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# SEPARATION OF SCALES: A QUANTUM-CLASSICAL APPROACH FOR COMPLEX SYSTEMS AND A SYSTEM-BATH ANSATZ

IRENE BURGHARDT, RÉMI CARLES, CLOTILDE FERMANIAN KAMMERER,  
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ABSTRACT. We consider quantum-dynamical systems that consist of two different parts as in system-bath Hamiltonians, and assume that each of these parts is described by a set of parameters. We investigate régimes of intersystem couplings that are partially flat because they vary slowly with respect to one set of variables (those of the bath, for example). We also study the situation where one of the sets of variables is semiclassically scaled; this case effectively leads to a quantum-classical formulation, due to the classical parameter evolution. In both situations, we propose two kinds of schemes of dimension reduction : one based on the Taylor expansion and the other one based on partial averaging. We analyze the error estimates in terms of appropriate norms of the coupling both for the wave function and for the action of observables, showing that the choice between an approach by Taylor expansion or by average is more a question of numerical convenience than a matter of errors, since both approaches lead to comparable estimates.

## 1. INTRODUCTION

We consider quantum-dynamical systems that consist of two different parts and investigate various régimes of intersystem couplings as well as effective dynamical descriptions, that simplify the original, fully quantum-mechanical formulation. The time-dependent Schrödinger equation

$$(1.1) \quad i\partial_t\psi = H\psi \quad ; \quad \psi|_{t=0} = \psi_0,$$

is governed by a Hamiltonian of the form

$$(1.2) \quad H = H_x + H_y + W(x, y), \text{ where} \\ H_x = -\frac{1}{2}\Delta_x + V_1(x), \quad H_y = -\frac{1}{2}\Delta_y + V_2(y),$$

where the potentials  $V_1(x)$ ,  $V_2(y)$  and the coupling potential  $W(x, y)$  are all smooth functions, that satisfy subquadratic estimates. Such a set-up guarantees existence and uniqueness of the solution to the Schrödinger equation (1.1) for a rather general set of initial data, see Assumption 2.1 for precise statements of our assumptions, as well as Remark 2.2 for other possible frameworks. The overall set of space variables is denoted as  $(x, y) \in \mathbb{R}^n \times \mathbb{R}^d$  such that the total dimension of the configuration space is  $n + d$ . The wave function depends on time  $t \geq 0$  and both space variables, that is,  $\psi = \psi(t, x, y)$ . We will abbreviate the Lebesgue spaces for the different variables  $x$ ,  $y$ , and  $(x, y)$  by

$$L_x^2 = L^2(\mathbb{R}^n) \quad , \quad L_y^2 = L^2(\mathbb{R}^d) \quad , \quad L^2 = L^2(\mathbb{R}^{n+d}).$$

We suppose that initially scales are separable, that is, we work with initial data of product form

$$(1.3) \quad \psi(0, x, y) = \psi_0(x, y) = \varphi_0^x(x)\varphi_0^y(y),$$

where the functions  $\varphi_0^x \in L_x^2$  and  $\varphi_0^y \in L_y^2$  are square-integrable and satisfy additional growth and regularity assumptions as given in Section 2.1 (typically, Schwartz class). In the simple case without coupling, that is,  $W \equiv 0$ , the solution stays separated,  $\psi(t, x, y) = \varphi^x(t, x)\varphi^y(t, y)$  for all time, where

$$\begin{aligned} i\partial_t \varphi^x &= H_x \varphi^x & ; & \quad \varphi^x|_{t=0} = \varphi_0^x, \\ i\partial_t \varphi^y &= H_y \varphi^y & ; & \quad \varphi^y|_{t=0} = \varphi_0^y, \end{aligned}$$

and this is an exact formula. Here, we aim at investigating the case of an actual coupling with  $\partial_x \partial_y W \neq 0$  and look for approximate solutions of the form

$$\psi_{\text{app}}(t, x, y) = \varphi^x(t, x)\varphi^y(t, y),$$

where the individual components satisfy evolution equations that account for the coupling between the variables. We investigate the size of the difference between the true and the approximate solution in the  $L^2$ -norm

$$\|\psi(t) - \psi_{\text{app}}(t)\|_{L^2},$$

and in Sobolev norms. We present error estimates that explicitly depend on derivatives of the coupling potential  $W(x, y)$  and on moments of the approximate solution. As an additional error measure we also consider the deviation of true and approximate expectation values

$$\langle \psi(t), A\psi(t) \rangle - \langle \psi_{\text{app}}(t), A\psi_{\text{app}}(t) \rangle,$$

for self-adjoint linear operators  $A$ . Roughly speaking, the estimates we obtain for observables depend on one more derivative of the coupling potential than the norm estimates. This means that in many situations expectation values are more accurately described than the wave function itself.

**1.1. Partially flat coupling.** In the first part of our paper we will investigate the accuracy, when the full Hamiltonian  $H$  is replaced by an approximate one,  $H_{\text{app}} = H_x + H_y + W_{\text{app}}(x, y)$ , that keeps the factored form of the wave function. We consider a brute-force approach, where we collocate partially at the origin and set

$$W_{\text{app}}(x, y) = W(x, 0) + W(0, y) - W(0, 0).$$

A possible criterion for choosing a collocation point different from the origin is presented in Remark 3.1. In comparison, following the more conventional time-dependent Hartree approach, we perform partial averages of the coupling potential,

$$\begin{aligned} \langle W \rangle_y &= \int_{\mathbb{R}^d} W(x, y) |\varphi^y(t, y)|^2 dy / \int_{\mathbb{R}^d} |\varphi^y(t, y)|^2 dy, \\ \langle W \rangle_x &= \int_{\mathbb{R}^n} W(x, y) |\varphi^x(t, x)|^2 dx / \int_{\mathbb{R}^n} |\varphi^x(t, x)|^2 dx, \end{aligned}$$

and the full average

$$\langle W \rangle = \int_{\mathbb{R}^{n+d}} W(x, y) |\varphi^x(t, x)\varphi^y(t, y)|^2 dx dy / \int_{\mathbb{R}^{n+d}} |\varphi^x(t, x)\varphi^y(t, y)|^2 dx dy.$$

We then set

$$W_{\text{app}}(t, x, y) = \langle W \rangle_y(t, x) + \langle W \rangle_x(t, y) - \langle W \rangle(t).$$

This approach will be referred to as mean-field approximation in the following.

For both approximations, the brute-force and the mean-field approximation, we derive various types of estimates for the error in  $L^2$ -norm. Our key finding is that both methods come with error bounds that are qualitatively the same, since they draw from either evaluations or averages of the function

$$\delta W(x, x', y, y') = W(x, y) - W(x, y') - W(x', y) + W(x', y').$$

Depending on whether one chooses to control the auxiliary function  $\delta W$  in terms of  $\nabla_x W$ ,  $\nabla_y W$  or  $\nabla_x \nabla_y W$ , the estimate requires a balancing with corresponding moments of the approximate solution, see Proposition 3.4. The extension of the  $L^2$ -results to first order Sobolev norms is given in Proposition 3.8. In addition, we analyse the deviation of the true and the approximate expectation values in a similar vein. For the expectation values, we again obtain qualitatively similar error estimates for both methods. The upper bounds differ from the norm bounds in so far as they involve one more derivative of the coupling potential  $W$  and low order Sobolev norms of the approximate solution, see Proposition 3.10.

We may therefore summarize that from the perspective of approximation accuracy, the brute force and the mean-field approach differ only slightly. Therefore, other assessment criteria are needed for explaining the prevalence of the Hartree method in many applications. Specifically, the time-dependent Hartree approximation (with its partial averages) stems from the time-dependent Dirac–Frenkel variational principle applied to the manifold of product functions, see for example [23, §3.2], and thus automatically conserves energy with the same energy as the original system. We provide a non-variational proof of this crucial conservation property in Lemma 3.12.

**1.2. Dimension reduction via semiclassical analysis.** In the second part of the paper we turn to a specific case of the previous general class of coupled Hamiltonians  $H^\varepsilon = H_x + H_y^\varepsilon + W(x, y)$  and consider for one part of the system a semiclassically scaled Schrödinger operator

$$H_y^\varepsilon = -\frac{\varepsilon^2}{2}\Delta_y + V_2(y), \quad \varepsilon > 0.$$

The initial data are still a product of the form (1.3), but the  $y$ -factor is chosen as

$$\varphi_0^y(y) = \varepsilon^{-d/4} a \left( \frac{y - q_0}{\sqrt{\varepsilon}} \right) e^{ip_0 \cdot (y - q_0)/\varepsilon},$$

that is,  $\varphi_0^y$  is a semiclassical wave packet with a smooth and rapidly decaying amplitude function  $a \in \mathcal{S}(\mathbb{R}^d)$ , and an arbitrary phase space center  $(q_0, p_0) \in \mathbb{R}^{2d}$ . We will choose a semiclassical wave packet approximation for  $\varphi^y(t, y)$  exploring two different choices for the center  $(q(t), p(t))$ . As a first option we consider the classical trajectory

$$\dot{q} = p \quad , \quad \dot{p} = -\nabla V_2(q),$$

and as a second option the corresponding trajectory resulting from the averaged gradient of the potential  $V_2$ ,

$$\langle \nabla V_2 \rangle (t) = \int_{\mathbb{R}^d} \nabla_y V_2(y) |\varphi^y(t, y)|^2 dy / \int_{\mathbb{R}^d} |\varphi^y(t, y)|^2 dy.$$

Correspondingly, the approximative factor  $\varphi^x(t, x)$  is evolved by the partial Hamiltonian  $H_x + W_{\text{eff}}$  with

$$W_{\text{eff}}(t, x) = W(x, q(t)) \quad \text{or} \\ W_{\text{eff}}(t, x) = \int_{\mathbb{R}^d} W(x, y) |\varphi^y(t, y)|^2 dy / \int_{\mathbb{R}^d} |\varphi^y(t, y)|^2 dy.$$

We obtain error estimates in  $L^2$ -norm, see Proposition 4.4, Sobolev norms, see Remark 4.6, and expectation values, see Proposition 4.9. These estimates are given in terms of the semiclassical parameter  $\varepsilon$  and derivatives of the coupling potential.

**1.3. Relation to earlier work.** The first motivation for our investigation came from the variational treatment of mixed quantum-classical molecular dynamics in [28]. To the best of our knowledge, the rather general mathematical analysis of scale separation in quantum systems we are developing here is new. Previous mathematical work we are aware of is concerned with rather specific coupling models, as for example the coupling of Hartree–Fock and classical equations in [6], or the time-dependent self-consistent field equations in [18], or with adiabatic approximations which rely on eigenfunctions for one part of the system, see for example [32, 26]. In the chemical physics literature, related approaches to dimension reduction have been proposed in the context of mean-field methods [13, 14], and the quantum-classical mean-field Ehrenfest approach [12, 2]. Also, quantum-classical formulations have been derived in a Wigner phase space [25, 19] setting and a quantum hydrodynamic setting [15, 5, 30]. The present formulation circumvents formal difficulties of these approaches [9, 31, 29], by preserving a quantum wavefunction description for the entire system.

**1.4. Outline of the paper.** In the next Section 2, we present the main assumptions we will work with (Section 2.1) and the type of arguments we will use (Section 2.3). We also develop the example of system-bath Hamiltonians with anharmonic couplings that has motivated our study (Section 2.2). Then, in Section 3, we present the reduction of dimension that can be achieved by taking advantage of a flat coupling. We discuss both the brute-force (Section 3.1) and the mean-field approach (Section 3.2), and give error estimates, for the wave function (Section 3.3) and for observables (Section 3.5), paying special attention to initial data presenting higher Sobolev regularity (Section 3.4), and to energy conservation (Section 3.6). Finally, in Section 4, we consider initial data which fit with a semiclassical setting and present the semiclassical approach that we motivate by a scaling procedure (Section 4.1). We introduce two types of ansatz (Section 4.2), one obtained by Taylor expansion (Section 4.3) and the other one by partial averaging (Section 4.4), the convergence of which we study in both cases for the wave function and for observables that only act on the semiclassical variable. These results are stated in Section 4.5 and proved in Sections 4.6 and 4.7, respectively.

## 2. ASSUMPTIONS AND TECHNICAL PRELIMINARIES

We describe here the mathematical setting that will be ours, discuss it in the context of system-bath Hamiltonians [34, 3], and provide the main technical lemma that we will use for deriving precise error estimates.

**2.1. Assumptions on regularity and growth of the potentials.** The regularity and the growth of the potential is an important feature of the subject. We choose a very classical set of assumptions since our focus is more on finding appropriate ways to approximate the solution in a standard framework than on treating specific situations.

**Assumption 2.1.** *All the potentials that we consider are smooth, real-valued, and at most quadratic in their variables:*

$$V_1 \in C^\infty(\mathbb{R}^n; \mathbb{R}), \quad V_2 \in C^\infty(\mathbb{R}^d; \mathbb{R}), \quad W \in C^\infty(\mathbb{R}^{n+d}; \mathbb{R}),$$

and, for  $\alpha \in \mathbb{N}^n$ ,  $\beta \in \mathbb{N}^d$ ,

$$\partial_x^\alpha V_1 \in L^\infty \text{ provided that } |\alpha| \geq 2,$$

$$\partial_y^\beta V_2 \in L^\infty \text{ provided that } |\beta| \geq 2,$$

$$\partial_x^\alpha \partial_y^\beta W \in L^\infty \text{ provided that } |\alpha| + |\beta| \geq 2.$$

All the initial data we consider are smooth and rapidly decaying, that is, Schwartz class functions:

$$\varphi_0^x \in \mathcal{S}(\mathbb{R}^n; \mathbb{C}), \quad \varphi_0^y \in \mathcal{S}(\mathbb{R}^d; \mathbb{C}) \quad (\text{hence } \psi_0 \in \mathcal{S}(\mathbb{R}^{n+d}; \mathbb{C})).$$

Under the above assumption, it is well-known that  $H_x$ ,  $H_y$  and  $H$  are essentially self-adjoint on  $L^2(\mathbb{R}^N)$ , with  $N = n, d$  and  $n + d$ , respectively (as a consequence of Faris-Lavine Theorem, see e.g. [27, Theorem X.38]).

The assumptions on the growth and smoothness of the potentials and the regularity of the initial data call for comments.

