by $\sum_{n=2}^{N+2} \epsilon^n \, \phi_n^{\perp}(w,y,0)$.

We now state our main Theorem:

Theorem 4.1 *Assume hypotheses* $\mathbf{H}_0$ *and* $\mathbf{H}_1$ *and consider the above construction. For all sufficiently small choices of* $g>0$*, there exist* $C(g)>0$ *and* $\Gamma(g)>0$ *such that, for* $N(\epsilon)=[\![g^2/\epsilon^2]\!]$*, the vector* $\Psi_*(X,t,\epsilon) = \hat{\Psi}_{N(\epsilon)}(X-a(t),(X-a(t))/\epsilon,t)$ *satisfies*

$$\left\| \, e^{-itH(\epsilon)/\epsilon^2}\,\Psi_*(X,0,\epsilon) \;-\; \Psi_*(X,t,\epsilon)\,\right\|_{L^2(\mathbb{R}^d,\mathcal{H}_{el})} \;\le\; C(g)\; e^{-\Gamma(g)/\epsilon^2},$$

for all $t\in[0,\,T]$*, as* $\epsilon\to 0$*.*
Moreover, we have the following exponential localization result. For any $b>0$ *and a sufficiently small choice of* $g>0$ *(that depends on* b*), there exist* $c(g)$ *and* $\gamma(g)>0$*, such that*

$$\left(\int_{|x-a(t)|>b} \|\Psi_*(X,t,\epsilon)\|^2_{\mathcal{H}_{el}}\, dx\right)^{1/2} \;\le\; c(g)\; e^{-\gamma(g)/\epsilon^2},$$

for all $t\in[0,\,T]$*, as* $\epsilon\to 0$*.*

The strategy of the proof is as follows: We consider the approximation $\hat{\Psi}_N(X-a(t),(X-a(t))/\epsilon,t)$ and the exact solution to the Schrödinger equation with the same initial conditions. We estimate the norm of the error (that is the difference between these two quantities) as a function of both N and ϵ. Apart from some subtleties, the norm of the error is bounded by $C\epsilon^N(\tau N^{1/2})^N$, for some constants C and $\tau>0$. We minimize the error estimate over all choices of N. This yields $N\simeq g^2/\epsilon^2$, for sufficiently small $g>0$, and an estimate of order $\mathrm{e}^{-\Gamma(g)/\epsilon^2}$ for the norm of the error.

We prove two extensions of this result in Section 8. In the first extension, we consider the validity of our approximation on the Ehrenfest time scale, *i.e.*, when $T=T(\epsilon)\simeq\ln(1/\epsilon)$. In the second extension, we study the dependence of our construction on J, in order to extend our main result to a wider class of initial conditions. We refer the reader to Section 8 for the precise statements.

5 Analyticity Properties

Our estimates depend on analyticity in $t\in\Omega$ of the vectors $c_n(w,t)\in l^2(\mathbf{N}^d,\mathbb{C})$ and $d_n(w,t)\in l^2(\mathbf{N}^d,\mathcal{H}_{\rm el})$, where Ω is the particular simply connected open complex neighborhood of the real interval $[0,\,T]$ mentioned at the beginning of Section 4.

To construct Ω, we begin with several observations. Our hypotheses imply that the eigenvalue $E(X)$ is analytic in $\Sigma_{\delta'}$, so the solutions $a(t)$, $\eta(t)$, $A(t)$, $B(t)$, and $S(t)$ are well defined for all $t \in [0, T]$. Moreover, by standard arguments [6], these functions all have analytic continuations from $[0, T]$ to a simply connected open set Ω_1 that contains $[0, T]$. We assume without loss of generality that $\Omega_1 = \overline{\Omega_1}$, where $\overline{\Omega_1}$ denotes the conjugate of Ω_1.

We note that $A^*(t)$ and $B^*(t)$ also have analytic continuations from $[0, T]$ to Ω_1. To see this for $A^*(t)$, note that for $t \in [0, T]$, $A^*(t) = A^*(\overline{t})$, and $A^*(\overline{t})$ has an analytic continutation to $\overline{\Omega_1}$. The argument for $B^*(t)$ is similar.

It now follows easily from the definitions that for each X, $\varphi_j(A(t), B(t), \epsilon^2, a(t), \eta(t), X)$ and $\overline{\varphi_j(A(t), B(t), \epsilon^2, a(t), \eta(t), X)}$ have analytic continuations from $[0, T]$ to some simply connected open set Ω_2. For $t \in [0, T]$, the real part of $B(t)A(t)^{-1}$ is strictly positive. This positivity will remain true for the real part of the analytic continuation of $B(t)A(t)^{-1}$ on some simply connected subset $\Omega \subset \Omega_1 \cap \Omega_2$ that contains $[0, T]$. We assume without loss of generality that $\Omega = \overline{\Omega}$ and we can assume that Ω has the form $\{ t \,:\, -a < \mathrm{Re}\, t < b \text{ and } |\mathrm{Im}\, t| < c \}$ where $a > c > 0$ and $b > T + c$. It follows that for $t \in \Omega$, both $\varphi_j(A(t), B(t), \epsilon^2, a(t), \eta(t), x)$ and $\overline{\varphi_j(A(t), B(t), \epsilon^2, a(t), \eta(t), x)}$ have analytic continuations from $[0, T]$ to Ω as elements of $L^2(I\!\!R^d)$.

Using these results and carefully examining the constructions of the vectors $c_n(w, t)$ and $d_n(w, t)$, we see that they are analytic in t for $t \in \Omega$, also.

Our hypotheses on $h(\cdot)$ and the above results also show that each of the following quantities is analytic in t for $t \in \Omega$ and each fixed $w \in \Sigma_\delta \subset \mathbb{C}^d$, for sufficiently small δ:

$$\begin{aligned}
&r(w,t) \;=\; [\, h(a(t)+w) \;-\; E(a(t)+w)\,]_r^{-1}, \\
&\Phi(w,t), \\
&(D_w^\alpha \Phi)(w,t), \quad \text{for } |\alpha| \le 2, \\
&D_w^\alpha E(a(t)), \quad \text{for all} \quad \alpha \\
&P_\perp(w,t).
\end{aligned}$$

By explicit computation of the phase corresponding to (3.2) it is easy to check that $\Phi(w, t)$ and its derivatives are also analytic for $t \in \Omega$.

Moreover, if $f_i(w, t)$, (i in some finite set) represents any of these quantities, f_i is analytic in $w \in \Sigma_\delta$, for any fixed $t \in \Omega$. Thus, by the Cauchy integral formula, we can assume that the following bounds hold (with the appropriate norm in each case):

$$\| \, (D_w^\alpha f_i)(w,t) \, \| \;\le\; c_i \; G_i^{|\alpha|} \; \frac{\alpha!}{(1+|\alpha|)^{d+1}}, \tag{5.1}$$

for some c_i, G_i, $w \in \Sigma_\delta$, and α ranges over all multi-indices.

We can assume here that all $G_i \le D_2$ for some constant

$$D_2 \;\ge\; 1,$$

and we associate the prefactors c_i in (5.1) with the different functions according to the rules

$$\begin{array}{ccccccc}
c_1 & \leftrightarrow & rP_\perp & \qquad & c_2 & \leftrightarrow & \Phi \\
c_3 & \leftrightarrow & \dot{\Phi} & & c_4 & \leftrightarrow & \nabla_w \Phi \\
c_5 & \leftrightarrow & \Delta_w \Phi & & c_6 & \leftrightarrow & E \\
c_4' & \leftrightarrow & \langle \Phi,\, \nabla_w \Phi \rangle & & c_5' & \leftrightarrow & \langle \Phi,\, \Delta_w \Phi \rangle.
\end{array}$$

6 Structure and Estimates of the $c_n(w,t)$ and $d_n(w,t)$

In this section, we decompose the functions g_n and $\phi_n^\perp$ of Section 3 into pieces, each of which satisfies various estimates.

Throughout this section, all w–dependent quantities are defined for w in the support of the cut-off function F. Furthermore, all the results of this section are claimed to hold only on the support of F.

Our decompositions of $g_n(w,y,t)$ and $\phi_n^\perp(w,y,t)$ have the following forms:

$$\begin{aligned} & g_n(w,y,t) \\ = \; & \epsilon^{-d/2} \sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\le n} \sum_{|l|+k\le p+\frac{n}{2}} \sum_{|j|\le J+n+2(p-|l|-k)} c_{n,p,l,k,\beta,j}(w,t)\; \varphi_j(A(t),B(t),1,0,0,y). \end{aligned} \tag{6.1}$$

and

$$\begin{aligned} & \phi_n^\perp(w,y,t) \;= \\ & \epsilon^{-d/2} \sum_{\beta\in\mathcal{B}_{n,2}} \sum_{p\le n-1} \sum_{|l|+k\le p+\frac{n-1}{2}} \sum_{|j|\le J+(n-1)+2(p-|l|-k)} d_{n,p,l,k,\beta,j}(w,t)\; \varphi_j(A(t),B(t),1,0,0,y). \end{aligned} \tag{6.2}$$

In (6.1), n, k and p are non-negative integers; j and l are multi-indices; and the index β runs over a finite set $\mathcal{B}_{n,1}$. The number J is fixed by the initial conditions. Each $c_{n,p,l,k,\beta,j}$ is a complex valued function.
In (6.2), $n\ge 2$, k and p are non-negative integers; j and l are multi-indices; and the index β runs over a finite set $\mathcal{B}_{n,2}$. Each $d_{n,p,l,k,\beta,j}(w,t)$ takes values in $\mathcal{H}_{el}$.

We let $c_{n,p,l,k,\beta}(w,t)$ and $d_{n,p,l,k,\beta}(w,t)$ respectively denote vectors in $l^2(\mathbf{N}^d,\mathbb{C})$ and $l^2(\mathbf{N}^d,\mathcal{H}_{el})$ whose components are $c_{n,p,l,k,\beta,j}(w,t)$ and $d_{n,p,l,k,\beta,j}(w,t)$.

The crucial step in the proof of Theorem 4.1 is the following:

Proposition 6.1 *There is a recursive construction of the coefficients $c_{n,p,l,k,\beta,j}(w,t)$ and $d_{n,p,l,k,\beta,j}(w,t)$ for w on the support of F.*
The indices for $c_{n,p,l,k,\beta,j}(w,t)$ are non-negative and satisfy

$$\begin{aligned} \beta &\in \mathcal{B}_{n,1} \\ p &\le n, \\ |l|+k &\le p+\frac{n}{2}, \\ |j| &\le J+n+2(p-|l|-k). \end{aligned}$$

The indices for $d_{n,p,l,k,\beta,j}(w,t)$ are non-negative and satisfy

$$\begin{aligned} n &\ge 2 \\ \beta &\in \mathcal{B}_{n,2}, \\ p &\le n-1 \\ |l|+k &\le p+\frac{n-1}{2} \\ |j| &\le J+(n-1)+2(p-|l|-k). \end{aligned}$$

Moreover, the following conditions are satisfied:
i) *For any* $n > 0$, $c_{n,0,l,k,\beta,j}(w,t) = 0$.
ii) *There exists* $K_0 > 0$*, such that the number of terms in both of the sums (6.1) and (6.2) is bounded by* $e^{K_0 n}$.
iii) *For* $t \in \Omega$*, let* $\mathrm{dist}(t)$ *be the distance from* t *to the complement of* Ω*. The coefficients* $c_{n,p,l,k,\beta}(w,t)$ *and* $d_{n,p,l,k,\beta}(w,t)$ *are analytic for* $t \in \Omega$*, and there exist constants* D_1 *and* D_2*, such that*

$$\begin{aligned} &\|(D_w^\alpha c_{n,p,l,k,\beta})(w,t)\| \\ &\leq\ D_1\ D_2^{|\alpha|+|l|+4n}\ \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\ \frac{|t|^p}{p!}\ \frac{k^k}{\mathrm{dist}(t)^k}\ \left[\frac{(J+n+2(p-|l|-k))!}{J!}\right]^{1/2}, \end{aligned} \tag{6.3}$$

and

$$\begin{aligned} &\|(D_w^\alpha d_{n,p,l,k,\beta})(w,t)\| \\ &\leq\ D_1 D_2^{|\alpha|+|l|+4(n-1)}\ \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\ \frac{|t|^p}{p!}\ \frac{k^k}{\mathrm{dist}(t)^k}\ \left[\frac{(J+(n-1)+2(p-|l|-k))!}{J!}\right]^{1/2}. \end{aligned} \tag{6.4}$$

Remark The complicated estimates (6.3) and (6.4) are motivated by estimates used in semiclassical approximations and adiabatic approximations. The factors on the right hand sides that explicitly involve J, n, and p occur in the semiclassical paper [15]. The factors that involve α and l appear in the adiabatic paper [25]. The factors that involve k occur in a proof of the adiabatic results of [25] that are based on Cauchy estimates instead of Nenciu's lemma [25] (that we generalize below as Lemma 6.4). We were unable to prove Proposition 6.1 without using a combination of all of these techniques. We estimate adiabatic error terms by using Nenciu's approach in the w variable and Cauchy estimates in the t variable.