- Remark 2.2.*
- (1) Concerning the growth of  $V_1$ ,  $V_2$  and  $W$ , the assumption that they are at most quadratic concerns the behavior at infinity and could be relaxed, up to suitable sign assumptions. Local behavior is rather free, for example a local double well is allowed, as long as it is not too confining at infinity. We choose to stick to the at most quadratic case, since bounded second order derivatives simplify the presentation.
  - (2) Concerning the smoothness, most of our results still hold assuming only smoothness of  $W$ , as long as the operators  $H_x$  and  $H_y$  are essentially self-adjoint on an adequate domain included in  $L^2(\mathbb{R}^N)$ , with  $N = n, d$ . For example,  $V_1$  and  $V_2$  could both present Coulomb singularities, and the results of Proposition 3.4 would still hold. In the semiclassical régime, we can also allow a Coulomb singularity for  $V_1$  and prove Proposition 4.4 and Proposition 4.9.
  - (3) Concerning the smoothness and the decay of the initial data, most of our results still hold, if the initial data are contained in one of the spaces  $\Sigma^k(\mathbb{R}^N)$  containing functions  $f$  whose norms

$$(2.1) \quad \|f\|_{\Sigma^k} = \sup_{\substack{z \in \mathbb{R}^N \\ |\alpha| + |\beta| \leq k}} \|z^\alpha \partial_z^\beta f\|_{L^2}$$

are bounded. Note that  $\mathcal{S}(\mathbb{R}^N) = \cap_{k \in \mathbb{N}} \Sigma^k$ . For example, Proposition 3.4 still holds for initial data in  $\Sigma^1$ , while Proposition 4.4 requires initial data in a semiclassically scaled  $\Sigma^3$  space.

**2.2. System-bath Hamiltonians.** An important class of coupled quantum systems are described by system-bath Hamiltonians [34, 3].

$$H_{\text{sb}} = -\frac{1}{2}\Delta_x - \frac{1}{2}\Delta_y + V_s(x) + V_b(y) + V_{\text{sb}}(x, y)$$

These are naturally given in the format required by (1.2). In the present discussion, we specify that the bath is described by a harmonic oscillator,  $V_b(y) = \frac{1}{2}k_2^0|y|^2$  (or a set of harmonic oscillators in more than one dimension) and the system-bath coupling  $V_{\text{sb}}(x, y) = W(x, y)$  is of cubic form, such that we obtain in the notation of (1.2),

$$H_x = -\frac{1}{2}\Delta_x + V_s(x) \quad , \quad H_y = -\frac{1}{2}\Delta_y + \frac{1}{2}k_2^0|y|^2 \quad , \quad W(x, y) = \frac{1}{2}\vec{\eta} \cdot x|y|^2.$$

where  $k_2^0 > 0$  and  $\vec{\eta} \in \mathbb{R}^d$ . The cubic, anharmonic coupling  $W(x, y)$  is a non-trivial case, which is employed, e.g., in the description of vibrational dephasing [22, 16] and Fermi resonances [4]. It is natural to assume smoothness and subquadratic growth for  $V_s(x)$ . However, the coupling potential  $W(x, y)$  clearly fails to satisfy the growth condition of Assumption 2.1. In the following, we will therefore also provide slight extensions of our estimates to accomodate this specific, but interesting type of coupling.

**2.3. Estimation lemma.** We shall consider two families of estimates, regarding the wave function itself on the one hand, and regarding observables on the other hand. Of course, error estimates on the wave function yield error estimates of observables, since

$$\begin{aligned} & |\langle \psi(t), A\psi(t) \rangle - \langle \psi_{\text{app}}(t), A\psi_{\text{app}}(t) \rangle| \\ & \leq \| \psi(t) - \psi_{\text{app}}(t) \|_{L^2} (\| A\psi(t) \|_{L^2} + \| A\psi_{\text{app}}(t) \|_{L^2}). \end{aligned}$$

However, it turns out that the averaging process, involved in the action of an observable on a wave function, allows to prove better estimates than those stemming from the norm estimates. Our first tool will be the following standard lemma that will be applied with either  $h = 1$  or  $h = \varepsilon$  a small parameter.

**Lemma 2.3.** *Let  $N \geq 1$ ,  $A$  be self-adjoint on  $L^2(\mathbb{R}^N)$ , and  $\psi$  solution to the Cauchy problem*

$$ih\partial_t\psi = A\psi + \Sigma \quad ; \quad \psi|_{t=0} = \psi_0,$$

where  $\psi_0 \in L^2(\mathbb{R}^N)$  and  $\Sigma \in L^1_{\text{loc}}(\mathbb{R}^+; L^2(\mathbb{R}^N))$ . Then for all  $t \geq 0$ ,

$$\| \psi(t) \|_{L^2(\mathbb{R}^N)} \leq \| \psi_0 \|_{L^2(\mathbb{R}^N)} + \frac{1}{h} \int_0^t \| \Sigma(s) \|_{L^2(\mathbb{R}^N)} ds.$$

*Proof.* In view of the self-adjointness of  $A$ , we have

$$\begin{aligned} \| \psi(t) \| \frac{d}{dt} \| \psi(t) \| &= \frac{1}{2} \frac{d}{dt} \langle \psi(t), \psi(t) \rangle = \text{Re} \langle \psi(t), \frac{1}{ih} (A\psi(t) + \Sigma(t)) \rangle \\ &= \frac{1}{h} \text{Im} \langle \psi(t), \Sigma(t) \rangle, \end{aligned}$$

and therefore, by the Cauchy–Schwarz inequality,

$$\frac{d}{dt}\|\psi(t)\| \leq \frac{1}{h}\|\Sigma(t)\|.$$

Integrating in time, we obtain

$$\|\psi(t)\| = \|\psi_0\| + \int_0^t \frac{d}{ds}\|\psi(s)\| ds \leq \|\psi_0\| + \frac{1}{h} \int_0^t \|\Sigma(s)\| ds.$$

□

In the context of observables, refined error estimates will follow from the application of the following lemma.

**Lemma 2.4.** *Let  $N \geq 1$ ,  $A_1, A_2, B$  be self-adjoint on  $L^2(\mathbb{R}^N)$ , and  $\psi^{(1)}, \psi^{(2)}$ , solutions to the homogeneous Cauchy problems*

$$ih\partial_t\psi^{(j)} = A_j\psi^{(j)} \quad ; \quad \psi|_{t=0} = \psi_0,$$

where  $\psi_0 \in L^2(\mathbb{R}^N)$ . Then, for all  $t \geq 0$

$$\left| \left\langle \psi^{(1)}(t), B\psi^{(1)}(t) \right\rangle - \left\langle \psi^{(2)}(t), B\psi^{(2)}(t) \right\rangle \right| \leq \frac{1}{h} \int_0^t |\rho(s, t)| ds,$$

with

$$\rho(s, t) = \left\langle \psi^{(1)}(s), [\exp(iA_2(t-s)/h)B \exp(-iA_2(t-s)/h), A_1 - A_2] \psi^{(1)}(s) \right\rangle.$$

*Proof.* We denote the unitary evolution operators by  $U_j(t) = \exp(-iA_j t/h)$  and calculate

$$\begin{aligned} & \left\langle \psi^{(1)}(t), B\psi^{(1)}(t) \right\rangle - \left\langle \psi^{(2)}(t), B\psi^{(2)}(t) \right\rangle \\ &= \langle U_1(t)\psi_0, BU_1(t)\psi_0 \rangle - \langle U_2(t)\psi_0, BU_2(t)\psi_0 \rangle \\ &= \int_0^t \frac{d}{ds} \langle \psi_0, U_1(s)^* U_2(t-s)^* BU_2(t-s) U_1(s) \psi_0 \rangle ds \\ &= \frac{1}{ih} \int_0^t \langle \psi_0, U_1(s)^* [U_2(t-s)^* BU_2(t-s), A_1 - A_2] U_1(s) \psi_0 \rangle ds \\ &= \frac{1}{ih} \int_0^t \left\langle \psi^{(1)}(s), [U_2(t-s)^* BU_2(t-s), A_1 - A_2] \psi^{(1)}(s) \right\rangle ds. \end{aligned}$$

□

### 3. PARTIALLY FLAT COUPLING

In this section, we do not assume special features in the scaling of the equation, except that  $W$  is flat in, say  $y$ . This flatness can be expressed mathematically as the smallness of  $\|\nabla_y W\|_{L^\infty}$  (or of  $\|\nabla_x \nabla_y W\|_{L^\infty}$ ). Depending on the regularity of the data that is considered, it could be relaxed on the smallness of  $\|\langle y \rangle^{-p} \nabla_y W\|_{L^\infty}$  for some  $p > 0$  (see Remark 3.6). We also emphasize that strictly speaking,  $\nabla_y W$  need not be bounded under Assumption 2.1. We present the ideas and error estimates under the assumption that  $\nabla_y W$  and its derivatives are bounded (and small), and explain in Remark 3.6 how to adapt the results when  $\nabla_y W$  is unbounded.

If  $\nabla_y W$  is small, we show that the coupling in  $(x, y)$  is negligible at leading order. We present two strategies to approximate the exact solution: brute-force approach, based on Taylor expansion, and the more standard approach, based on spatial averages. In each case, we provide an estimate measuring the error generated by the approximation. Throughout this section,  $\psi = \psi(t, x, y)$  denotes the solution to the initial value problem (1.1)–(1.3).

**3.1. Brute-force approach.** Set  $\tilde{\psi}_{\text{app}}(t, x, y) = \varphi^x(t, x)\varphi^y(t, y)$ , where

$$(3.1) \quad \begin{cases} i\partial_t \varphi^x = H_x \varphi^x + W(x, 0)\varphi^x & ; \quad \varphi^x|_{t=0} = \varphi_0^x, \\ i\partial_t \varphi^y = H_y \varphi^y + W(0, y)\varphi^y & ; \quad \varphi^y|_{t=0} = \varphi_0^y. \end{cases}$$

In view of Assumption 2.1, these equations have unique solutions  $\varphi^x \in C(\mathbb{R}; L_x^2)$ ,  $\varphi^y \in C(\mathbb{R}; L_y^2)$ , and higher regularity is propagated,  $\varphi^x \in C(\mathbb{R}; \Sigma_x^k)$ ,  $\varphi^y \in C(\mathbb{R}; \Sigma_y^k)$ , for all  $k \in \mathbb{N}$ , where we recall that  $\Sigma^k$  has been defined in (2.1). The approximate solution solves

$$i\partial_t \tilde{\psi}_{\text{app}} = H \tilde{\psi}_{\text{app}} + (-W(x, y) + W(x, 0) + W(0, y)) \tilde{\psi}_{\text{app}}.$$

This is not the right approximation: if  $W$  varies very little in  $y$ ,

$$\begin{aligned} W(x, y) - W(x, 0) - W(0, y) &\approx W(x, 0) - W(x, 0) - W(0, y) \\ &= -W(0, y) \approx -W(0, 0). \end{aligned}$$

This term is removed by considering instead  $\psi_{\text{app}} = \tilde{\psi}_{\text{app}} e^{itW(0,0)}$ . It solves

$$i\partial_t \psi_{\text{app}} = H \psi_{\text{app}} - \underbrace{(W(x, y) - W(x, 0) - W(0, y) + W(0, 0))}_{=: \Sigma_\psi} \psi_{\text{app}}.$$

The last term controls the error  $\psi - \psi_{\text{app}}$ , as we will see more precisely below. Saying that the coupling potential  $W$  is flat in  $y$  means that  $\nabla_y W$  is small, and we write

$$W(x, y) - W(x, 0) - W(0, y) + W(0, 0) = \underbrace{W(x, y) - W(x, 0)}_{\approx y \cdot \nabla_y W(x, 0)} - \underbrace{(W(0, y) - W(0, 0))}_{\approx y \cdot \nabla_y W(0, 0)}.$$

In the case where  $W$  is flat both in  $x$  and in  $y$ , it might be useful to write

$$\begin{aligned} &W(x, y) - W(x, 0) - W(0, y) + W(0, 0) \\ &= y \cdot \int_0^1 (\partial_y W(x, \eta y) - \partial_y W(0, \eta y)) d\eta = \int_0^1 \int_0^1 y \cdot \partial_x \partial_y W(\theta x, \eta y) x \, d\eta d\theta. \end{aligned}$$

*Remark 3.1.* For choosing another collocation point than the origin, one might use the matrix

$$M(x, y) = \partial_x \partial_y W(x, y) = (\partial_{x_j} \partial_{y_k} W(x, y))_{1 \leq j \leq n, 1 \leq k \leq d}.$$

We have for  $(x, y) \approx (0, 0)$ ,

$$W(x, y) - W(x, 0) - W(0, y) + W(0, 0) \approx (x \cdot M(0, 0)y)$$

corresponding to the standard normal mode expansion. Adopting polar decomposition, it is possible to write  $M(0, 0) = OQ$  with  $O$  a  $n \times d$  matrix of maximal rank

and  $Q = \sqrt{M(0,0)^t M(0,0)}$  a  $d \times d$  non-negative and symmetric. Then, in a basis where the matrix  $Q$  is diagonal, with diagonal elements  $\lambda_i \geq 0$ , we have

$$\begin{aligned} & W(x, y) - W(x, 0) - W(0, y) + W(0, 0) \\ & \approx \sum_j \lambda_j ({}^t O x)_j y_j = \sum_j \frac{\lambda_j}{4} \left( (({}^t O x)_j + y_j)^2 - (({}^t O x)_j - y_j)^2 \right). \end{aligned}$$

One then sees that it can be valuable to choose the collocation point as a point where the largest singular value of  $M(x_0, y_0)$  is minimal, that is, the quantity

$$(x_0, y_0) \mapsto \max_{|z|=1} \sqrt{z \cdot M(x_0, y_0)^t M(x_0, y_0) z},$$

attains a minimum.