6.1 The Toolbox

To prove Proposition 6.1, we repeatedly use the following very handy lemmas, whose proofs are given in Section 9. The first two lemmas deal with basic properties of analytic functions of one variable and are consequences of the Cauchy integral formula.

Lemma 6.1 *For* $k = 0$*, define* $k^k = 1$*. Suppose* g *is an analytic vector–valued function on the strip* $S_\delta = \{\, t : |\mathrm{Im}\, t| < \delta \,\}$*. If* g *satisfies*

$$\|g(t)\| \ \leq\ C\, k^k\, (\delta\ -\ |\mathrm{Im}\, t|)^{-k},$$

for some $k \geq 0$*, then* g' *satisfies*

$$\|g'(t)\| \ \leq\ C\, (k+1)^{k+1}\, (\delta\ -\ |\mathrm{Im}\, t|)^{-k-1},$$

for all $t \in S_\delta$.

Lemma 6.1 has a generalization to regions other than infinite strips. The generalization is needed if one wishes to study problems where analyticity holds only in a neighborhood of a finite time interval. The proof of the generalized lemma is similar to that of Lemma 6.1, but involves slightly more complicated geometry. The precise statement is the following:

Lemma 6.2 *For $k = 0$, define $k^k = 1$. Suppose g is an analytic vector–valued function in an open region $\Omega \subset \mathbb{C}$. For $t \in \Omega$, let* $\operatorname{dist}(t)$ *be the distance from t to Ω^C, the complement of Ω. If g satisfies*

$$\|g(t)\| \;\leq\; C\, k^k \,(\operatorname{dist}(t))^{-k},$$

for all $t \in \Omega$ and some $k \geq 0$, then g' satisfies

$$\|g'(t)\| \;\leq\; C\,(k+1)^{k+1}\,(\operatorname{dist}(t))^{-k-1},$$

for all $t \in \Omega$.

The next lemma gives estimates on indefinite integrals of certain analytic functions under stronger assumptions on the domain Ω.

Lemma 6.3 *Suppose f is an analytic vector–valued function in an open region $\Omega \subset \mathbb{C}$. For $t \in \Omega$, let* $\operatorname{dist}(t)$ *be the distance from t to Ω^C. We assume the domain is star-shaped with respect to the origin and that the origin is the most distant point to Ω^C, i.e.,* $\operatorname{dist}(0) \geq \operatorname{dist}(t)$, *for all $t \in \Omega$. Moreover, we assume that* $\operatorname{dist}(t)$ *is monotone decreasing along any line emanating from the origin. If f satisfies*

$$\|f(t)\| \;\leq\; C\ |t|^p\ (\operatorname{dist}(t))^{-k},$$

for all $t \in \Omega$ and some $k \geq 0$, then $\left\| \int_0^t f(s)\,ds \right\|$ satisfies

$$\left\| \int_0^t f(s)\,ds \right\| \;\leq\; C\ \frac{|t|^{p+1}}{p+1}\ (\operatorname{dist}(t))^{-k},$$

for all $t \in \Omega$.

Remark: In our situation, examples of sets Ω we can use that satisfy the conditions of Lemma 6.3 are infinite symmetrical horizontal strips or the rectangular regions chosen in Section 5.

A fourth tool we repeatedly use below is a multidimensional generalization of a lemma used in [25]. We warn the reader that the symbol for a norm means different things in different contexts, *e.g.*, for scalar–valued, operator–valued, and vector–valued functions, it respectively means absolute value, operator norm, and vector space norm.

Lemma 6.4 *The quantity*

$$\nu \;=\; \sup_{\alpha}\ \ (1+|\alpha|)^{d+1} \sum_{\{\,l\,:\,0\,\leq\, l_i\,\leq\,\alpha_i\,\}} \frac{1}{(1+|l|)^{d+1}}\ \frac{1}{(1+|\alpha-l|)^{d+1}}. \tag{6.5}$$

is finite.
Let Σ be an open subset of $\mathbb{C}^d$. Suppose $M(\cdot) \in C^\infty(\Sigma)$ is scalar–valued or operator–valued, and $N(\cdot) \in C^\infty(\Sigma)$ is either operator–valued or vector–valued. Assume these functions satisfy

$$\| \, (D^\alpha \, M) \, (x) \, \| \quad \leq \quad m(x) \;\, a(x)^{|\alpha+p|} \quad \frac{(\alpha+p)!}{(1+|\alpha|)^{d+1}} \tag{6.6}$$

$$\| \, (D^\alpha \, N) \, (x) \, \| \quad \leq \quad n(x) \;\, a(x)^{|\alpha+q|} \quad \frac{(\alpha+q)!}{(1+|\alpha|)^{d+1}} \tag{6.7}$$

for $x \in \Sigma$, all multi-indices α, and some fixed multi-indices p and q. Then

$$\| \, (\, D^\alpha \, (M \, N) \,) \, (x) \, \| \;\leq\; m(x) \;\, n(x) \;\, \nu \;\, a(x)^{|\alpha+p+q|} \quad \frac{(\alpha+p+q)!}{(1+|\alpha|)^{d+1}} \tag{6.8}$$

for each multi-index α, where ν is defined by (6.5).

6.2 Proof of Proposition 6.1

We prove Proposition 6.1 by induction and begin with the case $n = 0$. We construct $c_{0,0,0,0,\beta,j} \equiv c_{0,j}$ with $\beta = 1 \in \mathcal{B}_{0,1} \equiv \{1\}$. We note that there is no $d_{n,p,l,k,\beta}(w,t)$ for $n \leq 1$; the inequalities for its indices in the conclusion to the proposition cannot be satisfied by non-negative integers. Whenever $d_{n,p,l,k,\beta}(w,t)$ with $n \leq 1$ appears in any of the formal calculations below, it is understood to be zero.

We now assume that the estimates (6.3) and (6.4) on $c_{m,p,l,k,\beta}(w,t)$ and $d_{m,p,l,k,\beta}(w,t)$ are true for all $m \leq n-1$ and prove they still hold for $m = n$.

Our strategy is to show that each contribution Δ_i and Γ_i consists of a finite sum of terms that satisfy the required estimate. We estimate the number of terms by a separate argument. Our main tools are Lemmas 2.1, 6.2, 6.3, and 6.4.

The index β must be considered when counting the number of terms, but it plays no role in the estimates of the individual terms. To simplify the notation, we drop it while estimating the terms.

The Term Δ_1

We begin by considering the contribution to (6.4) from the term Δ_1 in (3.17).

By induction, each $d_{n-2,p,l,k,\beta}(w,t)$ is analytic for $t \in \Omega$ and has a p^{th} order zero at $t = 0$. It follows that $d_{n-2,p,l,k,\beta}(w,t) = t^p \, f(t)$, where f is analytic in Ω. When we take the time derivative, we obtain two terms, $p \, t^{p-1} \, f(t)$ and $t^p \, \dot{f}(t)$. These, respectively, give rise to two terms $d_{n,p-1,l,k,\beta'}(w,t)$ and $d_{n,p,l,k+1,\beta''}(w,t)$.

We consider all w-derivatives of $\Delta_1(w,t)$. We apply the induction hypothesis, Lemma 6.2, and Lemma 6.4 to obtain

$$\begin{aligned} & \left\| \, D_w^\alpha \left(r P_\perp(w,t) \dot{d}_{n-2,p,l,k}(w,t) \right) \right\| \\ & \leq \; c_1 \, \nu \, D_1 \, D_2^{|\alpha|+|l|+4(n-3)} \; \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \; \sqrt{\frac{(J+(n-3)+2(p-|l|-k))!}{J!}} \end{aligned}$$

$$\times \quad \left(\frac{|t|^{p-1}}{(p-1)!} \, \frac{k^k}{\mathrm{dist}(t)^k} \; + \; \frac{t^p}{p!} \, \frac{(k+1)^{k+1}}{\mathrm{dist}(t)^{k+1}} \right)$$

$$= \; c_1 \nu D_1 D_2^{-8} D_2^{|\alpha|+|l|+4(n-1)} \, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^{p'}}{p'!} \, \frac{k^k}{\mathrm{dist}(t)^k} \sqrt{\frac{(J+(n-1)+2(p'-|l|-k))!}{J!}}$$

$$+ \, c_1 \nu D_1 D_2^{-8} D_2^{|\alpha|+|l|+4(n-1)} \, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^{p}}{p!} \, \frac{k'^{k'}}{\mathrm{dist}(t)^{k'}} \sqrt{\frac{(J+(n-1)+2(p-|l|-k'))!}{J!}},$$

with $p' = p-1$ and $k' = k+1$. We check that

$$\begin{array}{rcccl} p & \leq & n-3 & < & n-1, \\ p' & \leq & n-4 & < & n-1, \\ |l|+k & \leq & p+(n-3)/2 & = & p'+(n-1)/2, \\ |l|+k' & \leq & p+(n-3)/2+1 & = & p+(n-1)/2, \end{array}$$

and the ranges of the components of each vector satisfy

$$\begin{array}{rcl} |j| & \leq & J+(n-1)+2(p'-|l|-k), \\ |j| & \leq & J+(n-1)+2(p-|l|-k'), \end{array}$$

as required. Hence, we get the desired bound for each of the two contributions, provided

$$D_2^8 \; \geq \; c_1 \, \nu.$$

The Term Δ_2

In the analysis of this term, we encounter an infinite matrix that represents multiplication by y^m in the basis of semiclassical wave packets. We denote this matrix by $\langle \varphi, \, y^m \varphi \rangle$. Its entries are $\langle \varphi_j, \, y^m \varphi_q \rangle(t)$, for multi-indices $m, j, q \in \mathbf{N}^d$. We recall that Lemma 2.1 gives bounds for these matrix elements and also states that $\langle \varphi_j, \, y^m \varphi_q \rangle(t) = 0$ if $|\,|j| - |q|\,| > |m|$.

We adopt the analogous notation for the infinite matrix $\langle \varphi, \, D_y^m \varphi \rangle$ that represents the operator D_y^m in the basis of semiclassical wave packets.

We define $d_0 = \sqrt{2}\, d$. Then, using (5.1), Lemmas 2.1, 6.3, 6.2, and 6.4, and some algebra, we obtain

$$\left\| D_w^\alpha \sum_{\tilde{m}=3}^{n} \sum_{|m|=\tilde{m}} \frac{D^m E(a(t))}{m!} \, \langle \varphi, \, y^m \varphi \rangle \, (r P_\perp)(w,t) \, d_{n-\tilde{m},p,l,k}(w,t) \right\|$$

$$\leq \; \sum_{\tilde{m}=3}^{n} \sum_{|m|=\tilde{m}} (d_0 \|A\|)^{\tilde{m}} \, \frac{c_6 \, c_1 \, \nu \, D_2^{\tilde{m}} \, m!}{(1+\tilde{m})^{d+1} \, m!} \sqrt{\frac{(J+(n-1)+2(p-|l|-k))!}{J!}}$$

$$\times \quad D_1 \, D_2^{|\alpha|+|l|+4(n-1-\tilde{m})} \, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^p}{p!} \, \frac{k^k}{\mathrm{dist}(t)^k}$$

$$\leq \sum_{\tilde{m}=3}^{n} \sum_{|m|=\tilde{m}} D_1\, c_6\, c_1 \nu \, \frac{(d_0\|A\|)^{\tilde{m}}}{D_2^{3\tilde{m}}} \, D_2^{|\alpha|+|l|+4(n-1)} \, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^p}{p!} \, \frac{k^k}{\mathrm{dist}(t)^k}$$
$$\times \sqrt{\frac{(J+n-1+2(p-|l|-k))!}{J!}}.$$

We also verify the constraints on the parameters and components of the vectors:

$$\begin{aligned} p &\leq n-1-\tilde{m} &&\leq n-1, \\ |l|+k &\leq p+(n-1-\tilde{m})/2 &&\leq p+(n-1)/2, \\ |j| &\leq J+(n-\tilde{m}-1)+2(p-|l|-k)+\tilde{m} &&\leq J+(n-1)+2(p-|l|-k). \end{aligned}$$

Hence, we see that each contribution from $\Delta_2(w,t)$ satisfies the required bound, provided the following two conditions are fulfilled

$$\begin{aligned} D_2^3 &\geq d_0\,\|A\|, \\ D_2^9 &\geq (d_0\|A\|)^3\, c_6\, c_1\, \nu. \end{aligned}$$

There are

$$\sum_{\tilde{m}=3}^{n} \sum_{|m|=\tilde{m}} 1 \;\leq\; \sum_{|m|\leq n} 1 \;=\; \binom{n+d}{d} \;\leq\; \sigma_0\, \mathrm{e}^{\sigma n}$$

such contributions, where $\sigma > 0$ can be chosen arbitrarily small (see [14]).

The Term Δ_3

For this term we make the laplacian explicit and write

$$\Delta_w d_{n-4,p,l,k}(w,t) \;=\; \sum_{i=1}^{d} (D_{w_i}^2 d_{n-4,p,l,k})(w,t).$$

We introduce $l_{i,2} \;=\; l+(0,0,\ldots 0,2,0,\ldots,0)$, where the 2 sits in the ith column.
We then estimate

$$\left\| D_w^\alpha \, \frac{1}{2} \, \left(rP_\perp(w,t)\Delta_w d_{n-4,p,l,k}(w,t)\,\right) \right\|$$
$$\leq \frac{1}{2} \sum_{i=1}^{d} c_1\, \nu\, D_1\, D_2^{|\alpha|+|l|+2+4(n-5)} \, \frac{(\alpha+l_{i,2})!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^p}{p!} \, \frac{k^k}{\mathrm{dist}(t)^k}$$
$$\times \sqrt{\frac{(J+(n-5)+2(p-|l|-k))!}{J!}}$$
$$= \sum_{i=1}^{d} \frac{c_1\, \nu\, D_1}{2\, D_2^{16}} \, D_2^{|\alpha|+|l_{i,2}|+4(n-1)} \, \frac{(\alpha+l_{i,2})!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^p}{p!} \, \frac{k^k}{\mathrm{dist}(t)^k}$$
$$\times \sqrt{\frac{(J+(n-1)+2(p-|l_{i,2}|-k))!}{J!}}.$$

Again, the constraints are satisfied since

$$\begin{array}{rcccl} p & \leq & n-5 & < & n-1 \\ |l_{i,2}|+k & \leq & p+(n-5)/2+2 & = & p+(n-1)/2 \\ |j| & \leq & J+(n-5)+2(p-|l|-k) & = & J+(n-1)+2(p-|l_{i,2}|-k) \end{array}$$

and each of the d contributions stemming from $\Delta_3(w,t)$ satisfies the required estimate, provided

$$D_2^{16} \;\geq\; c_1\nu/2.$$

We estimate each of the remaining terms $\Delta_i(w,t)$, $i=4,\ldots,8$, in the same fashion, using the same tools. Since this is straightforward, we only outline the arguments.

The Term Δ_4

We expand the dot product

$$(\nabla_w\Phi)\cdot(\nabla_w c_{n-4,p,l,k}) \;=\; \sum_{i=1}^{d}(D_{w_i}\Phi)\,(D_{w_i}c_{n-4,p,l,k})$$

and use the definition

$$l_{i,1}=l+(0,0,\ldots 0,1,0,\ldots,0),$$

where the 1 sits at the ith column. Recall that the estimates on the $c_{m,p,l,k}$'s differ from those on the $d_{m,p,l,k}$'s by a shift of 1 in the m dependence. We have

$$\begin{aligned} &\|\,D_w^\alpha\,(rP_\perp(w,t)\,\nabla_w\Phi(w,t)\cdot\nabla_w c_{n-4,p,l,k}(w,t))\,\| \\ \leq\;& \sum_{i=1}^{d}\frac{c_1\,c_4\,\nu^2\,D_1}{D_2^{12}}\;D_2^{|\alpha|+|l_{i,1}|+4(n-1)}\;\frac{(\alpha+l_{i,1})!}{(1+|\alpha|)^{d+1}}\;\frac{|t|^p}{p!}\;\frac{k^k}{\mathrm{dist}(t)^k} \\ &\qquad\qquad\times\;\sqrt{\frac{(J+(n-1)+2(p-|l_{i,1}|-k))!}{J!}}, \end{aligned}$$

with all constraints on $|j|, p, |l_{i,1}|, k$ satisfied. Thus each of the d contributions stemming from $\Delta_4(w,t)$ satisfies the required estimate, provided

$$D_2^{12} \;\geq\; c_1\,c_4\,\nu^2.$$

The Term Δ_5

This term is similar to the previous one. We obtain

$$\begin{aligned} &\left\|\,D_w^\alpha\left(\frac{1}{2}\,rP_\perp(w,t)\,(\Delta_w\Phi)(w,t)\,c_{n-4,p,l,k}(w,t)\right)\right\| \\ \leq\;& \frac{c_1\,c_5\,\nu^2\,D_1}{2D_2^{12}}\;D_2^{|\alpha|+|l|+4(n-1)}\;\frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\;\frac{|t|^p}{p!}\;\frac{k^k}{\mathrm{dist}(t)^k} \\ &\qquad\qquad\times\;\sqrt{\frac{(J+(n-1)+2(p-|l|-k))!}{J!}}, \end{aligned}$$

with all constraints on $|j|, p, |l|, k$ satisfied. Thus the contribution stemming from $\Delta_5(w,t)$ satisfies the required estimate, provided

$$D_2^{12} \;\geq\; c_1\, c_5\, \nu^2/2.$$

The Term Δ_6

At this point the matrices $\langle\varphi,\, D_{y_i}\varphi\rangle$ play a role that we control by the momentum space analog of Lemma 2.1. Expanding the dot product and introducing the matrices $\langle\varphi,\, D_{y_i}\varphi\rangle$, $i = 1, \ldots, d$ we have the following estimate for this term:

$$\begin{aligned}
&\left\| D_w^\alpha \sum_{i=1}^d (rP_\perp)(w,t)\; \langle\varphi,\, D_{y_i}\varphi\rangle\; D_{w_i} d_{n-3,p,l,k}(w,t) \right\| \\
&\leq\; \sum_{i=1}^d \frac{d_0\, c_1\, \nu\, \|B\|\, D_1}{D_2^{12}}\; D_2^{|\alpha|+|l_{i,1}|+4(n-1)}\; \frac{(\alpha+l_{i,1})!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^p}{p!}\; \frac{k^k}{\operatorname{dist}(t)^k} \\
&\qquad\qquad \times\; \sqrt{\frac{(J+(n-1)+2(p-|l_{i,1}|-k))!}{J!}}
\end{aligned}$$

with all constraints on $|j|, p, |l_{i,1}|, k$ satisfied. Thus, each of the d contributions stemming from $\Delta_6(w,t)$ satisfies the required estimate, provided

$$D_2^{12} \;\geq\; d_0\, c_1\, \nu \|B\|.$$

The Term Δ_7

Similarly,

$$\begin{aligned}
&\left\| D_w^\alpha \sum_{i=1}^d (rP_\perp)(w,t)\; \langle\varphi,\, D_{y_i}\varphi\rangle\; (D_{w_i}\Phi)(w,t)\; c_{n-3,p,l,k}(w,t) \right\| \\
&\leq\; \sum_{i=1}^d \frac{d_0\, c_1\, c_4\, \nu^2\, \|B\|\, D_1}{D_2^{8}}\; D_2^{|\alpha|+|l|+4(n-1)}\; \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^p}{p!}\; \frac{k^k}{\operatorname{dist}(t)^k} \\
&\qquad\qquad \times\; \sqrt{\frac{(J+(n-1)+2(p-|l|-k))!}{J!}},
\end{aligned}$$

with all constraints on $|j|, p, |l|, k$ satisfied. Thus, each of the d contributions stemming from $\Delta_7(w,t)$ satisfies the required estimate, provided

$$D_2^{12} \;\geq\; d_0\, c_1\, c_4\, \nu^2\, \|B\|.$$

The Term Δ_8

Finally,

$$\left\| \, D_w^\alpha \, \Big(r P_\perp(w,t) \, \dot{\Phi}(w,t) \, c_{n-2,p,l,k}(w,t) \Big) \, \right\|$$

$$\leq \; \frac{c_1 \, c_3 \, \nu^2 \, D_1}{D_2^4} \, D_2^{|\alpha|+|l|+4(n-1)} \, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^p}{p!} \, \frac{k^k}{\mathrm{dist}(t)^k} \, \sqrt{\frac{(J+(n-1)+2(p-|l|-k))!}{J!}},$$

with all constraints on $|j|, p, |l|, k$ satisfied. Thus the contribution stemming from $\Delta_8(w,t)$ satisfies the required estimate, provided

$$D_2^4 \;\geq\; c_1 \, c_3 \, \nu^2.$$

We now perform a similar analysis for the quantities $\Gamma_i(w,t)$ that appear in the expression for $\dot{c}_{n,p,l,k}(w,t)$. We integrate these terms with respect to t and apply Lemma 6.3. According to the lemma, integration of a term with a given value of p gives rise to a term with $p' = p+1$ in the estimates. We also note that the estimates we want to prove for the c's differ from those for the d's by the replacement of $n-1$ by n.

The Term $\int_0^t \Gamma_1$

We use the same techniques above to obtain

$$\left\| D_w^\alpha \, \int_0^t \, \frac{1}{2} \, (\Delta_w c_{n-2,p,l,k})(w,s) \, ds \right\|$$

$$\leq \; \sum_{i=1}^d \, \frac{D_1}{2 D_2^8} \, D_2^{|\alpha|+|l_{i,2}|+4n} \, \frac{(\alpha+l_{i,2})!}{(1+|\alpha|)^{d+1}} \, \frac{t^{p'}}{p'!} \, \frac{k^k}{\mathrm{dist}(t)^k} \, \sqrt{\frac{(J+n+2(p'-|l_{i,2}|-k))!}{J!}}.$$

We check that the constraints are satisfied:

$$\begin{array}{rcccl} p' & \leq & n-1 & < & n, \\ |l_{i,2}|+k & \leq & p+(n-2)/2+2 & = & p'+n/2 \\ |j| & \leq & J+(n-2)+2(p-|l|-k) & = & J+n+2(p'-|l_{i,2}|-k). \end{array}$$

Thus, each of the d contributions stemming from $\Gamma_1(w,t)$ satisfies the required estimate provided

$$D_2^8 \;\geq\; 1/2.$$

The Term $\int_0^t \Gamma_2$

Similarly, with $p' = p+1$,

$$\left\| D_w^\alpha \, \int_0^t \, \sum_{i=1}^d \, \langle \Phi, \, D_{w_i} \Phi \rangle (w,s) \; D_{w_i} c_{n-2,p,l,k}(w,s) \, ds \right\|$$

$$\leq \; \sum_{i=1}^d \, \frac{c_4' \, \nu \, D_1}{D_2^8} \, D_2^{|\alpha|+|l_{i,1}|+4n} \, \frac{(\alpha+l_{i,1})!}{(1+|\alpha|)^{d+1}} \, \frac{|t|^{p'}}{p'!} \, \frac{k^k}{\mathrm{dist}(t)^k} \, \sqrt{\frac{(J+n+2(p'-|l_{i,1}|-k))!}{J!}},$$

with all constraints on $|j|, p', |l_{i,1}|, k$ satisfied. Thus each of the d contributions stemming from $\Gamma_2(w,t)$ satisfies the required estimate, provided

$$D_2^8 \;\geq\; c_4'\,\nu.$$

The Term $\int_0^t \Gamma_3$

Again, with $p' = p+1$,

$$\left\| D_w^\alpha \int_0^t \frac{1}{2}\langle \Phi,\, (\Delta_w \Phi)\rangle(w,s)\; c_{n-2,p,l,k}(w,s)\, ds \right\|$$

$$\leq\; \frac{c_5'\,\nu\, D_1}{2\, D_2^8}\; D_2^{|\alpha|+|l|+4n}\; \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^{p'}}{p'!}\; \frac{k^k}{\mathrm{dist}(t)^k}\; \sqrt{\frac{(J+n+2(p'-|l|-k))!}{J!}},$$

with all constraints on $|j|, p', |l|, k$ satisfied. Thus, the contribution stemming from $\Gamma_3(w,t)$ satisfies the required estimate, provided

$$D_2^8 \;\geq\; c_5'\,\nu/2.$$

The Term $\int_0^t \Gamma_4$

Recall that the matrices $\langle \varphi,\, D_{y_i}\varphi\rangle$ are controlled by an analog of Lemma 2.1.