**3.2. Mean-field approach.** Instead of pointwise evaluations of the coupling potential, we might also use partial averages for an approximation. We consider

$$(3.2) \quad \begin{aligned} i\partial_t \phi^x &= H_x \phi^x + \langle W \rangle_y(t) \phi^x & ; & \quad \phi^x|_{t=0} = \varphi_0^x, \\ i\partial_t \phi^y &= H_y \phi^y + \langle W \rangle_x(t) \phi^y & ; & \quad \phi^y|_{t=0} = \varphi_0^y, \end{aligned}$$

where we have denoted

$$\begin{aligned} \langle W \rangle_y &= \langle W \rangle_y(t, x) = \frac{\int W(x, y) |\phi^y(t, y)|^2 dy}{\int |\phi^y(t, y)|^2 dy} = \frac{\int W(x, y) |\phi^y(t, y)|^2 dy}{\int |\varphi_0^y(y)|^2 dy}, \\ \langle W \rangle_x &= \langle W \rangle_x(t, y) = \frac{\int W(x, y) |\phi^x(t, x)|^2 dx}{\int |\phi^x(t, x)|^2 dx} = \frac{\int W(x, y) |\phi^x(t, x)|^2 dx}{\int |\varphi_0^x(x)|^2 dx}, \end{aligned}$$

where we have used the fact that the  $L^2$ -norms of  $\phi^x$  and  $\phi^y$  are independent of time, since  $W$  is real-valued. Note that (3.2) is a nonlinear system of equations. Contrary to the brute-force approach,  $L^2$  regularity does not suffice to define partial averages in general. In view of Assumption 2.1, a fixed point argument (very similar to the proof of e.g. [7, Lemma 13.10]) shows that this system has a unique solution  $(\phi^x, \phi^y) \in C(\mathbb{R}; \Sigma_x^1 \times \Sigma_y^1)$ , and higher regularity is propagated,  $\phi^x \in C(\mathbb{R}; \Sigma_x^k)$ ,  $\phi^y \in C(\mathbb{R}; \Sigma_y^k)$ , for all  $k \geq 2$ . The approximate solution is then

$$\phi_{\text{app}}(t, x, y) = \phi^x(t, x) \phi^y(t, y) e^{i \int_0^t \langle W \rangle ds},$$

with the phase given by the full average

$$\langle W \rangle = \langle W \rangle(t) = \frac{\int W(x, y) |\phi^x(t, x) \phi^y(t, y)|^2 dx dy}{\int |\varphi_0^x(x) \varphi_0^y(y)|^2 dx dy}.$$

It solves the equation

$$i\partial_t \phi_{\text{app}} = H \phi_{\text{app}} - \Sigma_\phi, \quad \Sigma_\phi := \left( W - \langle W \rangle_x - \langle W \rangle_y + \langle W \rangle \right) \phi_{\text{app}}.$$

*Remark 3.2.* The correcting phase  $e^{i \int_0^t \langle W \rangle ds}$  seems to be crucial if we want to compute the wave function. On the other hand, since it is a purely time dependent phase factor, it does not affect the usual quadratic observables.

In the case where  $W(x, y) = W_1(x)W_2(y)$ , the above formulas become

$$\langle W \rangle_y(t, x) = W_1(x) \langle W_2 \rangle_y(t), \quad \langle W \rangle_x(t, y) = \langle W_1 \rangle_x(t) W_2(y),$$

with

$$(3.3) \quad \begin{aligned} \langle W_2 \rangle &= \langle W_2 \rangle_y(t) = \frac{\int W_2(y) |\phi^y(t, y)|^2 dy}{\int |\phi^y(t, y)|^2 dy} = \frac{\int W_2(y) |\phi^y(t, y)|^2 dy}{\int |\varphi_0^y(y)|^2 dy}, \\ \langle W_1 \rangle &= \langle W_1 \rangle_x(t) = \frac{\int W_1(x) |\phi^x(t, x)|^2 dx}{\int |\phi^x(t, x)|^2 dx} = \frac{\int W_1(x) |\phi^x(t, x)|^2 dx}{\int |\varphi_0^x(x)|^2 dx}, \end{aligned}$$

and the residual is

$$\Sigma_\phi = (W_1 - \langle W_1 \rangle) (W_2 - \langle W_2 \rangle) \phi_{\text{app}}.$$

**3.3. Error estimate and approximation result.** Denote by

$$r_\psi = \psi - \psi_{\text{app}}, \quad r_\phi = \psi - \phi_{\text{app}},$$

the errors corresponding to each of the previous two approximations. They solve

$$(3.4) \quad i\partial_t r_\psi = H r_\psi + \Sigma_\psi \quad ; \quad i\partial_t r_\phi = H r_\phi + \Sigma_\phi \quad ; \quad r_\psi|_{t=0} = r_\phi|_{t=0} = 0.$$

We note that both approximations and their components are norm-conserving for all times  $t \geq 0$ , that is,

$$\|\phi^x(t)\|_{L_x^2} = \|\varphi^x(t)\|_{L_x^2} = \|\varphi_0^x\|_{L_x^2}, \quad \|\phi^y(t)\|_{L_y^2} = \|\varphi^y(t)\|_{L_y^2} = \|\varphi_0^y\|_{L_y^2}.$$

- In the case of the brute-force approach, according to the Taylor expansion that we consider:

$$(3.5) \quad \|\Sigma_\psi\|_{L^2} \leq \begin{cases} 2\|\nabla_y W\|_{L^\infty} \|y\psi_{\text{app}}\|_{L^2} = \|\nabla_y W\|_{L^\infty} \|\varphi^x\|_{L_x^2} \|y\varphi^y\|_{L_y^2}, \\ \|\nabla_x \nabla_y W\|_{L^\infty} \|x\varphi^x\|_{L_x^2} \|y\varphi^y\|_{L_y^2}. \end{cases}$$

- In the mean-field approach, we note that for  $(t, x, y) \in \mathbb{R} \times \mathbb{R}^{n+d}$ ,

$$\begin{aligned} & \left( \int |\varphi_0^x(x') \varphi_0^y(y')|^2 dx' dy' \right) \left( W - \langle W \rangle_x - \langle W \rangle_y + \langle W \rangle \right) (t, x, y) \\ &= \int_{\mathbb{R}^{n+d}} \underbrace{(W(x, y) - W(x, y') - W(x', y) + W(x', y'))}_{=: \delta W(x, x', y, y')} |\phi^x(t, x') \phi^y(t, y')|^2 dx' dy' \end{aligned}$$

Like we did in the brute-force approach, we may use either of the estimates

$$|\delta W(x, x', y, y')| \leq \begin{cases} 2|y - y'| \times \|\nabla_y W\|_{L^\infty}, \\ |x - x'| \times |y - y'| \times \|\nabla_x \nabla_y W\|_{L^\infty}. \end{cases}$$

In the first case, we come up with

$$\begin{aligned} \|\Sigma_\phi\|_{L^2}^2 &\leq 4\|\nabla_y W\|_{L^\infty}^2 \|\varphi_0^x\|_{L_x^2}^2 \times \\ &\quad \times \int_{\mathbb{R}^d} \left( \int_{\mathbb{R}^d} |y - y'| |\phi^y(t, y')|^2 dy' \right)^2 |\phi^y(t, y)|^2 dy / \|\varphi_0^y\|_{L_y^2}^4. \end{aligned}$$

Now we have

$$\begin{aligned} \int_{\mathbb{R}^d} |y - y'| |\phi^y(t, y')|^2 dy' &\leq \int_{\mathbb{R}^d} (|y| + |y'|) |\phi^y(t, y')|^2 dy' \\ &\leq |y| \|\varphi_0^y\|_{L_y^2}^2 + \|y\phi^y(t)\|_{L_y^2} \|\varphi_0^y\|_{L_y^2}, \end{aligned}$$

where we have used Cauchy-Schwarz inequality for the last term. We infer

$$\left( \int_{\mathbb{R}^d} |y - y'| |\phi^y(t, y')|^2 dy' \right)^2 \leq 2|y|^2 \|\varphi_0^y\|_{L_y^2}^4 + 2\|y\phi^y(t)\|_{L_y^2}^2 \|\varphi_0^y\|_{L_y^2}^2,$$

hence

$$\|\Sigma_\phi\|_{L^2}^2 \leq 8\|\nabla_y W\|_{L^\infty}^2 \|\varphi_0^x\|_{L_x^2}^2 \left( \int_{\mathbb{R}^d} |y|^2 |\phi^y(t, y)|^2 dy + \|y\phi^y(t)\|_{L_y^2}^2 \right),$$

and finally

$$(3.6) \quad \|\Sigma_\phi\|_{L^2} \leq 4\|\nabla_y W\|_{L^\infty} \|y\phi^y(t)\|_{L_y^2} \|\varphi_0^x\|_{L_x^2}.$$

In the case of the second type approximation for  $\delta W$ , we find similarly

$$\|\Sigma_\phi\|_{L^2} \leq 4\|\nabla_x \nabla_y W\|_{L^\infty} \|x\phi^x(t)\|_{L_x^2} \|y\phi^y(t)\|_{L_y^2}.$$

*Remark 3.3.* If  $V_1$  is confining,  $V_1(x) \gtrsim |x|^2$  for  $|x| \geq R$  (for instance,  $V_1(x) = c|x|^{2k}$ ,  $c > 0$  and  $k$  a positive integer, a typical case where  $V_1$  may be super-quadratic while  $H_x$  and  $H$  remain self-adjoint), then we can estimate  $\|x\phi^x\|_{L_x^2}$  uniformly in time. If  $V_1 = 0$ , or more generally if  $V_1(x) \rightarrow 0$  as  $|x| \rightarrow \infty$ , we must expect some linear growth in time

$$\|x\phi^x(t)\|_{L_x^2} \lesssim \langle t \rangle,$$

and the order of magnitude in  $t$  is sharp, corresponding to a dispersive phenomenon.

Invoking Lemma 2.3, we readily obtain:

**Proposition 3.4.** *Under Assumption 2.1, we have the following error estimates:*

- *Brute-force approach:* for  $\psi_{\text{app}}(t, x, y) = \varphi^x(t, x)\varphi^y(t, y)e^{itW(0,0)}$ , where  $\varphi^x$  and  $\varphi^y$  are given by (3.1)

$$\|\psi(t) - \psi_{\text{app}}(t)\|_{L^2} \leq \begin{cases} 2\|\nabla_y W\|_{L^\infty} \|\varphi_0^x\|_{L_x^2} \int_0^t \|y\varphi^y(s)\|_{L_y^2} ds, \\ \|\nabla_x \nabla_y W\|_{L^\infty} \int_0^t \|x\varphi^x(s)\|_{L_x^2} \|y\varphi^y(s)\|_{L_y^2} ds. \end{cases}$$

- *Mean-field approach:* for

$$\phi_{\text{app}}(t, x, y) = \phi^x(t, x)\phi^y(t, y)e^{i \int_0^t \langle W \rangle ds}$$

defined by (3.2)–(3.3),

$$\|\psi(t) - \phi_{\text{app}}(t)\|_{L^2} \leq \begin{cases} 4\|\nabla_y W\|_{L^\infty} \|\varphi_0^x\|_{L_x^2} \int_0^t \|y\phi^y(s)\|_{L_y^2} ds, \\ 4\|\nabla_x \nabla_y W\|_{L^\infty} \int_0^t \|x\phi^x(s)\|_{L_x^2} \|y\phi^y(s)\|_{L_y^2} ds. \end{cases}$$

We see that the smallness of  $\|\nabla_y W\|_{L^\infty}$  guarantees that the error between the exact solution  $\psi$  and the approximate solution ( $\psi_{\text{app}}$  or  $\phi_{\text{app}}$ ) is small.

*Example 3.5.* (1) In the case  $W(x, y) = W_1(x)W_2(y)$ , we obviously have

$$\begin{aligned} \|\nabla_y W\|_{L^\infty} &= \|W_1\|_{L_x^\infty} \|\nabla_y W_2\|_{L_y^\infty}, \\ \|\nabla_x \nabla_y W\|_{L^\infty} &= \|\nabla_x W_1\|_{L_x^\infty} \|\nabla_y W_2\|_{L_y^\infty}, \end{aligned}$$

and Proposition 3.4 yields error estimates provided that the above norms are finite.

- (2) An important class of examples consists of those where  $W$  is slowly varying in  $y$ :  $W(x, y) = w(x, \eta y)$  with  $\eta \ll 1$  and  $w$  bounded, as well as its derivatives. In that case

$$\|\nabla_y W\|_{L^\infty} = \eta \|\nabla_y w\|_{L^\infty}.$$

*Remark 3.6.* (1) Suppose we are in case (1) of the preceding examples, and  $W_2(y) = \eta|y|^2$ , with  $\eta$  small:  $\nabla W_2$  is not bounded, but we can adapt the above lines to get

$$\|\Sigma_\phi\|_{L^2} \leq 8\eta\sqrt{2}\|W_1\|_{L^\infty_x}\|\varphi_0^x\|_{L^2_x}\| |y|^2\phi^y\|_{L^2_y},$$

that is, the extra power of  $y$  is transferred to the  $\phi^y$  term.

(2) In the same spirit, with a flat coupling  $W(x, y)$  such that e.g.  $\|\langle y \rangle^{-p}\nabla_y W\|_{L^\infty}$  is small for some  $p > 0$ , we could get an estimate of the form

$$\|\Sigma_\phi\|_{L^2} \leq 4\|\langle y \rangle^{-p}\nabla_y W\|_{L^\infty}\|\varphi_0^x\|_{L^2_x}\|\langle y \rangle^{p+1}\phi^y\|_{L^2_y}.$$

**3.4. Error estimates at higher regularity.** Proposition 3.4 provides  $L^2$ -error estimates. To prove error estimates in  $H^1(\mathbb{R}^{n+d})$ , we differentiate (3.4) in space, and two aspects must be considered:

- The operator  $\nabla_{x,y}$  does not commute with  $H$  (in our framework),
- We must estimate  $\nabla_{x,y}\Sigma_\psi$  and  $\nabla_{x,y}\Sigma_\phi$ .

Indeed, we compute

$$i\partial_t\nabla_x r_\psi = H\nabla_x r_\psi + [\nabla_x, H]r_\psi + \nabla_x\Sigma_\psi,$$

and

$$[\nabla_x, H] = \nabla_x H - H\nabla_x = \nabla_x V_1 + \nabla_x W.$$

In the typical case where  $V_1$  is harmonic,  $\nabla_x V_1$  is linear in  $x$ , and so  $xr_\psi$  appears as a source term. Note that in the general setting of Assumption 2.1,  $|\nabla_x V_1(x)| \lesssim \langle x \rangle$ .

*Remark 3.7.* If  $\nabla_x V_1$  and  $\nabla_x W$  are bounded, then Lemma 2.3 yields

$$\|\nabla_x r_\psi(t)\|_{L^2} \leq \int_0^t (C\|r_\psi(s)\|_{L^2} + \|\nabla_x\Sigma_\psi(s)\|_{L^2}) ds.$$

The term  $\|r_\psi(s)\|_{L^2}$  is estimated in Proposition 3.4, and  $\|\nabla_x\Sigma_\psi(s)\|_{L^2}$  is estimated below.

Multiplying (3.4) by  $x$ , we find similarly

$$i\partial_t(xr_\psi) = H(xr_\psi) + [x, H]r_\psi + x\Sigma_\psi = H(xr_\psi) + \nabla_x r_\psi + x\Sigma_\psi.$$

Energy estimates provided by Lemma 2.3 applied to the equation for  $\nabla_x r_\psi$  and  $xr_\psi$  then yield a closed system of estimates:

$$\begin{aligned} \|\nabla_x r_\psi(t)\|_{L^2} + \|xr_\psi(t)\|_{L^2} &\leq \int_0^t (\|(\nabla_x V_1 + \nabla_x W)r_\psi(s)\|_{L^2} + \|\nabla_x r_\psi(s)\|_{L^2}) ds \\ &\quad + \int_0^t (\|\nabla_x\Sigma_\psi(s)\|_{L^2} + \|x\Sigma_\psi(s)\|_{L^2}) ds \\ &\leq C \int_0^t (\|xr_\psi(s)\|_{L^2} + \|\nabla_x r_\psi(s)\|_{L^2}) ds \\ &\quad + \int_0^t (\|\nabla_x\Sigma_\psi(s)\|_{L^2} + \|x\Sigma_\psi(s)\|_{L^2}) ds, \end{aligned}$$

where we have used the estimate  $|\nabla_x V_1 + \nabla_x W| \leq C(1 + |x|)$ , and the uncertainty principle (uncertainty in  $x$ , Cauchy-Schwarz in  $y$ ),

$$\|f\|_{L^2}^2 \leq \frac{2}{n}\|\nabla_x f\|_{L^2}\|xf\|_{L^2}.$$

The Gronwall Lemma then yields

$$\|\nabla_x r_\psi(t)\|_{L^2} + \|x r_\psi(t)\|_{L^2} \leq \int_0^t e^{Cs} (\|\nabla_x \Sigma_\psi(s)\|_{L^2} + \|x \Sigma_\psi(s)\|_{L^2}) ds,$$

for some  $C > 0$ . We compute

$$\nabla_x \Sigma_\psi = (\nabla_x W(x, y) - \nabla_x W(x, 0)) \psi_{\text{app}} + \delta W(x, 0, y, 0) \nabla_x \psi_{\text{app}}.$$

The first term is controlled by  $|y| \|\nabla_x \nabla_y W\|_{L^\infty} |\psi_{\text{app}}|$ . The second term is controlled like in Section 3.3, by replacing  $\psi_{\text{app}}$  with  $\nabla_x \psi_{\text{app}}$ . We can of course resume the same approach when considering  $\nabla_y r_\psi$ , and the analogue of the above first term is now controlled by  $|x| \|\nabla_x \nabla_y W\|_{L^\infty} |\psi_{\text{app}}|$ . Finally, in the case of  $r_\phi$ , computations are similar (we do not keep track of the precise dependence of multiplicative constants here), and we have (using the second family of estimate from Proposition 3.4):

**Proposition 3.8.** *Under Assumption 2.1, there exists a constant  $C > 0$  such that we have the following error estimates:*

- *Brute-force approach: for  $\psi_{\text{app}}(t, x, y) = \varphi^x(t, x) \varphi^y(t, y) e^{itW(0,0)}$ , where  $\varphi^x$  and  $\varphi^y$  are given by (3.1)*

$$\begin{aligned} & \|\nabla_x \psi(t) - \nabla_x \psi_{\text{app}}(t)\|_{L^2} + \|x \psi(t) - x \psi_{\text{app}}(t)\|_{L^2} \leq C \|\nabla_x \nabla_y W\|_{L^\infty} \times \\ & \times \int_0^t e^{Cs} \|y \varphi^y(s)\|_{L_y^2} (\|x \varphi^x(s)\|_{L_x^2} + \|\nabla_x \varphi^x(s)\|_{L_x^2} + \|x |\nabla_x \varphi^x(s)|\|_{L_x^2}) ds, \end{aligned}$$

$$\begin{aligned} & \|\nabla_y \psi(t) - \nabla_y \psi_{\text{app}}(t)\|_{L^2} + \|y \psi(t) - y \psi_{\text{app}}(t)\|_{L^2} \leq C \|\nabla_x \nabla_y W\|_{L^\infty} \times \\ & \times \int_0^t e^{Cs} \|x \varphi^x(s)\|_{L_x^2} (\|y \varphi^y(s)\|_{L_y^2} + \|\nabla_y \varphi^y(s)\|_{L_y^2} + \|y |\nabla_y \varphi^y(s)|\|_{L_y^2}) ds. \end{aligned}$$

- *Mean-field approach: for*

$$\phi_{\text{app}}(t, x, y) = \phi^x(t, x) \phi^y(t, y) e^{i \int_0^t \langle W \rangle ds}$$

*defined by (3.2)–(3.3), then  $\psi(t) - \phi_{\text{app}}(t)$  satisfies similar estimates.*

*Remark 3.9.* The approach presented above can be iterated to infer error estimates in Sobolev spaces of higher order,  $H^k(\mathbb{R}^{n+d})$  for  $k \geq 2$ , provided that we consider momenta of the same order, that is, provided that we consider  $\langle x \rangle^k r_\psi$  and  $\langle y \rangle^k r_\psi$ , which explains the interest in the functional spaces  $\Sigma^k$ . Error estimates in such spaces can also be obtained by first proving that  $\psi$  and the approximate solution(s) remain in  $\Sigma^k$ , and then interpolating with the  $L^2$  error estimate from Proposition 3.4.

**3.5. Error estimates for quadratic observables.** For obtaining quadratic estimates, we consider observables such as the energy or the momenta, that is, operators that are differential operators of order at most 2 with bounded smooth coefficients. These differential operators have their domain in  $H^2(\mathbb{R}^{n+d})$ , as the operator  $H$ . More generally, we could consider pseudo-differential operators  $B = \text{op}(b)$  associated with a smooth function  $b = b(Z)$  with  $Z = (z, \zeta) \in \mathbb{R}^{2(n+d)}$ , whose action on functions  $f \in \mathcal{S}(\mathbb{R}^{n+d})$  is given by

$$\text{op}(b)f(z) = (2\pi)^{-(n+d)} \int_{\mathbb{R}^{2(n+d)}} b\left(\frac{z+z'}{2}, \zeta\right) e^{i\zeta \cdot (z-z')} f(z') d\zeta dz'.$$

We assume that  $b$  satisfies the Hörmander condition

$$(3.7) \quad \forall \alpha, \beta \in \mathbb{N}^{n+d}, \exists C_{\alpha, \beta} > 0, \quad |\partial_z^\beta \partial_\zeta^\alpha b(z, \zeta)| \leq C_{\alpha, \beta} \langle \zeta \rangle^{2-|\alpha|},$$

that is,  $b$  is a symbol of order 2, see e.g. [1, Chapter I.2]. We shall also consider observables that depend only on the variable  $x$  or the variable  $y$ . Let  $B = \text{op}(b)$  be such an observable satisfying (3.7). We focus on a posteriori estimates and work first on the difference for the brute force approach

$$e_\psi(t) = \langle \psi(t), B\psi(t) \rangle - \langle \psi_{\text{app}}(t), B\psi_{\text{app}}(t) \rangle.$$

We use Lemma 2.4 for the operators  $H$  and the approximate Hamiltonian  $H_{\text{bf}}$ ,

$$(3.8) \quad H_{\text{bf}} = H_x + H_y + W(x, 0) + W(0, y) - W(0, 0),$$

to obtain

$$|e_\psi(t)| \leq \int_0^t |\rho_\psi(t, s)| ds,$$

where

$$\rho_\psi(t, s) = \langle \psi_{\text{app}}(s), [B(t-s), H - H_{\text{bf}}] \psi_{\text{app}}(s) \rangle, \quad B(\sigma) = e^{i\sigma H} B e^{-i\sigma H}.$$

By Egorov Theorem, see [35, Theorem 11.1], the operator  $B(\sigma)$  is also a pseudo-differential operator, that is,  $B(\sigma) = \text{op}(b(\sigma))$  for some function  $b(\sigma)$  that satisfies the growth condition (3.7). We have

$$H - H_{\text{bf}} = W(x, y) - W(x, 0) - W(0, y) + W(0, 0) = \delta W(x, 0, y, 0) =: \delta W(x, y),$$

with the notations of Section 3.3. Then, by the direct estimate of Lemma A.1,

$$\begin{aligned} & \| [B(\sigma), \delta W] \psi_{\text{app}}(s) \|_{L^2} \leq \\ & C_{b(\sigma)} \left( \| \nabla(\delta W) \psi_{\text{app}}(s) \|_{H^1} + C_2(\delta W) \| \psi_{\text{app}}(s) \|_{L^2} \right), \end{aligned}$$

where  $C_{b(\sigma)} > 0$  depends on derivative bounds for the function  $b(\sigma)$  and

$$C_2(\delta W) = \sum_{2 \leq |\alpha| \leq N_n + d} \| \partial^\alpha \delta W \|_{L^\infty}.$$

We therefore obtain

$$|\rho_\psi(t, s)| \leq C_{b(t-s)} \left( \| \nabla(\delta W) \psi_{\text{app}}(s) \|_{H^1} + C_2(\delta W) \| \psi_0 \|_{L^2} \right) \| \psi_0 \|_{L^2}.$$

Using the rectangular  $n \times d$  matrix  $M(x, y)$  introduced in Remark 3.1, the gradient of  $\delta W(x, y)$  can be written as

$$\nabla(\delta W)(x, y) = \begin{pmatrix} \nabla_x W(x, y) - \nabla_x W(x, 0) \\ \nabla_y W(x, y) - \nabla_y W(0, y) \end{pmatrix} = \begin{pmatrix} \int_0^1 M(x, \eta y) y d\eta \\ \int_0^1 {}^t M(\theta x, y) x d\theta \end{pmatrix}$$

We estimate the Sobolev norm by

$$\begin{aligned} \| \nabla(\delta W) \psi_{\text{app}}(s) \|_{H^1} & \leq \| \nabla M \|_{L^\infty} (\| x \psi_{\text{app}}(s) \|_{L^2} + \| y \psi_{\text{app}}(s) \|_{L^2}) \\ & \quad + \| M \|_{L^\infty} (\| \nabla(x \psi_{\text{app}}(s)) \|_{L^2} + \| \nabla(y \psi_{\text{app}}(s)) \|_{L^2}), \end{aligned}$$

so that integration in time provides

$$\begin{aligned} |e_\psi(t)| &\leq C_b \|\nabla M\|_{L^\infty} \|\psi_0\|_{L^2} \int_0^t (\|x\psi_{\text{app}}(s)\|_{L^2} + \|y\psi_{\text{app}}(s)\|_{L^2}) ds \\ &\quad + C_b \|M\|_{L^\infty} \|\psi_0\|_{L^2} \int_0^t (\|\nabla(x\psi_{\text{app}}(s))\|_{L^2} + \|\nabla(y\psi_{\text{app}}(s))\|_{L^2}) ds \\ &\quad + C_b C_2(\delta W) t \|\psi_0\|_{L^2}^2, \end{aligned}$$

where the constant  $C_b = \max_{\sigma \in [0, t]} C_{b(\sigma)}$  depends on derivatives of  $b$ . In the mean-field case, the approximate Hamiltonian is time-dependent,

$$(3.9) \quad H_{\text{mf}}(t) = H_x + H_y + \langle W \rangle_y(t) + \langle W \rangle_x(t) - \langle W \rangle(t).$$

The difference of the Hamiltonians is also a function, which is now time-dependent,

$$H - H_{\text{mf}}(t) = W + \langle W \rangle(t) - \langle W \rangle_x(t) - \langle W \rangle_y(t).$$

However, it is easy to check that a similar estimate can be performed, leading to an analogous conclusion. We sum up these results in a statement.

**Proposition 3.10.** *Under Assumption 2.1, for  $b \in C^\infty(\mathbb{R}^{n+d})$  satisfying (3.7) and  $B = \text{op}(b)$ , there exists a constant  $C_b > 0$  such that we have the following error estimates:*

- *Brute-force approach: for  $\psi_{\text{app}}(t, x, y) = \varphi^x(t, x)\varphi^y(t, y)e^{itW(0,0)}$ , where  $\varphi^x$  and  $\varphi^y$  are given by (3.1), the error*

$$e_\psi(t) = \langle \psi(t), B\psi(t) \rangle - \langle \psi_{\text{app}}(t), B\psi_{\text{app}}(t) \rangle$$

*satisfies*

$$|e_\psi(t)| \leq C_b \sup_{|\beta| \leq N_{n+d}} \|\nabla^\beta M\|_{L^\infty} \|\psi_0\|_{L^2} (\mathcal{N}(\psi_{\text{app}}) + t\|\psi_0\|_{L^2}),$$

*where*

$$\begin{aligned} \mathcal{N}(\psi_{\text{app}}) &= \|\varphi_0^x\|_{L_x^2} \int_0^t \|y\varphi^y(s)\|_{L_y^2} ds + \|\varphi_0^y\|_{L_y^2} \int_0^t \|x\varphi^x(s)\|_{L_x^2} ds \\ &\quad + \|\varphi_0^x\|_{L_x^2} \int_0^t \|\nabla(y\varphi^y(s))\|_{L_y^2} ds + \|\varphi_0^y\|_{L_y^2} \int_0^t \|\nabla(x\varphi^x(s))\|_{L_x^2} ds \\ &\quad + \int_0^t \|x\varphi^x(s)\|_{L_x^2} \|\nabla\varphi^y(s)\|_{L_y^2} ds + \int_0^t \|\nabla\varphi^x(s)\|_{L_x^2} \|y\varphi^y(s)\|_{L_y^2} ds \end{aligned}$$

*and  $M(x, y) = \partial_x \partial_y W(x, y)$ , while  $N_{n+d} > 0$  depends on  $n + d$ .*

- *Mean-field approach: for*

$$\phi_{\text{app}}(t, x, y) = \phi^x(t, x)\phi^y(t, y)e^{i \int_0^t \langle W \rangle ds}$$

*defined by (3.2)–(3.3), then  $\langle \psi(t), B\psi(t) \rangle - \langle \phi_{\text{app}}(t), B\phi_{\text{app}}(t) \rangle$  satisfies similar estimates.*

*Remark 3.11.* We point out that the error is governed by derivatives of second order in  $W$ , involving a derivative in the  $y$  variable that is supposed to be small. Besides, note that the direct use of an estimate on the wave function itself would have involved  $H^2$  norms of  $\psi_{\text{app}}(s)$ , while this estimate only requires  $H^1$  norms. This first improvement is due to the averaging process present in Egorov Theorem.