$$\left\| D_w^\alpha \int_0^t \sum_{i=1}^d \langle \varphi,\, D_{y_i}\varphi\rangle\; D_{w_i} c_{n-1,p,l,k}(w,s)\, ds \right\|$$

$$\leq\; \sum_{i=1}^d \frac{d_0\,\|B\|\, D_1}{D_2^4}\; D_2^{|\alpha|+|l_{i,1}|+4n}\; \frac{(\alpha+l_{i,1})!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^{p'}}{p'!}\; \frac{k^k}{\mathrm{dist}(t)^k}\; \sqrt{\frac{(J+n+2(p'-|l_{i,1}|-k))!}{J!}},$$

with all constraints on $|j|, p', |l_{i,1}|, k$ satisfied. Thus each of the d contributions stemming from $\Gamma_4(w,t)$ satisfies the required estimate, provided

$$D_2^4 \;\geq\; d_0\,\|B\|.$$

The Term $\int_0^t \Gamma_5$

For this term we obtain

$$\left\| D_w^\alpha \int_0^t \sum_{i=1}^d \langle \Phi,\, (D_{w_i}\Phi)(w,s)\rangle\, \langle \varphi,\, D_{y_i}\varphi\rangle\; c_{n-1,p,l,k}(w,s)\, ds \right\|$$

$$\leq\; \sum_{i=1}^d \frac{c_4'\,\nu\, d_0\,\|B\|\, D_1}{D_2^4}\; D_2^{|\alpha|+|l|+4n}\; \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^{p'}}{p'!}\; \frac{k^k}{\mathrm{dist}(t)^k}\; \sqrt{\frac{(J+n+2(p'-|l|-k))!}{J!}},$$

with all constraints on $|j|, p', |l|, k$ satisfied. Thus each of the d contributions stemming from $\Gamma_5(w,t)$ satisfies the required estimate, provided

$$D_2^4 \;\geq\; c_4'\, \nu\, d_0\, \|B\|.$$

The Term $\int_0^t \Gamma_6$

In this term, we encounter the sum over all previous c's. As in the similar contribution from Δ_2, we obtain

$$\left\| D_w^\alpha \sum_{\tilde m=3}^{n+2} \sum_{|m|=\tilde m} \int_0^t \frac{D^m E(a(t))}{m!}\, \langle \varphi,\, y^m \varphi\rangle\; c_{n+2-\tilde m,p,l,k}(w,s) \right\|$$

$$\leq \sum_{\tilde m=3}^{n} \sum_{|m|=\tilde m} D_1\; c_6\; D_2^8\, \frac{(d_0\, \|A\|)^{\tilde m}}{D_2^{3\tilde m}}\; D_2^{|\alpha|+|l|+4n}\; \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^{p'}}{p'!}\; \frac{k^k}{\mathrm{dist}(t)^k}$$

$$\times\; \sqrt{\frac{(J+n+2(p'-|l|-k))!}{J!}}.$$

We check that the constraints on the parameters and components of the vectors are satisfied

$$\begin{aligned} p' &\leq n-\tilde m+3 && \leq && n \\ |l|+k &\leq p+(n-\tilde m+2)/2 && \leq && p'+n/2 \\ |j| &\leq J+n+2+2(p-|l|-k) && = && J+n+2(p'-|l|-k). \end{aligned}$$

Hence we see that each contribution from $\Delta_2(w,t)$ satisfies the required bound, provided the following two conditions are fulfilled

$$\begin{aligned} D_2^3 &\geq d_0\, \|A\| \\ D_2 &\geq (d_0\, \|A\|)^3\, c_6. \end{aligned}$$

There are $\displaystyle\sum_{\tilde m=3}^{n} \sum_{|m|=\tilde m} 1 \;\leq\; \sigma_0\, \mathrm{e}^{\sigma n}$ such contributions, where $\sigma>0$ can be chosen arbitrarily small.

The Term $\int_0^t \Gamma_7$

This terms depends on the d's. Recall the estimates are a little different for them.

$$\left\| D_w^\alpha \int_0^t \sum_{i=1}^{d} \frac{1}{2}\, \langle \Phi(w,s),\, (D_{w_i}^2 d_{n-2,p,l,k})(w,s)\rangle\, ds \right\|$$

$$\leq \sum_{i=1}^{d} \frac{c_2\, \nu\, D_1}{2\, D_2^{12}}\; D_2^{|\alpha|+|l_{i,2}|+4n}\; \frac{(\alpha+l_{i,2})!}{(1+|\alpha|)^{d+1}}\; \frac{|t|^{p'}}{p'!}\; \frac{k^k}{\mathrm{dist}(t)^k}\; \sqrt{\frac{(J+n+2(p'-|l_{i,2}|-k))!}{J!}},$$

with all constraints on $|j|, p', |l_{i,2}|, k$ satisfied. Thus each of the d contributions stemming from $\Gamma_7(w,t)$ satisfies the required estimate, provided

$$D_2^{12} \;\geq\; c_2\,\nu/2.$$

The Term $\int_0^t \Gamma_8$

Similarly,

$$\left\| D_w^\alpha \int_0^t \sum_{i=1}^d \langle \varphi,\, D_{y_i}\varphi\rangle \;\langle \Phi(w,s),\, (D_{w_i} d_{n-1,p,l,k})(w,s)\rangle\, ds \right\|$$

$$\leq \sum_{i=1}^d \frac{c_2\,\nu\, d_0\, \|B\|\, D_1}{D_2^8}\, D_2^{|\alpha|+|l_{i,1}|+4n}\, \frac{(\alpha+l_{i,1})!}{(1+|\alpha|)^{d+1}}\, \frac{|t|^{p'}}{p'!}\, \frac{k^k}{\mathrm{dist}(t)^k}\, \sqrt{\frac{(J+n+2(p'-|l_{i,1}|-k))!}{J!}},$$

with all constraints on $|j|, p', |l_{i,1}|, k$ satisfied. Thus, each of the d contributions stemming from $\Gamma_8(w,t)$ satisfies the required estimate provided

$$D_2^8 \;\geq\; c_2\,\nu\, d_0\, \|B\|.$$

The Term $\int_0^t \Gamma_9$

Finally,

$$\left\| D_w^\alpha \int_0^t \langle \dot{\Phi}(w,s),\, d_{n,p,l,k}(w,s)\rangle\, ds \right\|$$

$$\leq \;\; \frac{c_3\,\nu\, D_1}{D_2^4}\, D_2^{|\alpha|+|l|+4n}\, \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}}\, \frac{|t|^{p'}}{p'!}\, \frac{k^k}{\mathrm{dist}(t)^k}\, \sqrt{\frac{(J+n+2(p'-|l|-k))!}{J!}},$$

with all constraints on $|j|, p', |l|, k$ satisfied. Thus, the contribution stemming from $\Gamma_9(w,t)$ satisfies the required estimate, provided

$$D_2^4 \;\geq\; c_3\,\nu.$$

By choosing D_2 large enough, all conditions are satisfied. This completes the induction for part iii) of Proposition 6.1.

The integration required to construct the c's shows that we obtain non-zero results for $c_{n,p,l,k,\beta}$ for $n>0$ only when $p \geq 1$. This proves part i) of Proposition 6.1.

We now turn to the proof of part ii) of Proposition 6.1.

6.3 Counting the Number of Terms that Occur in Our Expansion

In our Born–Oppenheimer expansion, the n^{th} order term has the form $\phi_n(w,y,t) \;=\; g_n(w,y,t)\Phi(w,t) + \phi_n^\perp(w,y,t)$. The way we compute $g_n(w,y,t)$ and $\phi_n^\perp(w,y,t)$, they decompose naturally as sums over the parameter β. We define u_n to be the number of such terms in $g_n(w,y,t)$ and v_n to be the number of terms in $\phi_n^{\perp}(w,y,t)$.

An examination of our construction shows that u_n and v_n satisfy the recursive estimates

$$u_{n+1} \;\leq\; \sum_{j=0}^{3} a_j\, u_{n-j} \,+\, \sum_{j=0}^{3} b_j\, v_{n-j} \,+\, \sum_{j=0}^{n} c_1\, \gamma_1^j\, u_{n-j} \,+\, \sum_{j=0}^{n} c_2\, \gamma_2^j\, v_{n-j} \,+\, v_{n+1} \tag{6.9}$$

$$v_{n+1} \;\leq\; \sum_{j=0}^{3} d_j\, u_{n-j} \,+\, \sum_{j=0}^{3} e_j\, v_{n-j} \,+\, \sum_{j=0}^{n} c_3\, \gamma_3^j\, u_{n-j} \,+\, \sum_{j=0}^{n} c_4\, \gamma_4^j\, v_{n-j}, \tag{6.10}$$

where a_i, b_i, c_i, d_i, e_i and γ_i are fixed numbers. The exponentials γ_i^j arise from an estimate (proven in the proof of Lemma 5.2 of [15]) for the number of Taylor series terms of any given order in the expansion of $E(a(t)+\epsilon y)$.

We substitute (6.10) for the last term in (6.9) and add the result to (6.10). By some simple estimates this leads to a recursive estimate for the single quantity $z_n = u_n + v_n$ of the form

$$z_{n+1} \;\leq\; \sum_{j=0}^{3} \widetilde{a}_j\, z_{n-j} \;+\; \sum_{j=0}^{n} \widetilde{c}\, \widetilde{\gamma}^j\, z_{n-j}.$$

An easy induction on n shows that this implies that z_n grows at most like e^{kn} for a sufficiently large value of k. The quantity z_n is the number of terms in $\phi_n(w,y,t)$, so this proves the assertion. ■

Proposition 6.1 now follows easily. ■

7 Exponential Error Bounds

In this section, we prove Theorem 4.1.

7.1 The Explicit Error Term

We use the following abstract lemma, whose proof is an easy application of Duhamel's formula (see e.g. [13]).

Lemma 7.1 *Suppose $H(\hbar)$ is a family of self-adjoint operators for $\hbar > 0$. Suppose $\psi(t,\,\hbar)$ belongs to the domain of $H(\hbar)$, is continuously differentiable in t, and approximately solves the Schrödinger equation in the sense that*

$$i\,\hbar\,\frac{\partial\psi}{\partial t}(t,\,\hbar) \;=\; H(\hbar)\,\psi(t,\,\hbar) \;+\; \xi(t,\,\hbar),$$

where $\xi(t,\,\hbar)$ satisfies

$$\|\,\xi(t,\,\hbar)\,\| \;\leq\; \mu(t,\,\hbar).$$

Then, for $t > 0$,

$$\|\, e^{-itH(\hbar)/\hbar}\,\psi(0,\,\hbar) \;-\; \psi(t,\,\hbar)\,\| \;\leq\; \hbar^{-1}\int_0^t \mu(s,\,\hbar)\,ds.$$

The analogous statement holds for $t < 0$.

We substitute our approximate solution (4.1)
$F\ \mathrm{e}^{iS/\epsilon^2}\ \mathrm{e}^{i\eta\cdot y/\epsilon}\ \left(\sum_{n=0}^{N}\epsilon^n\,\phi_n\ +\ \epsilon^{N+1}\,\phi_{N+1}^{\perp}\ +\ \epsilon^{N+2}\,\phi_{N+2}^{\perp}\right)$ into the Schrödinger equation and compute the residual term ξ_N.

It is more convenient to write this term in the multiple scales notation. We also use the notation $\epsilon^m\,\dfrac{E^{(m)}(a(t))}{m!}y^m$ to denote the Taylor series term $\sum_{|j|=m}\epsilon^{|j|}\,\dfrac{(D^jE)(a(t))}{j!}\,y^j$.

In this notation, the residual $\xi_N(w,y,t)$ is given, up to a phase factor, by two sums of terms. The first one contains all terms that do not involve derivatives of the cut-off. The second contains all terms that do involve derivatives of the cut-off.
The first sum is $F(w)$ times the following:

$$\frac{\epsilon^{N+3}}{2}\,\left(\Delta_w g_{N-1}\right)\,\Phi \tag{7.1}$$

$$+\quad\frac{\epsilon^{N+4}}{2}\,\left(\Delta_w g_N\right)\,\Phi \tag{7.2}$$

$$+\quad\epsilon^{N+3}\,\left(\nabla_w g_{N-1}\right)\cdot\left(\nabla_w\Phi\right) \tag{7.3}$$

$$+\quad\epsilon^{N+4}\,\left(\nabla_w g_N\right)\cdot\left(\nabla_w\Phi\right) \tag{7.4}$$

$$+\quad\frac{\epsilon^{N+3}}{2}\,g_{N-1}\,\left(\Delta_w\Phi\right) \tag{7.5}$$

$$+\quad\frac{\epsilon^{N+4}}{2}\,g_N\,\left(\Delta_w\Phi\right) \tag{7.6}$$

$$+\quad\frac{\epsilon^{N+3}}{2}\,\left(\Delta_w\phi_{N-1}^{\perp}\right) \tag{7.7}$$

$$+\quad\frac{\epsilon^{N+4}}{2}\,\left(\Delta_w\phi_N^{\perp}\right) \tag{7.8}$$

$$+\quad\epsilon^{N+3}\,\left(\nabla_w\cdot\nabla_y g_N\right)\Phi \tag{7.9}$$

$$+\quad\epsilon^{N+3}\,\left(\nabla_y g_N\right)\cdot\left(\nabla_w\Phi\right) \tag{7.10}$$

$$+\quad\epsilon^{N+3}\,\left(\nabla_w\cdot\nabla_y\phi_N^{\perp}\right) \tag{7.11}$$

$$+\quad i\,\epsilon^{N+3}\,\dot{\phi}_{N+1}^{\perp} \tag{7.12}$$

$$+\quad i\,\epsilon^{N+4}\,\dot{\phi}_{N+2}^{\perp} \tag{7.13}$$

$$+\quad\frac{\epsilon^{N+5}}{2}\,\left(\Delta_w\phi_{N+1}^{\perp}\right) \tag{7.14}$$

$$+\quad\frac{\epsilon^{N+6}}{2}\,\left(\Delta_w\phi_{N+2}^{\perp}\right) \tag{7.15}$$

$$+ \quad \epsilon^{N+4} \left(\nabla_w \cdot \nabla_y \phi_{N+1}^{\perp}\right) \tag{7.16}$$

$$+ \quad \epsilon^{N+5} \left(\nabla_w \cdot \nabla_y \phi_{N+2}^{\perp}\right) \tag{7.17}$$

$$+ \quad \frac{\epsilon^{N+3}}{2} \left(\Delta_y \phi_{N+1}^{\perp}\right) \tag{7.18}$$

$$+ \quad \frac{\epsilon^{N+4}}{2} \left(\Delta_y \phi_{N+2}^{\perp}\right) \tag{7.19}$$

$$- \quad \frac{\epsilon^{N+3}}{2} E^{(2)}(a(t))\, y^2\, \phi_{N+1}^{\perp} \tag{7.20}$$

$$- \quad \frac{\epsilon^{N+4}}{2} E^{(2)}(a(t))\, y^2\, \phi_{N+2}^{\perp} \tag{7.21}$$

$$- \quad \sum_{n=0}^{N} \epsilon^{N-n} \left(E(a(t)+\epsilon y) \; - \sum_{m \leq 2+n} \epsilon^m \, \frac{E^{(m)}(a(t))}{m!}\, y^m \right) g_{N-n}\, \Phi \tag{7.22}$$

$$- \quad \sum_{n=0}^{N} \epsilon^{N-n} \left(E(a(t)+\epsilon y) \; - \sum_{m \leq 2+n} \epsilon^m \, \frac{E^{(m)}(a(t))}{m!}\, y^m \right) \phi_{N-n}^{\perp} \tag{7.23}$$

$$- \quad \epsilon^{N+1} \left(E(a(t)+\epsilon y) \; - \sum_{m \leq 2} \epsilon^m \, \frac{E^{(m)}(a(t))}{m!}\, y^m \right) \phi_{N+1}^{\perp} \tag{7.24}$$

$$- \quad \epsilon^{N+2} \left(E(a(t)+\epsilon y) \; - \sum_{m \leq 2} \epsilon^m \, \frac{E^{(m)}(a(t))}{m!}\, y^m \right) \phi_{N+2}^{\perp}. \tag{7.25}$$

The second sum arises from terms in which the cut-off $F(w)$ is differentiated. It is

$$\sum_{n=0}^{N} \frac{\epsilon^{n+4}}{2} \left(\Delta_w F\right) g_n\, \Phi \tag{7.26}$$

$$+ \quad \sum_{n=0}^{N+2} \frac{\epsilon^{n+4}}{2} \left(\Delta_w F\right) \phi_n^{\perp} \tag{7.27}$$

$$+ \quad \sum_{n=0}^{N} \epsilon^{n+4} \left(\nabla_w F\right) \cdot \left(\nabla_w g_n\right) \Phi \tag{7.28}$$

$$+ \quad \sum_{n=0}^{N} \epsilon^{n+4}\, g_n \left(\nabla_w F\right) \cdot \left(\nabla_w \Phi\right) \tag{7.29}$$

$$+ \quad \sum_{n=0}^{N+2} \epsilon^{n+4} \left(\nabla_w F\right) \cdot \left(\nabla_w \phi_n^{\perp}\right) \tag{7.30}$$

$$+ \quad \sum_{n=0}^{N} \epsilon^{n+3} \left(\nabla_w F\right) \cdot \left(\nabla_y g_n\right) \Phi \tag{7.31}$$

$$+\sum_{n=0}^{N+2} \epsilon^{n+3}\, (\nabla_w F)\cdot(\nabla_y \phi_n^{\perp}) \tag{7.32}$$

7.2 Optimal Truncation

Each error term in the first sum (7.1)–(7.25) can be written as a uniformly bounded function times one of the following two forms:

$$\begin{aligned}
\mathcal{A} &= \Psi(w,t) \sum_r \sum_{|j|\le\rho(r)} c_{r,j}(w,t)\ \varphi_j(y,t)\\
\mathcal{B} &= \sum_{r'} \sum_{|j|\le\rho'(r')} d_{r',j}(w,t)\ \varphi_j(y,t),
\end{aligned}$$

where $\Psi(w,t)\in\mathcal{H}_{\rm el}$, $\varphi_j(y,t)=\epsilon^{-d/2}\varphi_j(A(t),B(t),1,0,0,y)$, r, r' denote a collective set of indices that belong to some finite set, and $\rho(r)$ and $\rho'(r')$ limit the number of multi-indices j allowed in the second sum.

The error term $\xi(w,y,t)\in\mathcal{H}_{\rm el}$ needs to be estimated for $t\in I\!R$, in the following norm

$$\begin{aligned}
\|\xi(t)\| &= \left\{\int_{\mathbb{R}^d} \|\xi\,(x-a(t),\,(x-a(t))/\epsilon,\,t)\,\|^2_{\mathcal{H}_{\rm el}}\,dx\right\}^{1/2}\\
&= \left\{\int_{\mathbb{R}^d} \|\xi(w,w/\epsilon,t)\|^2_{\mathcal{H}_{\rm el}}\,dw\right\}^{1/2}.
\end{aligned}$$

With that norm, using the Cauchy–Schwarz inequality and the $L^2(I\!R^d)$ orthonormality of the $\varphi_j(y,t)$, we obtain the following estimate for the norm of $\mathcal{A}$ in terms of the norm of vector $c_r(w,t)\in l^2(\mathbf{N}^d,\mathbb{C})$,

$$\|\mathcal{A}\| \le \sum_r \sup_{w\in{\rm supp}\,F} \|\Psi(w,t)\|_{\mathcal{H}_{\rm el}} \sup_{w\in{\rm supp}\,F} \|c_r(w,t)\| \left(\sum_{|j|\le\rho(r)} 1\right)^{1/2}. \tag{7.33}$$

By similar arguments we get the following estimate for the norm of $\mathcal{B}$ in terms of the norm of the vector $d_{r'}(w,t)\in l^2(\mathbf{N}^d,\mathcal{H}_{\rm el})$,

$$\|\mathcal{B}\| \le \sum_{r'} \sup_{w\in{\rm supp}\,F} \|d_{r'}(w,t)\| \left(\sum_{|j|\le\rho'(r')} 1\right)^{1/2}. \tag{7.34}$$

Note also that

$$\sum_{|j|\le\rho'(r')} 1 \le \binom{\rho'(r')+d}{d}, \tag{7.35}$$

which grows at most polynomially with $\rho'(r')$.

Lemma 7.2 *For $t \in [0,\, T]$, and for any $\alpha \in \mathbf{N}^d$ and $\gamma \in \mathbf{N}^d$, there exist $C_0 > 0$ and $\tau_0 > 0$, such that*

$$\sum_{\beta \in \mathcal{B}_{n,1}} \ \sum_{p \leq n} \ \sum_{k+|l| \leq p+\frac{n}{2}} \ \sum_{|j| \leq J+n+2(p-|l|-k)} \left\| \Psi(w,t) D_w^\alpha D_y^\gamma c_{n,p,l,k,\beta,j}(w,t) \varphi_j(y,t) \right\|$$

$$\leq \ C_0 \left\{ n^{1/2} \tau_0 \right\}^n \tag{7.36}$$

and

$$\sum_{\beta \in \mathcal{B}_{n,2}} \ \sum_{p \leq n-1} \ \sum_{k+|l| \leq p+\frac{n-1}{2}} \ \sum_{|j| \leq J+n-1+2(p-|l|-k)} \left\| D_w^\alpha D_y^\gamma d_{n,p,l,k,\beta,j}(w,t) \varphi_j(y,t) \right\|$$

$$\leq \ C_0 \left\{ n^{1/2} \tau_0 \right\}^n \tag{7.37}$$

If the operator D_y^γ is replaced by the operator y^γ, the same bounds are valid.

Proof: We begin with (7.36). We have

$$\sum_{|j| \leq J+n+2(p-|l|-k)} D_w^\alpha c_{n,p,l,k,\beta,j}(w,t) \ D_y^\gamma \varphi_j(y,t)$$

$$= \sum_{|\tilde{k}| \leq J+|\gamma|+n+2(p-|l|-k)} \Big(\langle \varphi,\, D_y^\gamma \varphi \rangle \ D_w^\alpha c_{n,p,l,k,\beta}(w,t) \Big)_{\tilde{k}} \ \varphi_{\tilde{k}}(y,t).$$

We know that the vector $\langle \varphi,\, D_y^\gamma \varphi \rangle \ D_w^\alpha c_{n,p,l,k,\beta}(w,t)$ satisfies the estimate

$$\Big\| \langle \varphi,\, D_y^\gamma \varphi \rangle \, D_w^\alpha c_{n,p,l,k,\beta}(w,t) \Big\| \ \leq \ D_1 \, D_2^{|\alpha|+|l|+4n} \ \frac{(\alpha+l)!}{(1+|\alpha|)^{d+1}} \ \frac{|t|^p}{p!} \ \frac{k^k}{\delta^k} \ (\|B\| \, d_0)^{|\gamma|}$$

$$\times \ \sqrt{\frac{(J+|\gamma|+n+2(p-|l|-k))!}{J!}}. \tag{7.38}$$

Here $\delta > 0$ is the distance in the complex plane from $[0,\, T]$ to the complement of Ω.

Since the number of indices in $\mathcal{B}_{n,1}$ is bounded by $\mathrm{e}^{K_0 n}$, $D_2 \geq 1$, and $(\alpha+l)! \leq (|\alpha|+|l|)!$, we can estimate the sum (7.36) by

$$\frac{D_1 \, D_2^{|\alpha|} \, (\|B\| d_0)^{|\gamma|}}{\sqrt{J!} \, (1+|\alpha|)^{d+1}} \ \mathrm{e}^{K_0 n} \ D_2^{11n/2} \ \sum_{p \leq n} \frac{|t|^p}{p!} \ \sum_{|l|+k \leq p+n/2} \left(\frac{k}{\delta} \right)^k$$

$$\times \quad (|\alpha|+|l|)! \ \sqrt{(J+|\gamma|+n+2(p-|l|-k))!}. \tag{7.39}$$

Then, using $a!b! \leq (a+b)!$, the fact that $(a+2p)!/(p!)^2$ is increasing in p, and $p \leq n$, we have

$$\frac{(J+|\gamma|+n+2(p-|l|-k))! \ ((|\alpha|+|l|)!)^2}{(p!)^2} \ \leq \ \frac{(J+|\gamma|+2|\alpha|+3n-2k)!}{(n!)^2},$$

so that (7.39) is bounded by

$$\frac{D_1\, D_2^{|\alpha|}\, (\|B\| d_0)^{|\gamma|}}{n!\, \sqrt{J!}\, (1+|\alpha|)^{d+1}}\ \mathrm{e}^{K_0 n}\ D_2^{11n/2} \sum_{p\le n} |t|^p \sum_{k=0}^{p+n/2} \left(\frac{k}{\delta}\right)^k$$
$$\times\ \sqrt{(J+|\gamma|+2|\alpha|+3n-2k)!} \sum_{|l|\le p+n/2-k} 1.$$

The last term is bounded by $\binom{[\![3n/2]\!]+d}{d} \le \sigma_0\, \mathrm{e}^{3\sigma n/2}$, where $[\![x]\!]$ denotes the integer part of x.