**3.6. Energy conservation.** In comparison with the brute-force approximation, the mean-field approximation has the advantage to be both norm-preserving and energy-preserving, even though the mean-field Hamiltonian  $H_{\text{mf}}(t)$  depends on time (see (3.9)). If one derives the mean-field approach from the time-dependent Dirac–Frenkel variational principle, then energy conservation is immediate, see [24, Chapter II.1.5]. Here we give an elementary ad-hoc proof.

**Lemma 3.12.** *Under Assumption 2.1 and considering the mean-field approach:*

$$\phi_{\text{app}}(t, x, y) = \phi^x(t, x)\phi^y(t, y)e^{i \int_0^t \langle W \rangle(s) ds},$$

defined by (3.2)–(3.3), then

$$\langle \phi_{\text{app}}(t), H_{\text{mf}}(t)\phi_{\text{app}}(t) \rangle = \langle \psi_0, H\psi_0 \rangle \quad \text{for all } t \geq 0.$$

*Proof.* A first observation is that

$$\langle \phi_{\text{app}}(t), H_{\text{mf}}(t)\phi_{\text{app}}(t) \rangle = \langle \psi_0, H_{\text{mf}}(0)\psi_0 \rangle \quad \text{for all } t \geq 0.$$

Indeed,

$$\begin{aligned} & \frac{d}{dt} \langle \phi_{\text{app}}(t), H_{\text{mf}}(t)\phi_{\text{app}}(t) \rangle \\ &= \langle \phi_{\text{app}}(t), \partial_t H_{\text{mf}}(t)\phi_{\text{app}}(t) \rangle = \langle \phi_{\text{app}}(t), \partial_t W_{\text{app}}(t)\phi_{\text{app}}(t) \rangle \end{aligned}$$

with  $W_{\text{app}}(t) = \langle W \rangle_y(t) + \langle W \rangle_x(t) - \langle W \rangle(t)$ . We deduce

$$\begin{aligned} & \frac{d}{dt} \langle \phi_{\text{app}}(t), H_{\text{mf}}(t)\phi_{\text{app}}(t) \rangle \\ &= \int W(x, y) (\partial_t |\phi^x(t, x)|^2 |\phi^y(t, y)|^2 + |\phi^x(t, x)|^2 \partial_t |\phi^y(t, y)|^2) dx dy \\ &\quad - \int W(x, y) \partial_t (|\phi^x(t, x)|^2 |\phi^y(t, y)|^2) dx dy = 0, \end{aligned}$$

where we have used the self-adjointness of  $H_{\text{mf}}(t)$  and norm-conservation in the multiplicative components. Secondly, since

$$\begin{aligned} \langle \psi_0, W_{\text{app}}(0)\psi_0 \rangle &= \left\langle \psi_0, \left( \langle W \rangle_x(0) + \langle W \rangle_y(0) - \langle W \rangle(0) \right) \psi_0 \right\rangle \\ &= 2 \langle W \rangle(0) - \langle W \rangle(0) = \langle W \rangle(0), \end{aligned}$$

the approximate energy coincides with the actual energy, and we obtain the result of Lemma 3.12.  $\square$

*Remark 3.13.* In the brute-force case,  $H_{\text{bf}}$ , defined by (3.8), is time-independent, and we have

$$\langle \psi_{\text{app}}(t), H_{\text{bf}}\psi_{\text{app}}(t) \rangle = \langle \psi_0, H_{\text{bf}}\psi_0 \rangle \quad \text{for all } t \geq 0.$$

However this conserved value does not correspond to the exact energy of (1.1), but only to an approximation of it.

## 4. DIMENSION REDUCTION VIA SEMICLASSICAL ANALYSIS

In this section, we consider coupled systems, where one part is governed by a semiclassically scaled Hamiltonian, that is,  $H_y = H_y^\varepsilon$  with

$$H_y^\varepsilon = -\frac{\varepsilon^2}{2}\Delta_y + V_2(y).$$

First we motivate such a partial semiclassical scaling in the context of system-bath Hamiltonians and introduce wave packets as natural initial data for the semiclassical part of the system. We explore partial semiclassical wave packet dynamics guided by classical trajectories and by trajectories with averaged potentials. Thus, the partially highly-oscillatory evolution of a PDE in dimension  $n+d$  is reduced to a less-oscillatory PDEs in dimensions  $n$ , and ODEs in dimension  $d$ . The corresponding error estimates in § 4.5 compare the true and the approximate product solution in norm and with respect to expectation values.

**4.1. Semiclassical scaling.** We reconsider the system-bath Hamiltonian with cubic coupling of §2.2, now formulated in physical coordinates  $(X, Y)$ , that is,

$$H_{\text{sb}} = -\frac{\hbar^2}{2\mu_1}\Delta_X + V_s(X) - \frac{\hbar^2}{2\mu_2}\Delta_Y + \frac{\mu_2\omega_2^2}{2}|Y|^2 + \frac{1}{2}\vec{\eta} \cdot X|Y|^2,$$

where the coordinates  $X$  and  $Y$  of the system and the bath part are prescaled, resulting in the single mass parameters  $\mu_1, \mu_2$  for each subsystem and one single harmonic frequency  $\omega_2$  for the bath (noting that, alternatively, several harmonic bath frequencies  $\omega_{2,j}$  could be introduced, without modifying the conclusions detailed below). The corresponding time-dependent Schrödinger equation reads

$$i\hbar\partial_\tau\Psi(\tau, X, Y) = H_{\text{sb}}\Psi(\tau, X, Y).$$

We perform a local harmonic expansion of the potential  $V_s(X)$  around the origin  $X = 0$  and assume that it is possible to determine a dominant frequency  $\omega_1$ . We then define the natural length scale of the system as

$$a = \sqrt{\frac{\hbar}{\mu_1\omega_1}}.$$

Rescaling coordinates as  $(x, y) = \frac{1}{a}(X, Y)$ , we obtain

$$H_{\text{sb}} = \hbar\omega_1 \left( -\frac{1}{2}\Delta_x + V_1(x) - \frac{\varepsilon^2}{2}\Delta_y + \frac{1}{2}\frac{\varpi^2}{\varepsilon^2}|y|^2 + \frac{1}{2}\vec{\eta}' \cdot x|y|^2 \right),$$

where we have introduced the dimensionless parameters

$$\varepsilon = \sqrt{\frac{\mu_1}{\mu_2}} \quad , \quad \varpi = \sqrt{\frac{\omega_2}{\omega_1}} \quad ,$$

and denoted

$$V_1(x) = \frac{1}{\hbar\omega_1}V_s(ax) \quad , \quad \vec{\eta}' = \frac{a}{\mu_1\omega_1^2}\vec{\eta}.$$

The rescaling of the system potential  $V_s(X)$  and the coupling vector  $\vec{\eta}$  do not alter their role in the Hamiltonian, whereas the two dimensionless parameters  $\varepsilon$  and  $\varpi$  deserve further attention. We now consider the régime where both the mass ratio  $\varepsilon$  between system and bath and the frequency ratio  $\varpi$  between bath and system are small, that is, where the system is viewed as “light” and “fast” when compared to the “heavy” and “slow” bath.

*Example 4.1.* For the hydrogen molecule  $\text{H}_2$ , where the electrons are considered as the quantum subsystem while the interatomic vibration is considered as the classical subsystem, we have  $\mu_1 = m_e$  and  $\mu_2 = 918.6m_e$ . Further, the characteristic electronic energy is of the order of  $\hbar\omega_1 = 1E_h$  while the first vibrational level is found at  $\hbar\omega_2 = 0.02005E_h$ . Hence the dimensionless parameters are both small,  $\varepsilon = 0.03299$  and  $\varpi = 0.1416$ .

*Example 4.2.* As a second example, we consider coupled molecular vibrations, exemplified by the  $\text{H}_2$  molecule in a “bath” of rare-gas atoms, here chosen as krypton (Kr) atoms. The  $\text{H}_2$  vibration is now considered as a quantum system interacting with weak intermolecular vibrations. The reduced masses are given as  $\mu_1(\text{H-H}) = 0.5 \text{ u} = 911.44 m_e$  (where u refers to atomic mass units),  $\mu_2(\text{Kr-Kr}) = 41.9 \text{ u} = 76.379 \times 10^3 m_e$ , and  $\mu_3(\text{H}_2\text{-Kr}) = 1.953 \text{ u} = 3560.10 m_e$ . The vibrational quanta associated with these vibrations are  $\hbar\omega_1(\text{H-H}) = 4159.2\text{cm}^{-1} = 0.0189E_h$ ,  $\hbar\omega_2(\text{Kr-Kr}) = 21.6\text{cm}^{-1} = 9.82 \times 10^{-5} E_h$ , and  $\hbar\omega_3(\text{H}_2\text{-Kr}) = 26.8\text{cm}^{-1} = 1.22 \times 10^{-4} E_h$  (see Refs. [17, 33]). The resulting dimensionless mass ratios are given as  $\epsilon_{12} = \sqrt{\mu_1/\mu_2} = 0.109$  and  $\epsilon_{13} = \sqrt{\mu_1/\mu_3} = 0.51$ , and the corresponding frequency ratios are  $\varpi_{12} = \sqrt{\omega_2/\omega_1} = 0.072$  and  $\varpi_{13} = \sqrt{\omega_3/\omega_1} = 0.080$ . In the case of the  $\text{H}_2$ -Kr relative motion, note that the frequency ratio  $\varpi_{13}$  is indeed small whereas the mass ratio is  $\epsilon_{13} \sim 0.5$ ; this shows that the quantum-classical boundary is less clearly defined than in the first example of coupled electronic-nuclear motions. In such cases, different choices can be made in defining the quantum-classical partitioning.

In an idealized setting, where  $\varepsilon$  is considered as a small positive parameter whose size can be arbitrarily small, we would say that

$$\varpi = \mathcal{O}(\varepsilon) \quad \text{as } \varepsilon \rightarrow 0,$$

and view the system-bath Hamiltonian  $H_{\text{sb}}$  as an instance of a partially semiclassical operator

$$H^\varepsilon = -\frac{1}{2}\Delta_x + V_1(x) - \frac{\varepsilon^2}{2}\Delta_y + V_2(y) + W(x, y),$$

whose potentials  $V_1(x)$  and  $V_2(y)$  are independent of the semiclassical parameter  $\varepsilon$  and satisfy the growth conditions of Assumption 2.1 (see also Remark 2.2). As emphasized in §2.2, the cubic coupling potential does not satisfy the subquadratic estimate, but can be controlled by additional moments of the approximate solution. A corresponding rescaling of time,  $t = \tau/(\varepsilon\omega_1)$ , translates the time-dependent Schrödinger equation to its semiclassical counterpart

$$(4.1) \quad i\varepsilon\partial_t\psi^\varepsilon(t, x, y) = H^\varepsilon\psi^\varepsilon(t, x, y),$$

where the physical and the rescaled wave functions are related via

$$\psi^\varepsilon(t, x, y) = a^{(n+d)/2} \Psi(\tau/(\varepsilon\omega_1), aX, aY).$$

**4.2. Semiclassical initial data and ansatz.** As before, the initial data separate scales,

$$(4.2) \quad \psi^\varepsilon(0, x, y) = \varphi_0^x(x)\mathbf{g}^\varepsilon(y),$$

where we now assume that  $\mathbf{g}^\varepsilon$  is a semiclassically scaled wave packet,

$$(4.3) \quad \mathbf{g}^\varepsilon(y) = \frac{1}{\varepsilon^{d/4}} a \left( \frac{y - q_0}{\sqrt{\varepsilon}} \right) e^{ip_0 \cdot (y - q_0)/\varepsilon},$$

with  $(q_0, p_0) \in \mathbb{R}^{2d}$ ,  $a$  rapidly decreasing, i.e.  $a \in \mathcal{S}(\mathbb{R}^d; \mathbb{C}) = \bigcap_{k \in \mathbb{N}} \Sigma^k$  ( $a$  is typically a complex Gaussian but not necessarily).

We now seek an approximate solution of the form

$$(4.4) \quad \psi_{\text{app}}^\varepsilon(t, x, y) = \psi_1^\varepsilon(t, x) \psi_2^\varepsilon(t, y),$$

where  $\psi_2^\varepsilon$  is a semiclassically scaled wave packet for all time,

$$(4.5) \quad \psi_2^\varepsilon(t, y) = \frac{1}{\varepsilon^{d/4}} u_2 \left( t, \frac{y - q(t)}{\sqrt{\varepsilon}} \right) e^{ip(t) \cdot (y - q(t)) / \varepsilon + iS(t) / \varepsilon}.$$

Here,  $(q(t), p(t)) \in \mathbb{R}^{2d}$ , the phase  $S(t) \in \mathbb{R}$ , and the amplitude  $u_2(t) \in \mathcal{S}(\mathbb{R}^d, \mathbb{C})$  must be determined. Denote by

$$z = \frac{y - q(t)}{\sqrt{\varepsilon}}$$

the space variable for  $u_2$ , or, equivalently,  $y = q(t) + z\sqrt{\varepsilon}$ . Set

$$(4.6) \quad u_{\text{app}}^\varepsilon(t, x, z) = \psi_1^\varepsilon(t, x) u_2(t, z)$$

the part of the approximate solution that just contains the amplitude. With this notation,

$$\psi_{\text{app}}^\varepsilon(t, x, y) = \frac{1}{\varepsilon^{d/4}} u_{\text{app}}^\varepsilon(t, x, z) e^{ip(t) \cdot z / \sqrt{\varepsilon} + iS(t) / \varepsilon} \Big|_{z = \frac{y - q(t)}{\sqrt{\varepsilon}}}.$$

The analysis developed in the two next sections allows to derive two different approximations, based on ordinary differential equations governing the semiclassical wave packet part, which are justified in Section 4.5 (see Proposition 4.4).