Using $k^{2k} \le (2k)^{2k}$, $a^a b^b \le (a+b)^{a+b}$ and $a! \le a^a$ we have

$$(J+|\gamma|+2|\alpha|+3n-2k)!\ k^{2k}\ \le\ (J+|\gamma|+2|\alpha|+3n)^{J+|\gamma|+2|\alpha|+3n}.$$

Since we can assume without loss that $\delta < 1$, this implies

$$\begin{aligned}
&\sum_{k=0}^{p+n/2} \left(\frac{k}{\delta}\right)^k \sqrt{(J+|\gamma|+2|\alpha|+3n-2k)!} \\
\le\ & (J+|\gamma|+2|\alpha|+3n)^{\frac{J+|\gamma|+2|\alpha|+3n}{2}} \sum_{k=0}^{p+n/2} \delta^{-k} \\
\le\ & (J+|\gamma|+2|\alpha|+3n)^{\frac{J+|\gamma|+2|\alpha|+3n}{2}}\ \frac{K_1}{\delta^{n/2}}\ \delta^{-p},
\end{aligned} \tag{7.40}$$

for some constant K_1 that satisfies $\delta^{-1} - 1 \ge K_1^{-1}\delta^{-1}$. Together with

$$\sum_{p\le n} (t/\delta)^p\ \le\ \begin{cases} K_2 (t/\delta)^n & \text{if}\quad t/\delta > 1 \\ K_2^n & \text{if}\quad t/\delta = 1 \\ K_2 & \text{if}\quad t/\delta < 1, \end{cases} \tag{7.41}$$

where K_2 is constant, we get (in the first case above)

$$\begin{aligned}
&\sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\le n} \sum_{k+|l|\le p+\frac{n}{2}} \left\| \langle \varphi,\, D_y^\gamma \varphi\rangle\, D_w^\alpha c_{n,p,l,k,\beta}(w,t) \right\| \\
\le\ & \frac{\sigma_0 K_1 K_2 D_1 D_2^{|\alpha|} (\|B\| d_0)^{|\gamma|}}{(1+|\alpha|)^{d+1}}\ \frac{\mathrm{e}^{(K_0+3\sigma/2)n} D_2^{11n/2} t^n}{\sqrt{J!}\, n!\, \delta^{3n/2}}\ (J+|\gamma|+2|\alpha|+3n)^{\frac{J+|\gamma|+2|\alpha|+3n}{2}}.
\end{aligned}$$

We postpone the study of the dependence of our estimates on t and J to Section 8. So, using the above,

$$(J+|\gamma|+2|\alpha|+3n)^{\frac{J+|\gamma|+2|\alpha|+3n}{2}} \le (J+|\gamma|+2|\alpha|+3n)^{\frac{J+|\gamma|+2|\alpha|}{2}} ((J+|\gamma|+2|\alpha|+3)n)^{\frac{3n}{2}},$$

and the existence of $0 < a < b$, such that $a^n n^n \le n! \le b^n n^n$, we learn the existence of positive constants (*i.e.*, independent of n) K_3, K_4 and K_5, such that

$$\sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\le n} \sum_{k+|l|\le p+\frac{n}{2}} \left\| \langle \varphi,\, D_y^\gamma \varphi\rangle\ D_w^\alpha c_{n,p,l,k,\beta}(w,t) \right\|\ \le\ K_3\, K_4^n\, \frac{n^{3n/2}}{a^n n^n}\ \le\ K_3\, (K_5\, n^{1/2})^n.$$

This yields the result with $C_0 = K_3$ and $\tau_0 = K_5$.

The second sum is dealt with in the same manner, since the vectors $d_{n,p,l,k,\beta}(w,t)$ satisfy the same bounds as $c_{n,p,l,k,\beta}(w,t)$ does with n replaced by $n-1$.

Finally, the replacement of D_y^γ by the operator y^γ means that the matrix $\langle\varphi|D_y^\gamma\varphi\rangle$ must be replaced by the matrix $\langle\varphi|y^\gamma\varphi\rangle$. But the latter has the same properties as the former; the bounds above remain true with $\|B\|$ replaced by $\|A\|$ from (7.38) onward. This affects the definition of C_0 only. ∎

The following lemma is the key to the proof of exponential accuracy of our approximation by means of optimal truncation.

Lemma 7.3 *For sufficiently small $g>0$, there exist $\Gamma(g)$ and $C(g)>0$ such that the choice $N(\epsilon) = [\![g^2/\epsilon^2]\!]$ implies that the norm of the error term $\xi_{N(\epsilon)}(t)$ given by (7.1) satisfies*

$$\|\xi_{N(\epsilon)}(t)\| \;\le\; C(g)\, e^{-\Gamma(g)/\epsilon^2}.$$

Proof: The previous lemma, formulas (7.33), (7.34) and (7.35) show that all terms in the first sum defining ξ_N except (7.12), (7.13), (7.22), and (7.23) are exponentially small, once we prove

$$C_0\epsilon^{N(\epsilon)}\;\left\{N(\epsilon)^{1/2}\;\tau_0\right\}^{N(\epsilon)} \;\le\; C\;\mathrm{e}^{-\Gamma/\epsilon^2}. \tag{7.42}$$

Because $g^2/\epsilon^2 - 1 \le N \le g^2/\epsilon^2$, if we choose $0 < g < 1/\tau_0$, the left hand side of this inequality is bounded by

$$C_0\left\{\epsilon N^{1/2}\tau_0\right\}^N \;\le\; C_0\left\{g\tau_0\right\}^N \;\le\; C_0\,\mathrm{e}^{-|\ln(g\tau_0)|N} \;\le\; C_0\,\mathrm{e}^{|\ln(g\tau_0)|}\;\mathrm{e}^{-|\ln(g\tau_0)|g^2/\epsilon^2}, \tag{7.43}$$

which gives

$$C(g) \;=\; C_0\,\mathrm{e}^{|\ln(g\tau_0)|} \quad\text{and}\quad \Gamma(g) \;=\; |\ln(g\tau_0)|\;g^2.$$

The terms (7.12) and (7.13) can be dealt with in a similar fashion once we have computed

$$\begin{aligned}\dot{\phi}^{\perp}_{N+1} \;=\; \sum_{\beta\in\mathcal{B}_{N+1,2}}\;\sum_{p\le N}\;\sum_{k+|l|\le p+\frac{N}{2}}\;\sum_{|j|\le J+N+2(p-|l|-k)} &\dot{d}_{N+1,p,l,k,\beta,j}(w,t)\,\varphi_j(y,t)\\ &+\;\; d_{N+1,p,l,k,\beta,j}(w,t)\,\dot{\varphi}_j(y,t),\end{aligned}$$

where the second term equals

$$\begin{aligned}\sum_{\beta\in\mathcal{B}_{N+1,2}}\;\sum_{p\le N}\;\sum_{k+|l|\le p+\frac{N}{2}}\;\sum_{|\tilde{k}|\le J+N+2+2(p-|l|-k)} &\left(\left(\frac{i}{2}\,\langle\varphi,\,\Delta_y\varphi\rangle\right.\right.\\ &\left.\left.-\;\;\frac{iE^{(2)}(a(t))}{2}\,\langle\varphi,\,y^2\varphi\rangle\right)\,d_{N+1,p,l,k,\beta}(w,t)\right)_{\tilde{k}}\;\varphi_{\tilde{k}}(y,t).\end{aligned}$$

Lemma 6.2 shows that $\dot{d}_{N+1,p,l,k,\beta}$ satisfies bounds similar to those satisfied by $d_{N+1,p,l,k,\beta,j}$ and the term above is taken care of by lemma 7.2. Similar statements are true for $\dot{\phi}^{\perp}_{N+2}$, and the analysis above also applies to these error terms.

Next consider (7.22). By the mean value theorem, there exists $\zeta_q(y,t,\epsilon) = a(t) + \theta_q(y,t,\epsilon)\,\epsilon y$, where $q \in \mathbf{N}^d$ and $\theta_q(y,t,\epsilon) \in (0,1)$, such that

$$E(a(t)+\epsilon y) \;-\; \sum_{m\le 2+n} \epsilon^m\, \frac{E^{(m)}(a(t))}{m!}\, y^m \;=\; \sum_{|q|=2+n+1} \epsilon^{|q|}\, \frac{D^q\, E(\zeta_q(y,t,\epsilon))}{q!}\, y^q.$$

Hence, we need to estimate

$$\sum_{n=0}^{N} \epsilon^{N+3} \sum_{|q|=2+n+1} \frac{D^q E(\zeta_q(y,t,\epsilon))}{q!}\, y^q \sum_{\beta,p,k,l,j} c_{N-n,p,l,k,\beta,j}(w,t)\, \varphi_j(y,t)\, \Phi, \tag{7.44}$$

with the following restrictions

$$\begin{aligned} |j| &\le J + (N-n) + 2(p-k-|l|) \\ k+|l| &\le p + (N-n)/2 \\ p &\le N-m \\ \beta &\in \mathcal{B}_{1,N-n}. \end{aligned} \tag{7.45}$$

We take a fixed value of $n\in[0,N]$, and consider the vectors

$$\frac{D^q E(\zeta_q(y,t,\epsilon))}{q!}\, \langle \varphi,\, y^q \varphi\rangle(t) c_{N-n,p,l,k,\beta}(w,t)$$

we have to estimate. Due to the presence of the cut-off function F (which we have omitted in the notation), we have

$$\frac{|D^q E(\zeta_q(y,t,\epsilon))|}{q!} \;\le\; \frac{c_6\, D_2^{|q|}}{(1+|q|)^{d+1}},$$

and with our bounds on the matrix $\langle \varphi,\, y^q\varphi\rangle(t)$ and on the vector $c_{N-n,p,l,k,\beta}(w,t)$, we can write

$$\left\| \frac{D^q\, E(\zeta_q(y,t,\epsilon))}{q!}\, \langle \varphi,\, y^q\varphi\rangle(t)\; c_{N-n,p,l,k,\beta}(w,t) \right\| \tag{7.46}$$

$$\le\; \frac{c_6\, D_2^{n+3}\, (d_0\|A\|)^{n+3}}{(1+(n+3))^{d+1}}\; \frac{\sqrt{(J+3+N+2(p-k-|l|))!}}{\sqrt{J!}}\; D_1\; D_2^{|l|+4(N-n)}\; l!\; \frac{|t|^p}{p!}\; \frac{k^k}{\delta^k}.$$

Then we use similar estimates to the above and the restrictions (7.45) to get

$$k^{2k}\, l!\, l!\, \frac{(J+3+N+2(p-k-|l|))!}{p!\, p!} \;\le\; (2k)^{2k}\; \frac{(J+3+N+2(p-k))!}{p!\, p!}$$

$$\le\; (2k)^{2k}\; \frac{(J+3+3N-2n-2k)!}{(N-n)!\,(N-n)!} \;\le\; \frac{(J+3+3N-2n)^{J+3+3N-2n}}{(N-n)!\;(N-n)!}.$$

Using this and $|l|\le 3(N-n)/2$, we see that (7.46) is bounded above by

$$\frac{c_6 D_1 (D_2 d_0 \|A\|)^3}{4^{d+1}\sqrt{J!}}\; D_2^{11N/2}\; \frac{(d_0\|A\|)^n}{D_2^{9n/2}}\; \frac{|t|^p}{(N-n)!\delta^k}\; (J+3+3N-2n)^{(J+3+3N)/2}.$$

Finally, with

$$\sum_{p=0}^{N-n} \sum_{k=0}^{p+\frac{N-m}{2}} |t|^p\, \delta^{-k} \;\leq\; K_1\, K_2\, \delta^{-(N-m)/2} \left(\frac{t}{\delta}\right)^{N-n},$$

(see (7.40), (7.41)), the bounds $\sum_{|l|\leq p+(N-n)/2} 1 \;\leq\; \sigma_0\, \mathrm{e}^{3\sigma(N-n)/2}$, $\sum_{|q|\leq n+3} 1 \;\leq\; \sigma_0\, \mathrm{e}^{\sigma(n+3)}$, and $|\mathcal{B}_{1,N-n}| \leq \mathrm{e}^{K_0(N-n)}$, we get (with the conditions (7.45) on the summations)

$$\begin{aligned}
&\sum_{|q|=2+n+1} \sum_{\beta,p,k,l,j} \epsilon^N \left\| \frac{D^q\, E(\zeta_q(y,t,\epsilon))}{q!}\, \langle \varphi,\, y^q \varphi\rangle(t)\, c_{N-n,p,l,k,\beta}(w,t) \right\| \\
\leq\;& \frac{\sigma_0^2 \mathrm{e}^{3\sigma} K_1 K_2 c_6 D_1 (D_2 d_0 \|A\|)^3}{4^{d+1}\sqrt{J!}}\, \mathrm{e}^{5\sigma N/2}\, D_2^N\, (d_0\|A\|)^N\, \epsilon^N\, (J+3+3N)^{(J+3+N)/2} \\
&\qquad \times \left(\frac{D_2^{9/2}}{\delta^{1/2}\, d_0\, \|A\|}\right)^{N-n} \left(\frac{t}{\delta}\right)^{N-n} \frac{(J+3+3N)^{N-n}}{(N-n)!}.
\end{aligned} \tag{7.47}$$

Postponing the study of the t and J dependence of our estimates, we use the bound $(J+3+3N)^{(J+3+N)/2} \;\leq\; N^{N/2}\,(J+3+3N)^{(J+3)/2}\,(J+6)^{N/2}$ to establish the existence of constants L_0, L_1, L_2, independent of N and n, such that (7.47) is bounded above by

$$L_0\, L_1^N\, \epsilon^N\, N^{N/2}\, \frac{(L_2\, N)^{N-n}}{(N-n)!}.$$

It remains for us to sum over n and use (7.33) to bound (7.44) by

$$\begin{aligned}
&\epsilon^3\, \sqrt{\sigma_0}\, \mathrm{e}^{3\sigma N/2}\, L_0\, L_1^N\, \epsilon^N N^{N/2} \sum_{n=0}^{N} \frac{(L_2\, N)^{N-n}}{(N-n)!} \\
\leq\;& \epsilon^3\, \sqrt{\sigma_0}\, L_0\, (\mathrm{e}^{3\sigma/2}\, L_1)^N\, \epsilon^N\, N^{N/2} \sum_{s=0}^{\infty} \frac{(L_2\, N)^s}{s!} \\
\leq\;& \epsilon^3\, \sqrt{\sigma_0}\, L_0\, (\mathrm{e}^{3\sigma/2}\, L_1\, \mathrm{e}^{L_2})^N\, \epsilon^N\, N^{N/2}.
\end{aligned}$$

If we choose $g < 1/(L_1\, \mathrm{e}^{L_2+3\sigma/2})$, we can apply the analysis (7.43) to obtain an exponentially small bound on (7.44) by the optimal truncation $N(\epsilon) \;=\; [\![g^2/\epsilon^2]\!]$.