*Remark 4.3.* We note that our approximation ansatz (4.4) differs from the adiabatic one, that would write the full Hamiltonian as  $H^\varepsilon = -\frac{\varepsilon^2}{2} \Delta_y + H_f(y)$ , where

$$H_f(y) = -\frac{1}{2} \Delta_x + V_1(x) + V_2(y) + W(x, y)$$

is an operator, that parametrically depends on the “slow” variable  $y$  and acts on the “fast” degrees of freedom  $x$ . From the adiabatic point of view, one would then construct an approximate solution as  $\psi_{\text{bo}}^\varepsilon(t, x, y) = \Phi(x, y) \psi_2^\varepsilon(t, y)$ , where  $\Phi(x, y)$  is an eigenfunction of the operator  $H_f(y)$ ; here, the subscript “bo” stands for Born-Oppenheimer. The result of Corollary 4.8 emphasizes the difference between these two points of view.

**4.3. Approximation by partial Taylor expansion.** Plugging the expression of  $\psi_{\text{app}}^\varepsilon(t, x, y)$  into (4.1), and using Taylor expansions

$$\begin{aligned} V_2(y) &= V_2(q(t) + z\sqrt{\varepsilon}) = V_2(q(t)) + \sqrt{\varepsilon} z \cdot \nabla V_2(q(t)) + \frac{\varepsilon}{2} \langle z, \nabla^2 V_2(q(t)) z \rangle + \mathcal{O}(\varepsilon^{3/2}), \\ W(x, y) &= W(x, q(t) + z\sqrt{\varepsilon}) = W(x, q(t)) + \sqrt{\varepsilon} z \cdot \nabla_y W(x, q(t)) + \mathcal{O}(\varepsilon), \end{aligned}$$

we find:

$$\begin{aligned}
i\varepsilon\partial_t\psi_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x\psi_{\text{app}}^\varepsilon + \frac{\varepsilon^2}{2}\Delta_y\psi_{\text{app}}^\varepsilon - V(x,y)\psi_{\text{app}}^\varepsilon &= \frac{1}{\varepsilon^{d/4}}e^{ip(t)\cdot z/\sqrt{\varepsilon}+iS(t)/\varepsilon} \times \\
&\left( \left( p \cdot \dot{q} - \dot{S} - \frac{|p|^2}{2} - V_2(q) - V_1(x) - W(x,q) \right) u_{\text{app}}^\varepsilon \right. \\
&\quad + \sqrt{\varepsilon} \left( -i\dot{q} \cdot \nabla_z u_{\text{app}}^\varepsilon - \dot{p} \cdot z u_{\text{app}}^\varepsilon + ip \cdot \nabla_z u_{\text{app}}^\varepsilon - z \cdot \nabla V_2(q) u_{\text{app}}^\varepsilon - z \cdot \nabla_y W(x,q) u_{\text{app}}^\varepsilon \right) \\
&\quad + \varepsilon \left( i\partial_t u_{\text{app}}^\varepsilon + \frac{1}{2\varepsilon}\Delta_x u_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_z u_{\text{app}}^\varepsilon - \frac{1}{2}\langle z, \nabla^2 V_2(q) z \rangle u_{\text{app}}^\varepsilon \right) \\
&\quad \left. + \mathcal{O}(\varepsilon^{3/2}) \right),
\end{aligned}$$

where the argument of  $u_{\text{app}}^\varepsilon$  and its derivatives are taken in  $z = \frac{y-q(t)}{\sqrt{\varepsilon}}$ . To cancel the first four terms in the  $\sqrt{\varepsilon}$  line, it is natural to require

$$(4.7) \quad \dot{q} = p, \quad q(0) = q_0, \quad \dot{p} = -\nabla V_2(q), \quad p(0) = p_0.$$

Now cancelling the first four terms in the first line of the right hand side yields

$$(4.8) \quad S(t) = \int_0^t \left( \frac{|p(s)|^2}{2} - V_2(q(s)) \right) ds.$$

In other words,  $(q(t), p(t))$  is the classical trajectory in  $y$ , and  $S(t)$  is the associated classical action. At this stage, we note that the term  $z \cdot \nabla_y W(x, q) u_{\text{app}}^\varepsilon$  is not compatible with decoupling the variables  $x$  and  $z$  (or equivalently,  $x$  and  $y$ ). Using that  $\|\nabla_y W\|_{L^\infty}$  is assumed to be small, the above computation becomes

$$\begin{aligned}
i\varepsilon\partial_t\psi_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x\psi_{\text{app}}^\varepsilon + \frac{\varepsilon^2}{2}\Delta_y\psi_{\text{app}}^\varepsilon - V(x,y)\psi_{\text{app}}^\varepsilon &= \frac{1}{\varepsilon^{d/4}}e^{ip(t)\cdot z/\sqrt{\varepsilon}+iS(t)/\varepsilon} \times \\
&\left( i\varepsilon\partial_t u_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x u_{\text{app}}^\varepsilon + \frac{\varepsilon}{2}\Delta_z u_{\text{app}}^\varepsilon - \left( \frac{\varepsilon}{2}\langle z, \nabla^2 V_2(q) z \rangle + V_1(x) + W(x,q) \right) u_{\text{app}}^\varepsilon \right. \\
&\quad \left. + \mathcal{O}(\varepsilon^{3/2} + \sqrt{\varepsilon}\|\nabla_y W\|_{L^\infty}) \right).
\end{aligned}$$

In view of (4.6), we set

$$(4.9) \quad i\varepsilon\partial_t\psi_1^\varepsilon + \frac{1}{2}\Delta_x\psi_1^\varepsilon = (V_1(x) + W(x,q))\psi_1^\varepsilon \quad ; \quad \psi_1^\varepsilon|_{t=0} = \varphi_0^\varepsilon$$

and

$$(4.10) \quad i\partial_t u_2 + \frac{1}{2}\Delta_z u_2 = \frac{1}{2}\langle z, \nabla^2 V_2(q) z \rangle u_2 \quad ; \quad u_2|_{t=0} = a.$$

Equation (4.10) is a Schrödinger equation with a time-dependent harmonic potential: it has a unique solution in  $L^2$  as soon as  $a \in L^2(\mathbb{R}^d)$ . In addition, since  $a \in \Sigma^k$  for all  $k \in \mathbb{N}$ ,  $u_2 \in C(\mathbb{R}; \Sigma_z^k)$  for all  $k \in \mathbb{N}$ . The approximate solution  $\psi_{\text{app}}^\varepsilon$  solves

$$i\varepsilon\partial_t\psi_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x\psi_{\text{app}}^\varepsilon + \frac{\varepsilon^2}{2}\Delta_y\psi_{\text{app}}^\varepsilon - V\psi_{\text{app}}^\varepsilon = \frac{1}{\varepsilon^{d/4}}e^{ip\cdot z/\sqrt{\varepsilon}+iS/\varepsilon} (r_1^\varepsilon + r_2^\varepsilon),$$

where the remainder  $r_1^\varepsilon$  is due to the Taylor expansion in  $V_2$ , and satisfies the pointwise estimate

$$|r_1^\varepsilon(t, x, z)| \leq \frac{1}{6} \times \varepsilon^{3/2} \|\nabla^3 V_2\|_{L^\infty} |\psi_1^\varepsilon(t, x)| \times |z|^3 |u_2(t, z)|,$$

while the remainder  $r_2^\varepsilon$  is due to the Taylor expansion in  $W$ , and satisfies the pointwise estimate

$$|r_2^\varepsilon(t, x, z)| \leq \sqrt{\varepsilon} \|\nabla_y W\|_{L^\infty} |\psi_1^\varepsilon(t, x)| \times |z| |u_2(t, z)|.$$

To summarize, the approximate solution obtained by Taylor expansion is given by

$$\begin{aligned} \psi_{\text{app}}^\varepsilon(t, x, y) &= \frac{1}{\varepsilon^{d/4}} e^{ip(t) \cdot (y - q(t)) / \varepsilon + iS(t) / \varepsilon} u_2 \left( t, \frac{y - q(t)}{\sqrt{\varepsilon}} \right) \psi_1^\varepsilon(t, x), \\ \text{with } i\varepsilon \partial_t \psi_1^\varepsilon + \frac{1}{2} \Delta_x \psi_1^\varepsilon &= (V_1(x) + W(x, q)) \psi_1^\varepsilon \quad ; \quad \psi_1^\varepsilon|_{t=0} = \varphi_0^x, \\ \text{and } i\partial_t u_2 + \frac{1}{2} \Delta_z u_2 &= \frac{1}{2} \langle z, \nabla^2 V_2(q) z \rangle u_2 \quad ; \quad u_2|_{t=0} = a, \\ \dot{q} &= p, \quad q(0) = q_0, \quad \dot{p} = -\nabla V_2(q), \quad p(0) = p_0, \quad S(t) = \int_0^t \left( \frac{|p(s)|^2}{2} - V_2(q(s)) \right) ds. \end{aligned}$$

The validity of this approximation is stated in Proposition 4.4 below. Note that if  $a$  is a Gaussian state, then  $u_2$  too and its (time-dependent) parameters – width matrix and center point – can be computed by solving ODEs (see e.g. [24, 7, 11, 21] and references therein).

**4.4. Approximation by partial averaging.** Following e.g. [10], we write

$$\begin{aligned} V_2(y) &= V_2(q(t) + z\sqrt{\varepsilon}) = \langle V_2 \rangle_y(t) + \sqrt{\varepsilon} z \cdot \langle \nabla V_2 \rangle_y(t) + \frac{\varepsilon}{2} z \cdot \langle \nabla^2 V_2 \rangle_y(t) z + v_1, \\ W(x, y) &= W(x, q(t) + z\sqrt{\varepsilon}) = \langle W(x, \cdot) \rangle_y(t) + v_2, \end{aligned}$$

where the averages are with respect to  $|\psi_2^\varepsilon(t, y)|^2$ . For example,

$$\begin{aligned} (4.11) \quad \langle \nabla^2 V_2 \rangle_y(t) &= \frac{\int \nabla^2 V_2(y) |\psi_2(t, y)|^2 dy}{\int |\psi_2(t, y)|^2 dy} = \frac{1}{\|a\|_{L^2(\mathbb{R}^d)}^2} \int_{\mathbb{R}^d} \nabla^2 V_2(q(t) + \sqrt{\varepsilon} z) |u_2(t, z)|^2 dz, \\ \langle W(x, \cdot) \rangle_y(t) &= \frac{1}{\|a\|_{L^2(\mathbb{R}^d)}^2} \int_{\mathbb{R}^d} W(x, q(t) + \sqrt{\varepsilon} z) |u_2(t, z)|^2 dz, \end{aligned}$$

where we anticipate the fact that the  $L_y^2$ -norm of  $\psi_2^\varepsilon(t)$  is independent of time. We almost literally repeat the previous argument and find that

$$\begin{aligned} i\varepsilon \partial_t \psi_{\text{app}}^\varepsilon + \frac{1}{2} \Delta_x \psi_{\text{app}}^\varepsilon + \frac{\varepsilon^2}{2} \Delta_y \psi_{\text{app}}^\varepsilon - V(x, y) \psi_{\text{app}}^\varepsilon &= \frac{1}{\varepsilon^{d/4}} e^{ip(t) \cdot z / \sqrt{\varepsilon} + iS(t) / \varepsilon} \times \\ &\left( \left( p \cdot \dot{q} - \dot{S} - \frac{|p|^2}{2} - \langle V_2 \rangle_y - V_1(x) - \langle W(x, \cdot) \rangle_y \right) u_{\text{app}}^\varepsilon \right. \\ &+ \sqrt{\varepsilon} \left( -i\dot{q} \cdot \nabla_z u_{\text{app}}^\varepsilon - \dot{p} \cdot z u_{\text{app}}^\varepsilon + ip \cdot \nabla_z u_{\text{app}}^\varepsilon - z \cdot \langle \nabla V_2 \rangle_y u_{\text{app}}^\varepsilon \right) \\ &+ \varepsilon \left( i\partial_t u_{\text{app}}^\varepsilon + \frac{1}{2\varepsilon} \Delta_x u_{\text{app}}^\varepsilon + \frac{1}{2} \Delta_z u_{\text{app}}^\varepsilon - \frac{1}{2} z \cdot \langle \nabla^2 V_2 \rangle_y z u_{\text{app}}^\varepsilon \right) \\ &\left. + \tilde{r}_1^\varepsilon + \tilde{r}_2^\varepsilon \right), \end{aligned}$$

with  $\tilde{r}_j^\varepsilon = v_j u_{\text{app}}^\varepsilon$ ,  $j = 1, 2$ , and  $z$  is taken as  $z = (y - q(t)) / \sqrt{\varepsilon}$ . To cancel the first four terms in the  $\sqrt{\varepsilon}$  line, it is natural to require

$$\dot{q} = p, \quad q(0) = q_0, \quad \dot{p} = -\langle \nabla V_2 \rangle_y(t), \quad p(0) = p_0.$$

Now cancelling the first four terms in the first line of the right hand side yields

$$\dot{S}(t) = \frac{|p(t)|^2}{2} - \langle V_2 \rangle_y(t).$$

With these choices the above equation becomes

$$\begin{aligned} i\varepsilon\partial_t\psi_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x\psi_{\text{app}}^\varepsilon + \frac{\varepsilon^2}{2}\Delta_y\psi_{\text{app}}^\varepsilon - V(x, y)\psi_{\text{app}}^\varepsilon &= \frac{1}{\varepsilon^{d/4}}e^{ip(t)\cdot z/\sqrt{\varepsilon}+iS(t)/\varepsilon} \times \\ \left( i\varepsilon\partial_t u_{\text{app}}^\varepsilon + \frac{1}{2}\Delta_x u_{\text{app}}^\varepsilon + \frac{\varepsilon}{2}\Delta_z u_{\text{app}}^\varepsilon - \left( \frac{\varepsilon}{2} z \cdot \langle \nabla^2 V_2 \rangle_y z + V_1(x) + \langle W(x, \cdot) \rangle_y \right) u_{\text{app}}^\varepsilon \right. \\ \left. + \tilde{r}_1^\varepsilon + \tilde{r}_2^\varepsilon \right) \Big|_{z=\frac{y-q(t)}{\sqrt{\varepsilon}}}. \end{aligned}$$