Since the estimates we have on the d's are similar to those we have on the c's, with the replacement of n by $n-1$, the same exponential bound is valid for (7.23), (see (7.34)) and the analysis of the the first collection of error terms is completed.

We now need to take into account the error terms (7.26) to (7.32) arising from the derivatives of the cut-off function F. Choose $F_0 > 0$ that satisfies

$$\max\, \{\, |\Delta_w F(w)|,\; \|\nabla_w F(w)\|\, \} \;\leq\; F_0,$$

uniformly in w, and recall that for any $i = 1, \ldots, d$,

$$\mathrm{supp}\, \partial_{w_i} F(w) \;\subseteq\; \{ w \in I\!\!R^d \;:\; b_0 < |w| < b_1 \} \tag{7.48}$$

for some $0 < b_0 < b_1 < \infty$. Now consider (7.26). We express g_n in terms of the c's to see that the norm of $\frac{2}{\epsilon^4}$ times (7.26) (in $L^2(I\!R^d, \mathcal{H}_{\rm el})$) can be bounded as follows:

$$\left\| \sum_{n=0}^{N} \Delta_w\, F(w)\, g_n(w,y,t)\, \epsilon^n\, \Phi(w,t) \right\|$$

$$\leq \sum_{n=0}^{N} \epsilon^n \sqrt{\int_{\mathbb{R}^d} \left| \Delta_w F(w) \sum_{|j|\leq J+3n} c_{n,j}(w,t)\, \epsilon^{-d/2}\, \varphi_j(w/\epsilon,t) \right|^2 \|\Phi(w,t)\|^2_{\mathcal{H}_{\rm el}}\, dw}$$

$$\leq F_0 \sum_{n=0}^{N} \epsilon^n \sup_{w\in {\rm supp} F\subseteq \mathbb{R}^d} \|c_n(w,t)\| \sqrt{\sum_{|j|\leq J+3n} \int_{|w|\geq b_0} \left|\epsilon^{-d/2}\, \varphi_j(w/\epsilon,t)\right|^2 dw}. \tag{7.49}$$

We know from Section 7 of [15] that there exists a constant $0 < \beta_d$ depending on the dimension d only, such that

$$\sqrt{2|j|+d} \;<\; b_0/(\|A\|\epsilon), \qquad \text{for all } |j| \leq J+3N$$

and $|j| \leq J+3N$ imply

$$\sqrt{\int_{|w|\geq b_0} \left|\epsilon^{-d/2}\, \varphi_j(w/\epsilon,t)\right|^2 dw} \;\leq\; {\rm e}^{\beta_d |j|}\; {\rm e}^{-\,(b_0^2)/(12\,\|A\|^2\,\epsilon^2)}.$$

All the conditions here will be satisfied if $N(\epsilon) = [\![g^2/\epsilon^2]\!]$, provided we choose g and ϵ to satisfy

$$\epsilon^2(d+2J) + 6g^2 < b_0^2/\|A\|^2.$$

For such a choice, using $\sum_{|j|\leq J+3n} {\rm e}^{2\,\beta_d\,|j|} \;\leq\; \sigma_0\, {\rm e}^{(\sigma+2\beta_d)(J+3n)}$, we get

$$\sqrt{\sum_{|j|\leq J+3n} \int_{|w|\geq b_0} \left|\epsilon^{-d/2} \varphi_j(w/\epsilon,t)\right|^2 dw} \;\leq\; \sqrt{\sigma_0}\; {\rm e}^{(\sigma+2\beta_d)(J+3n)/2}\; {\rm e}^{-\,(b_0^2)(12\|A\|^2\epsilon^2)}.$$

Moreover, by means of manipulations that by now are familiar,

$$\|c_n(w,t)\| \;\leq\; \sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\leq n} \sum_{k+|l|\leq p+\frac{n}{2}} \|c_{n,p,l,k,\beta}(w,t)\|$$

$$\leq \sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\leq n} \sum_{k+|l|\leq p+\frac{n}{2}} D_1\, D_2^{|l|+4n}\, l!\, \frac{|t|^p}{p!}\, \frac{k^k}{\delta^k}\, \frac{(J+n+2(p-|l|-k))}{\sqrt{J!}}$$

$$\leq \sum_{\beta\in\mathcal{B}_{n,1}} \sum_{p\leq n} \sum_{k+|l|\leq p+\frac{n}{2}} \frac{D_1 D_2^{11n/2}}{\sqrt{J!}}\, \frac{|t|^p}{\delta^k}\, \frac{(J+3n)^{(J+3n)/2}}{n!}$$

$$\leq {\rm e}^{K_0 n}\, \sigma_0\, {\rm e}^{3\sigma n/2}\, K_1 K_2\, \frac{1}{\delta^n} \left(\frac{|t|}{\delta}\right)^n \frac{D_1 D_2^{11n/2}}{\sqrt{J!}}\, (J+3n)^{J/2}\, \frac{(J+3N)^{3n/2}}{n!}.$$

Combining these estimates, we get the existence of positive constants M_0 and M_1, such that for $N = [\![g^2/\epsilon^2]\!]$,

$$\left\| \sum_{n=0}^{N} \Delta_w \, F(w) \, g_n(w,y,t) \, \epsilon^n \, \Phi(w,t) \right\| \;\leq\; \mathrm{e}^{-(b_0^2)/(12\,\|A\|^2\,\epsilon^2)} \sum_{n=0}^{N} M_0 \, \frac{(\epsilon M_1 N^{3/2})^n}{n!}$$

$$\leq\; \mathrm{e}^{-(b_0^2)/(12\|A\|^2\epsilon^2)} \, M_0 \, \mathrm{e}^{\epsilon M_1 N^{3/2}} \;\leq\; \mathrm{e}^{-(b_0^2)/(12\|A\|^2\epsilon^2)} \, M_0 \, \mathrm{e}^{M_1 g^3/\epsilon^2} \;\leq\; M_0 \, \mathrm{e}^{-(b_0^2)/(24\|A\|^2\epsilon^2)},$$

provided

$$M_1 g^3 < b_0^2/(24\|A\|^2).$$

All other terms in the list (7.26) to (7.32) can be estimated in a similar fashion under a similar condition on g.

This concludes the proof of our lemma. ■

Remark: It is not difficult to check that if we keep N fixed, then our approximation (4.1) $\hat{\psi}(w,y,t)$ is accurate up to an error of order ϵ^N, as expected.

A by–product of our estimates on the terms stemming from the introduction of the cutoff is that our approximation is exponentially localized in a ball centered at $a(t)$ of any radius b_0, as stated in the second part of Theorem 4.1.

Hence, we have completed the proof of Theorem 4.1. ■

8 Generalizations

As in [15], under some mild supplementary assumptions, we can extend our results to allow $0 \leq t \leq T(\epsilon)$ with $T(\epsilon) \simeq \ln(1/\epsilon^2)$. This proves the validity of our construction up to the Ehrenfest time scale.

Theorem 8.1 *In addition to the assumptions of Theorem 4.1, assume that a classical solution to the equation (2.4) exists for all $t \in \mathbb{R}$. Moreover, assume that for all z in a complex neighborhood of Ξ, the following bound is satisfied*

$$|E(z)| \;\leq\; N \, e^{M|z|},$$

and that $E(x)$ is bounded below. Suppose also that there exist L and $\lambda > 0$, such that for all $t \in \mathbb{R}$

$$\|A(t)\| + \|B(t)\| \;\leq\; L \, e^{\lambda t}.$$

Then, there exist τ', C', $T' > 0$, and $0 < \sigma, \sigma' < 2$ such that the approximation defined by choosing $N(\epsilon) \simeq 1/\epsilon^\sigma$ is accurate up to an error whose norm is bounded by $C' e^{-\tau'/\epsilon^{\sigma'}}$, uniformly for all times $0 \leq t \leq T' \ln(1/\epsilon^2)$.

Proof: It is enough to mimick the proof of the corresponding result for the semiclassical propagation of the Schrödinger equation in [15], since our hypotheses imply that nothing can happen on the adiabatic side of the problem. By the conservation of energy, the exponential bound on $E(z)$ and the assumed existence of a Liapunov exponent, we easily see from the proof of Lemmas 7.2 and 7.3, that the behavior in t of all constants (independent of N) is at worst exponential in t. From the conditions $D_2 \geq \mathrm{e}^{KT}$, with K some constant, we need to take $g(T) \leq g_0 \mathrm{e}^{-g_1 t}$ so that the optimal truncation procedure yields an error of the order $\mathrm{e}^{K_0 T} \mathrm{e}^{-g_0^2 \mathrm{e}^{-2g_1}/\epsilon^2}$. The choice $T(\epsilon) \leq T' \ln(1/\epsilon^2)$, with $T' > 0$ sufficiently small, gives the desired result. ■

Similarly, we can extend our results to allow initial conditions in a wider class of vectors. Indeed, we have been careful to make explicit the J dependence in all estimates so that we can control the error term as a function of J. Recall that J is fixed arbitrarily in (3.8) which gives the expansion in the basis $\varphi_j(A(0), B(0), \epsilon^2, a(0), \eta(0), x)$ of the nuclear part of the wave function that we take as an initial condition.

As in [15], for $(a, \eta) \in I\!\!R^{2d}$, we introduce the operator $\Lambda_\epsilon(a, \eta)$ such that

$$(\Lambda_\epsilon(a, \eta) f)(x) \;=\; \epsilon^{-d}\, \mathrm{e}^{i\eta\cdot(x-a)/\epsilon^2}\, f((x-a)/\epsilon).$$

We define a dense set $\mathcal{C}$ in $L^2(I\!\!R^d)$, that is contained in the set $\mathcal{S}$ of Schwartz functions, by

$$\mathcal{C} \;=\; \left\{ f(x) \;=\; \sum_j c_j\, \varphi_j(I\!\!I, I\!\!I, 1, 0, 0, x) \;\in\; \mathcal{S}, \quad \text{such that} \right.$$
$$\left. \text{there exists } K > 0 \text{ with } \sum_{|j|>J} |c_j|^2 \leq \mathrm{e}^{-KJ}, \text{ for large } J \right\}. \qquad (8.1)$$

Remark It is easy to check that the inequality in (8.1) is equivalent to the requirement that the coefficients of f satisfy

$$|c_j| \;\leq\; \mathrm{e}^{-K|j|},$$

for large $|j|$. Another equivalent definition of $\mathcal{C}$ is

$$\mathcal{C} \;=\; \cup_{t>0}\; \mathrm{e}^{-tH_{ho}}\, \mathcal{S},$$

where $H_{ho} \;=\; -\Delta/2 \,+\, x^2/2$ is the harmonic oscillator Hamiltonian. The set $\mathcal{C}$ is also called the set of analytic vectors [27] for the harmonic oscillator Hamiltonian.