We see that we can now define the approximate solution by:

$$(4.12) \quad i\varepsilon\partial_t\psi_1^\varepsilon + \frac{1}{2}\Delta_x\psi_1^\varepsilon = \left( V_1(x) + \langle W(x, \cdot) \rangle_y(t) \right) \psi_1^\varepsilon \quad ; \quad \psi_1^\varepsilon|_{t=0} = \varphi_0^x,$$

$$i\partial_t u_2 + \frac{1}{2}\Delta_z u_2 = \frac{1}{2} z \cdot \langle \nabla^2 V_2 \rangle_y(t) z u_2 \quad ; \quad u_2|_{t=0} = a.$$

Since the matrix  $\langle \nabla^2 V_2 \rangle_y(t)$  is real-valued, we infer that the  $L_z^2$ -norm of  $u_2(t)$  is independent of time, hence  $\|\psi_2(t)\|_{L_y^2} = \|u_2(t)\|_{L_z^2} = \|a\|_{L^2}$ . The equation in  $u_2$  is now nonlinear, and can be solved in  $\Sigma^1$ , since  $\nabla V_2$  is at most linear in its argument:  $u_2 \in C(\mathbb{R}; \Sigma_z^1)$ , as higher  $\Sigma^k$  regularity is propagated. To sum up, the approximation obtained by partial averaging reads:

$$\begin{aligned} \psi_{\text{app}}^\varepsilon(t, x, y) &= \frac{1}{\varepsilon^{d/4}}e^{ip(t)\cdot(y-q(t))/\varepsilon+iS(t)/\varepsilon} u_2 \left( t, \frac{y-q(t)}{\sqrt{\varepsilon}} \right) \psi_1^\varepsilon(t, x), \\ \text{with } i\varepsilon\partial_t\psi_1^\varepsilon + \frac{1}{2}\Delta_x\psi_1^\varepsilon &= \left( V_1(x) + \langle W(x, \cdot) \rangle_y(t) \right) \psi_1^\varepsilon \quad ; \quad \psi_1^\varepsilon|_{t=0} = \varphi_0^x, \\ \text{and } i\partial_t u_2 + \frac{1}{2}\Delta_z u_2 &= \frac{1}{2} z \cdot \langle \nabla^2 V_2 \rangle_y(t) z u_2 \quad ; \quad u_2|_{t=0} = a, \\ \dot{q} &= p, \quad q(0) = q_0, \quad \dot{p} = -\langle \nabla V_2 \rangle_y(t), \quad p(0) = p_0, \quad \dot{S}(t) = \frac{|p(t)|^2}{2} - \langle V_2 \rangle_y(t). \end{aligned}$$

Here again, if  $a$  is a Gaussian, then so is  $u_2$  and its width and center can be computed by solving ODEs (see [24, 7, 11]). Note also that, differently from the previous setting,  $u_2$  is now  $\varepsilon$  dependent via the quantity  $\langle \nabla^2 V_2 \rangle_y(t)$  (see (4.11)). However, this dependence is very weak since a Taylor expansion in (4.11) shows that  $u_2$  is close in any  $\Sigma^k$  norm from the solution of the equation

$$i\partial_t u + \frac{1}{2}\Delta_z u = \frac{1}{2} z \cdot \nabla^2 V_2(q(t)) z u \quad ; \quad u|_{t=0} = a.$$

For this reason, we do not keep memory of this  $\varepsilon$ -dependence and write  $u_2$ . By contrast, the  $\varepsilon$ -dependence of  $\psi_1^\varepsilon$  is strong since it implies oscillations features in time.

**4.5. The approximation results.** The main outcome of the above approximations can be stated as follows, and is proved in Section 4.6:

**Proposition 4.4.** *Let  $\psi^\varepsilon$  be the solution to (4.1)–(4.2), with  $\mathbf{g}^\varepsilon$  given by (4.3). Then with  $\psi_{\text{app}}^\varepsilon$  given either like in Section 4.3 or like in Section 4.4, there exist constants  $K_0, K_1$  independent of  $\varepsilon$  such that for all  $t \geq 0$ ,*

$$\|\psi^\varepsilon(t) - \psi_{\text{app}}^\varepsilon(t)\|_{L^2} \leq K_0 \left( \sqrt{\varepsilon} + \frac{\|\nabla_y W\|_{L^\infty}}{\sqrt{\varepsilon}} \right) e^{K_1 t}.$$

**Corollary 4.5.** *Assume  $\eta := \|\nabla_y W\|_{L^\infty} \ll \sqrt{\varepsilon}$ , then for all  $T > 0$ ,*

$$\sup_{t \in [0, T]} \|\psi^\varepsilon(t) - \psi_{\text{app}}^\varepsilon(t)\|_{L^2} = \mathcal{O} \left( \sqrt{\varepsilon} + \frac{\eta}{\sqrt{\varepsilon}} \right).$$

*Remark 4.6.* Using the same techniques as in Section 3.4, one can prove estimates on higher regularity norms, using  $\varepsilon$ -derivatives in  $y$  and standard ones in  $x$ . For example, if  $a \in \Sigma^4$ , then there exists  $K_0, K_1$  independent of  $\varepsilon$  such that

$$\begin{aligned} & \|\varepsilon \nabla_y \psi^\varepsilon(t) - \varepsilon \nabla_y \psi_{\text{app}}^\varepsilon(t)\|_{L^2} + \|y \psi^\varepsilon(t) - y \psi_{\text{app}}^\varepsilon(t)\|_{L^2} \\ & \leq K_0 \left( \sqrt{\varepsilon} \int_0^t e^{K_1 s} \|u_2(s)\|_{\Sigma^4} ds + \frac{\|\nabla_y W\|_{L^\infty}}{\sqrt{\varepsilon}} \int_0^t e^{K_1 s} \|u_2(s)\|_{\Sigma^2} ds \right). \end{aligned}$$

We refer to [8] (see also [7, Chapter 12]) for more detailed computations.

Note that, in both approximations, the evolution of  $u_2$  corresponds to the standard quadratic approximation. In particular, if  $a$  is Gaussian, then  $u_2$  is Gaussian at all time, and solving the equation in  $u_2$  amounts to solving ordinary differential equations. However, the equation (4.9) solved by  $\psi_1(t)$  is still quantum, however a reduction of the total space dimension of the quantum system has been made from  $n + d$  to  $n$ .

The latter equations (4.9) and (4.12) can be written as an adiabatic problem:

$$i\varepsilon \partial_t \psi_1^\varepsilon(t) = \mathfrak{h}(t) \psi_1^\varepsilon(t), \quad \psi_1^\varepsilon(0) = \varphi_0^x,$$

where  $\mathfrak{h}(t)$  is one of the time-dependent self-adjoint operators on  $L^2(\mathbb{R}^n)$

$$\mathfrak{h}_{\text{te}}(t) = -\frac{1}{2}\Delta + V_1(x) + W(x, q(t)) \quad \text{and} \quad \mathfrak{h}_{\text{pa}}(t) = -\frac{1}{2}\Delta + V_1(x) + \langle W(x, \cdot) \rangle_y(t).$$

We assume here that  $\mathfrak{h}(t)$  has a compact resolvent and thus, that its spectrum consists in a sequence of time-dependent eigenvalues

$$\Lambda_1(t) \leq \Lambda_2(t) \leq \dots \leq \Lambda_k(t) \xrightarrow[k \rightarrow +\infty]{} \infty.$$

We also assume that some eigenvalue  $\Lambda_j(t)$  is separated from the remainder of the spectrum for all  $t \in \mathbb{R}$  and that the initial datum  $\varphi_0^x$  is in the eigenspace of  $\mathfrak{h}(0)$  for the eigenvalue  $\Lambda_j(0)$ :

$$(4.13) \quad \mathfrak{h}(0) \varphi_0^x = \Lambda_j(0) \varphi_0^x.$$

Then adiabatic theory as developed by Kato [20] states that  $\psi_1(t)$  stays in the eigenspace of  $\Lambda_j(t)$  on finite time, up to a phase.

**Proposition 4.7** (Kato [20]). *Assume we have (4.13) and that  $\Lambda_j(0)$  is a simple eigenvalue of  $\mathfrak{h}(0)$  such that there exists  $\delta_0 > 0$  for which*

$$d(\{\Lambda_j(t)\}, \text{Sp}(\mathfrak{h}(t)) \setminus \{\Lambda_j(t)\}) \geq \delta_0.$$

*Denote by  $\Phi_j^x(t)$  a family of normalized eigenvectors of  $\mathfrak{h}(t)$  such that*

$$\Phi_j^x(0) = \varphi_0^x, \quad \langle \Phi_j^x(t), \partial_t \Phi_j^x(t) \rangle = 0.$$

Then, for all  $T > 0$ , there exists a constant  $C_T > 0$  such that

$$\left\| \psi_1^\varepsilon(t) - e^{-\frac{i}{\varepsilon} \int_0^t \Lambda_j(s) ds} \Phi_j^x(t, x) \right\|_{L_x^2} \leq C_T \varepsilon.$$

**Corollary 4.8.** *We then obtain the following approximate solution*

$$\psi_{\text{app}}^\varepsilon(t, x, y) = e^{-\frac{i}{\varepsilon} \int_0^t \Lambda_j(s) ds + \frac{i}{\varepsilon} S(t) + \frac{i}{\varepsilon} p(t) \cdot (y - q(t))} \Phi_j^x(t, x) u_2 \left( t, \frac{y - q(t)}{\sqrt{\varepsilon}} \right).$$

One then sees the difference with the Born-Oppenheimer approach recalled in Remark 4.3.

Of course, other methods can prove to be adequate for solving equation (4.12) depending on the specific context of the studied problem.

Let us now discuss the approximation of observables that we choose as acting only in the variable  $y$ . Due to the presence of the small parameter  $\varepsilon$ , we choose semiclassical observables and associate with  $b \in C_c^\infty(\mathbb{R}^{2d})$  ( $b$  smooth and compactly supported) the operator  $\text{op}_\varepsilon(b)$  whose action on functions  $f \in \mathcal{S}(\mathbb{R}^d)$  is given by

$$\text{op}_\varepsilon(b)f(y) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} b \left( \frac{y + y'}{2}, \varepsilon \xi \right) e^{i\varepsilon \cdot (y - y')} f(y') dy'.$$

As usual, the error estimate is better when quadratic observables are considered, instead of wave functions. More specifically, the error estimate from Proposition 4.4 is improved by a factor  $\sqrt{\varepsilon}$ . We apply Lemma 2.4 with now  $h = \varepsilon$ . Like in Section 3.5, we note that  $\psi_{\text{app}}^\varepsilon$  solves an equation of the form

$$i\varepsilon \partial_t \psi_{\text{app}}^\varepsilon = H_{\text{app}}^\varepsilon \psi_{\text{app}}^\varepsilon.$$

We prove the following result.

**Proposition 4.9.** *Let  $\psi^\varepsilon$  be the solution to (4.1)–(4.2), with  $\mathbf{g}^\varepsilon$  given by (4.3). Then with  $b \in C^\infty(\mathbb{R}^d)$  and  $\psi_{\text{app}}^\varepsilon$  given either like in Section 4.3 or like in Section 4.4, there exist a constant  $K$  independent of  $\varepsilon$  such that for all  $t \geq 0$ ,*

$$\left| \langle \text{op}_\varepsilon(b) \psi^\varepsilon(t), \psi^\varepsilon(t) \rangle - \langle \text{op}_\varepsilon(b) \psi_{\text{app}}^\varepsilon(t), \psi_{\text{app}}^\varepsilon(t) \rangle \right| \leq K t (\varepsilon + \|\nabla_y W\|_{L^\infty}).$$

*Remark 4.10.* Of course, we could have considered a mixed setting consisting of pseudodifferential operators as in Section 3.5 in the variable  $x$ , and semiclassical as above in the variable  $y$ . One would then obtain estimates mixing those of Proposition 3.10 and Proposition 4.9. Our interest here was in emphasizing the unmixed semiclassical aspect.

**4.6. Error estimates for the wave function.** In this section, we prove Proposition 4.4, and make comments on the constants  $K_0, K_1$ , which may be analyzed more explicitly in some cases.

4.6.1. *Approximation by partial Taylor expansion.* As we have seen before,

$$\begin{aligned} \|r_1^\varepsilon(t)\|_{L^2} &\leq \frac{\varepsilon^{3/2}}{6} \|\nabla^3 V_2\|_{L_y^\infty} \|\varphi_0^x\|_{L_x^2} \| |z|^3 u_2(t) \|_{L_x^2}, \\ \|r_2^\varepsilon(t)\|_{L^2} &\leq \sqrt{\varepsilon} \|\nabla_y W\|_{L^\infty} \|\varphi_0^x\|_{L_x^2} \|z u_2(t)\|_{L_x^2}. \end{aligned}$$

Lemma 2.3 then yields, with now  $h = \varepsilon$ ,

$$\begin{aligned} \|\psi^\varepsilon(t) - \psi_{\text{app}}^\varepsilon(t)\|_{L^2} &\leq \frac{\sqrt{\varepsilon}}{6} \|\nabla^3 V_2\|_{L_y^\infty} \|\varphi_0^x\|_{L_x^2} \int_0^t \| |z|^3 u_2(s) \|_{L_z^2} ds \\ &\quad + \frac{\|\nabla_y W\|_{L^\infty}}{\sqrt{\varepsilon}} \|\varphi_0^x\|_{L_x^2} \int_0^t \| |z| u_2(s) \|_{L_z^2} ds. \end{aligned}$$

According to the signature of  $\nabla^2 V_2(q(t))$ , the quantities  $\| |z|^3 u_2(s) \|_{L_z^2}$  and  $\| |z| u_2(s) \|_{L_z^2}$  may be bounded uniformly in  $s \geq 0$  or not. For instance, they are bounded if  $\nabla^2 V_2$  is uniformly positive definite, or at least uniformly positive definite along the trajectory  $q$ . On the other hand, we always have an exponential bound, even if it may not be sharp,

$$\| |z|^3 u_2(s) \|_{L_z^2} + \| |z| u_2(s) \|_{L_z^2} \leq C_0 e^{C_1 s},$$

for some constants  $C_0, C_1 > 0$ . This control is sharp in the case where  $\nabla^2 V_2$  is uniformly negative definite. See e.g. [7, Lemma 10.4] for a proof of the exponential control, and [7, Section 10.5] for a discussion on its optimality. In particular, for bounded time intervals, the (relative) error is small if  $\|\nabla_y W\|_{L^\infty} \ll \sqrt{\varepsilon} \ll 1$ .