Let $f \in \mathcal{C}$. We set

$$f_J(y,t) \;=\; \sum_{|j|\leq J} c_j\, \varphi_j(A(t), B(t), \epsilon^2, 0, 0, y), \qquad \text{and}$$
$$f(y,t) \;=\; \sum_j c_j\, \varphi_j(A(t), B(t), \epsilon^2, 0, 0, y)$$

where the classical quantities $a(t)$, $\eta(t)$, $A(t)$, $B(t)$, and $S(t)$ correspond to the initial conditions $a(0)$, $\eta(0)$, $A(0) = B(0) = I\!\!I$, and $S(0)$. We consider the construction described in Section 4 corresponding to the initial condition $g_0(0, y, t) \;=\; f_J(y, t)$, making explicit the dependence on J in the notation:

$$\hat{\Psi}_{J,N}(w, y, t)$$
$$=\; F(w)\, \mathrm{e}^{iS(t)/\epsilon^2}\, \mathrm{e}^{i\eta(t)\cdot y/\epsilon} \left(\sum_{n=0}^{N} \epsilon^n\, g_{n,J}(w, y, t)\, \Phi(w, t) \;+\; \sum_{n=2}^{N+2} \epsilon^n\, \phi_{n,J}^{\perp}(w, y, t) \right).$$

Recall that

$$
\begin{aligned}
& \hat{\Psi}_{J,N}(w,y,0) \\
= \quad & F(w)\, \mathrm{e}^{iS(0)/\epsilon^2}\, \mathrm{e}^{i\eta(0)\cdot y/\epsilon} \left(f_J(y,0)\, \Phi(w,0) \;+\; \sum_{n=2}^{N+2} \epsilon^n\, \phi_{n,J}^{\perp}(w,y,0) \right).
\end{aligned}
$$

Let $\nu > 0$, and consider $N(\epsilon) = [\![g^2/\epsilon^2]\!]$ and $J(\epsilon) = \nu N(\epsilon)$. We define our more general initial conditions as

$$
\begin{aligned}
& \hat{\Psi}_f(w,y,0) \\
= \quad & F(w)\, \mathrm{e}^{iS(0)/\epsilon^2}\, \mathrm{e}^{i\eta(0)\cdot y/\epsilon} \left(f(y,0)\, \Phi(w,0) \;+\; \sum_{n=2}^{N(\epsilon)+2} \epsilon^n\, \phi_{n,J(\epsilon)}^{\perp}(w,y,0) \right),
\end{aligned}
$$

which corresponds, when we get back to the variables (X,t), to an initial state $\hat{\Psi}_f(X - a(0), (X - a(0))/\epsilon, 0)$ whose projection along the electronic eigenvector $\tilde{\Phi}(X,0)$ yields a nuclear wave packet of the form $(\Lambda_\epsilon(a(0),\eta(0))f)(X)$. Note that the component of the initial state perpendicular to $\tilde{\Phi}(X,0)$ necessary to achieve exponential accuracy depends on ϵ. This component is determined by the coefficients of the function f.

We can now state our result for such general initial conditions

Theorem 8.2 *Assume the hypotheses of Theorem 4.1 and consider the above constructions. There exist sufficiently small $g > 0$ and positive constants $C(g)$, $\Gamma(g)$, such that with the definition*

$$\Psi_*(X,t,\epsilon) = \hat{\Psi}_{J(\epsilon),N(\epsilon)}(X - a(t), (X - a(t))/\epsilon, t),$$

we have

$$\left\| \, e^{-itH(\epsilon)/\epsilon^2}\, \Psi_f(X,0,\epsilon) \;-\; \Psi_*(X,t,\epsilon) \, \right\|_{L^2(\mathbb{R}^d, \mathcal{H}_{el})} \;\leq\; C(g)\, e^{-\Gamma(g)/\epsilon^2},$$

for all $t \in [0,T]$, as $\epsilon \to 0$.
Moreover, the result for times $T \simeq \ln(1/\epsilon^2)$ corresponding to Theorem 8.1 is also true for these initial conditions.

Proof: We have

$$
\begin{aligned}
& \mathrm{e}^{-itH(\epsilon)/\epsilon^2}\, \Psi_f(X,0,\epsilon) \\
= \quad & \mathrm{e}^{-itH(\epsilon)/\epsilon^2}\, (\Psi_f(X,0,\epsilon) - \Psi_*(X,0,\epsilon)) \;+\; \mathrm{e}^{-itH(\epsilon)/\epsilon^2}\, \Psi_*(X,0,\epsilon) \\
= \quad & \Psi_*(X,t,\epsilon) \;+\; O(\|\mathrm{e}^{-itH(\epsilon)/\epsilon^2}\, \Psi_*(X,0,\epsilon) - \Psi_*(X,t,\epsilon)\|_{L^2(\mathbb{R}^d,\mathcal{H}_{\mathrm{el}})}) \\
+ \quad & O\left(\|\Psi_f(X,0,\epsilon) - \Psi_*(X,0,\epsilon)\|_{L^2(\mathbb{R}^d,\mathcal{H}_{\mathrm{el}})} \right).
\end{aligned}
$$

By our choice of function f, the last term is exponentially small in $1/\epsilon^2$. The remaining norm to estimate corresponds to the situation of Theorem 4.1 in which we let the parameter J grow as $1/\epsilon^2$, according to our choice of $J(\epsilon)$. But, as in the proof of Theorem 3.6 in [15] for the corresponding result in semiclassical dynamics, we have made the dependence in J of all the key estimates explicit. It is enough to go through the proof of theorem 4.1 to check that with $J = \nu N$, all arguments can be repeated to get the same N and ϵ behavior for the estimates on the error terms, (see [15] for details). Hence, we see that for sufficiently small g, we can approximate the solution corresponding to these generalized initial conditions up to an error of order $\mathrm{e}^{-\Gamma(g)/\epsilon^2}$. The Ehrenfest time regime is dealt with similarly. ■

9 Technicalities

In this section we give the proofs of the auxiliary lemmas we used in the course of the main argument.

Proof of Lemma 6.1: We first consider the case $k \geq 1$. By Cauchy's formula, we can write

$$g'(t) \;=\; \frac{1}{2\pi i}\int_\Gamma \frac{g(s)}{(t-s)^2}\,ds, \tag{9.2}$$

where Γ is the circular contour with center t and radius $\dfrac{1}{k+1}\,(\delta\,-\,|\mathrm{Im}\,t|)$.

For s on Γ, we have $(\delta - |\mathrm{Im}\,s|) \,\geq\, \dfrac{k}{k+1}(\delta\,-\,|\mathrm{Im}\,t|)$. Thus,

$$\|g(s)\| \;\leq\; C\,k^k\,(\delta\,-\,|\mathrm{Im}\,s|)^{-k} \;\leq\; C\,k^k\,\left[\frac{k}{k+1}(\delta\,-\,|\mathrm{Im}\,t|)\right]^{-k}$$

So, by putting the norm inside the integral in (9.2), we have

$$\begin{aligned}\|g'(t)\| \;&\leq\; \frac{1}{2\pi}\,\frac{2\pi}{k+1}(\delta\,-\,|\mathrm{Im}\,t|)\;C\,k^k\left[\frac{k}{k+1}(\delta\,-\,|\mathrm{Im}\,t|)\right]^{-k}\;\left[\frac{1}{k+1}(\delta\,-\,|\mathrm{Im}\,t|)\right]^{-2}\\ &=\; C\,(k+1)^{k+1}\,(\delta\,-\,|\mathrm{Im}\,t|)^{-k-1}.\end{aligned}$$

For $k = 0$ we use the same argument with the radius of Γ replaced by $\alpha\,(\delta\,-\,|\mathrm{Im}\,t|)$ for any $\alpha < 1$. This yields the bound

$$\|g'(t)\| \;\leq\; C\,\alpha^{-1}\,(\delta\,-\,|\mathrm{Im}\,t|)^{-1}.$$

The lemma follows because $\alpha < 1$ is arbitrary. ■

Proof of Lemma 6.4: To prove the quantity ν is finite, we estimate

$$\sum_{\{\,l\,:\,0\leq l_i\leq\alpha_i\,\}}\frac{1}{(1+|l|)^{d+1}}\,\frac{1}{(1+|\alpha-l|)^{d+1}}$$

$$=\;\sum_{\substack{\{\,l\;:\;0\,\leq\,l_i\,\leq\,\alpha_i\,\}\\ |l|\,\leq\,[\![\frac{|\alpha|}{2}]\!]}}\frac{1}{(1+|l|)^{d+1}}\,\frac{1}{(1+|\alpha-l|)^{d+1}}$$

$$+ \sum_{\substack{\{\, l \,:\, 0 \le l_i \le \alpha_i \,\} \\ |l| > [\![\frac{|\alpha|}{2}]\!]}} \frac{1}{(1+|l|)^{d+1}} \, \frac{1}{(1+|\alpha-l|)^{d+1}}$$

$$\le \frac{2}{\left(1 + [\![\frac{|\alpha|}{2}]\!]\right)^{d+1}} \sum_{\substack{\{\, l \,:\, 0 \le l_i \le \alpha_i \,\} \\ |l| \le [\![\frac{|\alpha|}{2}]\!]}} \frac{1}{(1+|l|)^{d+1}}$$

$$\le \frac{2^{d+2}}{(1+|\alpha|\,)^{d+1}} \sum_{\substack{\{\, l \,:\, 0 \le l_i \le \alpha_i \,\} \\ |l| \le [\![\frac{|\alpha|}{2}]\!]}} \frac{1}{(1+|l|)^{d+1}}$$

$$\le \frac{2^{d+2}}{(1+|\alpha|\,)^{d+1}} \sum_{l} \frac{1}{(1+|l|)^{d+1}}.$$

Thus, $\quad \nu \;\le\; 2^{d+2} \sum_{l} (1+|l|)^{-d-1}.$

To see that the right hand side of this inequality is finite, we note that the number of multi-indices l with $|l| = L$ is the binomial coefficient $\begin{pmatrix} L+d-1 \\ d-1 \end{pmatrix}$, with the convention that $\begin{pmatrix} 0 \\ 0 \end{pmatrix} \;=\; 1$. Thus,

$$\nu \;\le\; 2^{d+2} \sum_{L=0}^{\infty} \begin{pmatrix} L+d-1 \\ d-1 \end{pmatrix} \frac{1}{(1+L)^{d+1}}$$

$$= \frac{2^{d+2}}{(d-1)!} \sum_{L=0}^{\infty} \frac{(L+d-1)(L+d-2)\cdots(L+1)}{(L+1)^{d+1}}.$$

For large L, $\quad \dfrac{(L+d-1)(L+d-2)\cdots(L+1)}{(L+1)^{d+1}}$ is asymptotic to L^{-2}, so ν is finite.

Since $D^{\alpha}\,(M\,N) \;=\; \sum_{\{\, l \,:\, 0 \le l_i \le \alpha_i \,\}} \left[\prod_{j=1}^{d} \begin{pmatrix} \alpha_j \\ l_j \end{pmatrix} \right] \left(D^{l}\,M\right) \left(D^{(\alpha-l)}\,N\right)$, we have

$$\|\,(\,D^{\alpha}\,(M\,N)\,)\,(x)\,\|$$

$$\le \sum_{\{\, l \,:\, 0 \le l_i \le \alpha_i \,\}} \left[\prod_{j=1}^{d} \begin{pmatrix} \alpha_j \\ l_j \end{pmatrix} \right] m(x)\; n(x)\; a(x)^{|\alpha+p+q|} \quad \frac{(l+p)!}{(1+|l|)^{d+1}} \, \frac{(\alpha-l+q)!}{(1+|\alpha-l|)^{d+1}}$$

$$= m(x)\; n(x)\; a(x)^{|\alpha+p+q|} \quad (\alpha+p+q)!$$

$$\times \sum_{\{\,l\,:\,0\le l_i\le\alpha_i\,\}} \left[\prod_{j=1}^{d} \begin{pmatrix}\alpha_j\\ l_j\end{pmatrix} \begin{pmatrix}\alpha_j+p_j+q_j\\ l_j+p_j\end{pmatrix}^{-1}\right] \frac{1}{(1+|l|)^{d+1}\,(1+|\alpha-l|)^{d+1}}.$$

Since $\begin{pmatrix}\alpha_j+p_j+q_j\\ l_j+p_j\end{pmatrix} \ge \begin{pmatrix}\alpha_j+q_j\\ l_j\end{pmatrix} \ge \begin{pmatrix}\alpha_j\\ l_j\end{pmatrix}$, we therefore have

$$\begin{aligned}\|\,(\,D^\alpha\,(M\,N)\,)\,(x)\,\| \;&\le\; m(x)\;n(x)\;a(x)^{|\alpha+p+q|}\quad(\alpha+p+q)!\\ &\qquad\times \sum_{\{\,l\,:\,0\le l_i\le\alpha_i\,\}} \frac{1}{(1+|l|)^{d+1}\,(1+|\alpha-l|)^{d+1}}.\\ &\le\; m(x)\;n(x)\;\nu\;a(x)^{|\alpha+p+q|}\quad\frac{(\alpha+p+q)!}{(1+|\alpha|)^{d+1}}.\end{aligned}$$ ∎

Proof of Lemma 6.3: If $f(t)$ satisfies $\|f(t)\| \le C\,|t|^p\,\mathrm{dist}(t)^{-k}$, for all $t\in\Omega$, there exists $g(t)$ analytic in Ω, such that $f(t)=t^p\,g(t)$ and $\|g(t)\|\le C\,\mathrm{dist}(t)^{-k}$. We use the integration path from 0 to $t\in\Omega$ parametrized by $\gamma(u)=tu$, with $u\in[0,1]$, to compute

$$\begin{aligned}\left\|\int_0^t f(s)\,ds\right\| \;&=\; \left\|\int_0^1 f(tu)\,du\right\| \;=\; \left\|\int_0^1 t\,(tu)^p\,g(tu)\,du\right\|\\ \le\; C\,|t|^{p+1}\int_0^1 \frac{u^p}{\mathrm{dist}(tu)^k}\,du \;&\le\; C\,\frac{|t|^{p+1}}{p+1}\,\mathrm{dist}(t)^{-k},\end{aligned} \tag{9.3}$$

since, by assumption, $\mathrm{dist}(ut)$ is a decreasing function of u. ∎

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