*Remark 4.11.* If  $\nabla_y W$  is not bounded, e.g.  $\nabla_y W(x, y) = \eta \langle x \rangle^\gamma$ , then we can replace the previous error estimate with

$$\begin{aligned} \|\psi^\varepsilon(t) - \psi_{\text{app}}^\varepsilon(t)\|_{L^2} &\leq \frac{\sqrt{\varepsilon}}{6} \|\nabla^3 V_2\|_{L_y^\infty} \|\varphi_0^x\|_{L_x^2} \int_0^t \| |z|^3 u_2(s) \|_{L_z^2} ds \\ &\quad + \frac{\eta}{\sqrt{\varepsilon}} \int_0^t \|\langle x \rangle^\gamma \psi_1^\varepsilon(s)\|_{L_x^2} \| |z| u_2(s) \|_{L_z^2} ds. \end{aligned}$$

In other words, the cause for the unboundedness of  $\nabla_y W$  is transferred to a weight for  $\psi_1^\varepsilon$ . Similarly, if  $\nabla_y W$  is unbounded in  $y$ , we may change the weight in the terms  $\| |z|^k u_2 \|_{L_z^2}$ , after substituting  $y$  with  $q + z\sqrt{\varepsilon}$ .

4.6.2. *Approximation by partial averaging.* To estimate the size of  $\tilde{r}_1$  and  $\tilde{r}_2$ , we might argue again via Taylor expansion. Indeed, we have

$$\begin{aligned} \|a\|_{L^2}^2 \langle V_2 \rangle_y &= \int V_2(q(t) + \sqrt{\varepsilon}z) |u_2(t, z)|^2 dz \\ &= V_2(q(t)) + \sqrt{\varepsilon} \nabla V_2(q(t)) \cdot \int z |u_2(t, z)|^2 dz + r_3^\varepsilon(t), \end{aligned}$$

where

$$|r_3^\varepsilon(t)| \leq \frac{\varepsilon}{2} \|\nabla^2 V_2\|_{L^\infty} \| |z| u_2(t, z) \|_{L_z^2}^2.$$

Hence, we have for all averages  $f = V_2, \nabla V_2, \nabla^2 V_2, W(x, \cdot)$  that

$$\langle f \rangle_y(t) = f(q(t)) + \mathcal{O}(\sqrt{\varepsilon}),$$

where the error constant depends on moments of  $|u_2|^2$ . In particular, if  $u_2(0)$  is Gaussian, the odd moments of  $|u_2(t, z)|^2$  vanish, and the above estimate improves to  $\mathcal{O}(\varepsilon)$ . Hence, the  $L^2$ -norm of  $\tilde{r}_1$  is  $\mathcal{O}(\sqrt{\varepsilon})$  close to the  $L^2$ -norm of  $r_1$ , and the  $L^2$ -norm of  $\tilde{r}_2$  is  $\mathcal{O}(\eta\sqrt{\varepsilon})$ ,  $\eta = \|\nabla_y W\|_{L^\infty}$ , close to the  $L^2$ -norm of  $r_2$  (with each time an extra  $\sqrt{\varepsilon}$  gain in the above mentioned Gaussian case). In particular, the order of magnitude for the difference between exact and approximate solution is the same as in the previous subsection, only multiplicative constants are affected. We emphasize that the constants  $C_0$  and  $C_1$  from the previous subsection are in

general delicate to assess. On the other hand, in specific cases (typically when  $u_2$  is Gaussian and  $\nabla^2 V_2$  is known), they can be computed rather explicitly.

**4.7. Error estimates for quadratic observables.** The proof of Proposition 4.9 is discussed in the next two sections.

4.7.1. *Approximation by Taylor expansion.* In that case, the time-dependent Hamiltonian  $H_{\text{app}}^\varepsilon$  is  $H_{\text{app}}^\varepsilon = H_{\text{te}}^\varepsilon$  with

$$H_{\text{te}}^\varepsilon := -\frac{1}{2}\Delta_x - \frac{\varepsilon^2}{2}\Delta_y + V_1(x) + W(x, q) + V_2(q) + (y - q) \cdot \nabla V_2(q) + \frac{1}{2}\langle y - q, \nabla^2 V_2(q)(y - q) \rangle,$$

where  $q = q(t)$ . In particular, the difference  $H^\varepsilon - H_{\text{te}}^\varepsilon$  is a function,

$$H^\varepsilon - H_{\text{te}}^\varepsilon = W(x, y) - W(x, q) + V_2(y) - V_2(q) - (y - q) \cdot \nabla V_2(q) - \frac{1}{2}\langle y - q, \nabla^2 V_2(q)(y - q) \rangle =: \delta W(t, x, y).$$

In view of Lemma 2.4, if  $B = \text{op}_\varepsilon(b)$  with  $b \in C_c^\infty(\mathbb{R}^{2d})$ , it yields (a posteriori estimate)

$$|\langle \psi^\varepsilon(t), B\psi^\varepsilon(t) \rangle - \langle \psi_{\text{app}}^\varepsilon(t), B\psi_{\text{app}}^\varepsilon(t) \rangle| \leq \frac{1}{\varepsilon} \int_0^t |\rho^\varepsilon(t, x)| ds,$$

where

$$\rho^\varepsilon(t, s) = \langle \psi_{\text{app}}^\varepsilon(s), [B(t-s), \delta W(s)] \psi_{\text{app}}^\varepsilon(s) \rangle, \quad B(\sigma) = e^{i\frac{\sigma}{\varepsilon}H} B e^{-i\frac{\sigma}{\varepsilon}H}.$$

By Egorov Theorem [35],  $B(\sigma) = \varepsilon \text{op}_\varepsilon(b(\sigma))$  for a function  $b(\sigma) \in C_c^\infty(\mathbb{R}^d)$ . Therefore, by semiclassical calculus,

$$\frac{1}{i\varepsilon} [B(t-s), \delta W(s)] = \text{op}_\varepsilon(\{b(t-s), \delta W(s)\}) + \varepsilon^2 \text{op}_\varepsilon(r^\varepsilon(s, t)),$$

where  $\|\text{op}_\varepsilon(r^\varepsilon(s, t))\|_{\mathcal{L}(L^2)}$  is bounded uniformly in  $\varepsilon$ , whence the estimate of Proposition 4.9.

4.7.2. *Approximation by partial averaging.* In that case, the time-dependent Hamiltonian  $H_{\text{app}}^\varepsilon$  is  $H_{\text{app}}^\varepsilon = H_{\text{pa}}^\varepsilon$  with

$$H_{\text{pa}}^\varepsilon = -\frac{1}{2}\Delta_x - \frac{\varepsilon^2}{2}\Delta_y + V_1(x) + \langle W(x, \cdot) \rangle_y + \langle V_2 \rangle_y(t) + (y - q) \cdot \langle \nabla V_2 \rangle_y(t) + \frac{1}{2}(y - q) \cdot \langle \nabla^2 V_2 \rangle_y(t)(y - q),$$

where  $q = q(t)$ . In particular, as in the preceding case, the difference  $H^\varepsilon - H_{\text{pa}}^\varepsilon$  is a time-dependent function

$$H^\varepsilon - H_{\text{pa}}^\varepsilon = W(x, y) - \langle W(x, \cdot) \rangle_y + V_2(y) - \langle V_2 \rangle_y(t) - (y - q) \cdot \langle \nabla V_2 \rangle_y(t) - \frac{1}{2}(y - q) \cdot \langle \nabla^2 V_2 \rangle_y(t)(y - q) =: \widetilde{\delta W}(t, x, y),$$

and the arguments developed above also apply.

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#### APPENDIX A. COMMUTATOR ESTIMATE

We provide a direct proof of the commutator estimate used for our analysis of quadratic observables.

**Lemma A.1.** *Let  $N \geq 1$  and  $b = b(z, \zeta)$  be a smooth function on  $\mathbb{R}^{2N}$  satisfying the Hörmander growth condition (3.7). Let  $\delta W$  be a smooth function on  $\mathbb{R}^N$  with bounded derivatives. Then, there exist constants  $C_b > 0$  and  $M_N > 0$  such that*

$$\|[\text{op}(b), \delta W]\psi\|_{L^2} \leq C_b \left( \|\nabla(\delta W)\psi\|_{H^1} + \sum_{2 \leq |\alpha| \leq M_N} \|\partial^\alpha(\delta W)\|_\infty \|\psi\|_{L^2} \right)$$

for all  $\psi \in H^1(\mathbb{R}^N)$ .

*Proof.* We explicitly write the commutator as

$$\begin{aligned} & [\text{op}(b), \delta W]\psi(z) = \\ & (2\pi)^{-N} \int_{\mathbb{R}^{2N}} b\left(\frac{z+z'}{2}, \zeta\right) e^{i\zeta \cdot (z-z')} (\delta W(z') - \delta W(z)) \psi(z') d\zeta dz'. \end{aligned}$$

We Taylor expand the function  $\delta W(z)$  around the point  $z'$ , so that

$$\delta W(z) - \delta W(z') = \nabla(\delta W)(z') \cdot (z - z') + (z - z') \cdot \delta R_2(z, z')(z - z')$$

with

$$\delta R_2(z, z') = \int_0^1 (1 - \vartheta) \nabla^2(\delta W)(z' + \vartheta(z - z')) d\vartheta.$$

Corresponding to the above decomposition, we write

$$[\text{op}(b), \delta W]\psi(z) = f_1(z) + f_2(z)$$

and estimate the two summands separately. We observe that  $(z - z')e^{i\zeta \cdot (z-z')} = -i\nabla_\zeta e^{i\zeta \cdot (z-z')}$  and perform an integration by parts to obtain

$$\begin{aligned} & \int_{\mathbb{R}^{2N}} b\left(\frac{z+z'}{2}, \zeta\right) e^{i\zeta \cdot (z-z')} \nabla(\delta W)(z') \cdot (z - z') \psi(z') d\zeta dz' \\ & = i \int_{\mathbb{R}^{2N}} \nabla(\delta W)(z') \cdot \nabla_\zeta b\left(\frac{z+z'}{2}, \zeta\right) e^{i\zeta \cdot (z-z')} \psi(z') d\zeta dz' \end{aligned}$$

Therefore,

$$\|f_1\|_{L^2} \leq C_b \|\nabla(\delta W)\psi\|_{H^1},$$

where the constant  $C_b > 0$  depends on derivative bounds of the function  $b$ . For the remainder term of the above Taylor approximation we write

$$\begin{aligned} & \int_{\mathbb{R}^{2N}} b\left(\frac{z+z'}{2}, \zeta\right) e^{i\zeta \cdot (z-z')} (z - z') \cdot \delta R_2(z, z')(z - z') \psi(z') d\zeta dz' \\ & = \int_{\mathbb{R}^{2N}} \text{tr} \left( \delta R_2(z, z') \nabla_\zeta^2 b\left(\frac{z+z'}{2}, \zeta\right) \right) e^{i\zeta \cdot (z-z')} \psi(z') d\zeta dz', \end{aligned}$$

and obtain that

$$\|f_2\|_{L^2} \leq C'_b \sum_{2 \leq |\alpha| \leq M_N} \|\partial^\alpha(\delta W)\|_\infty \|\psi\|_{L^2},$$

where  $C'_b > 0$  depends on derivative bounds of  $b$ , while  $M_N > 0$  depends only on the dimension  $N$ .  $\square$

#### REFERENCES

- [1] S. ALINHAC AND P. GÉRARD, *Pseudo-differential operators and the Nash-Moser theorem*, vol. 82 of Graduate Studies in Mathematics, American Mathematical Society, Providence, RI, 2007. Translated from the 1991 French original by Stephen S. Wilson.
- [2] G. D. BILLING, *On the use of Ehrenfest's theorem in molecular scattering*, Chem. Phys. Lett., 100 (1983), p. 535.
- [3] H. BREUER AND F. PETRUCCIONE, *The theory of open quantum systems*, Oxford University Press, Oxford, 2002.
- [4] P. R. BUNKER AND P. JENSEN, *Molecular symmetry and spectroscopy*, NRC Research Press, Ottawa, 2006.
- [5] I. BURGHARDT AND G. PARLANT, *On the dynamics of coupled Bohmian and phase-space variables: A new hybrid quantum-classical approach*, J. Chem. Phys., 120 (2004), p. 3055.
- [6] E. CANCÈS AND C. LE BRIS, *On the time-dependent Hartree-Fock equations coupled with a classical nuclear dynamics*, Math. Models Methods Appl. Sci., 9 (1999), pp. 963–990.
- [7] R. CARLES, *Semi-classical analysis for nonlinear Schrödinger equations: WKB analysis, focal points, coherent states*, World Scientific Publishing Co. Pte. Ltd., Hackensack, NJ, 2nd ed., xiv-352 p. 2021.
- [8] R. CARLES AND C. FERMANIAN KAMMERER, *Nonlinear coherent states and Ehrenfest time for Schrödinger equations*, Commun. Math. Phys., 301 (2011), pp. 443–472.
- [9] J. CARO AND L. SALCEDO, *Impediments to mixing classical and quantum dynamics*, Phys. Rev. A, 60 (1999), p. 842.
- [10] R. D. COALSON AND M. KARPLUS, *Multidimensional variational gaussian wave packet dynamics with application to photodissociation spectroscopy*, The Journal of Chemical Physics, 93 (1990), pp. 3919–3930.
- [11] M. COMBESCURE AND D. ROBERT, *Coherent states and applications in mathematical physics*, Theoretical and Mathematical Physics, Springer, Dordrecht, 2012.
- [12] J. B. DELOS, W. R. THORSON, AND S. K. KNUDSON, *Semiclassical theory of inelastic collisions. I. Classical picture and semiclassical formulation*, Phys. Rev. A, 6 (1972), p. 709.
- [13] P. A. M. DIRAC, *Note on exchange phenomena in the Thomas atom*, Proc. Cambridge Philos. Soc., 26 (1930), p. 376.
- [14] R. B. GERBER, V. BUCH, AND M. A. RATNER, *Time-dependent self-consistent field approximation for intramolecular energy transfer: I. Formulation and application to dissociation of van der Waals molecules*, J. Chem. Phys., 77 (1982), p. 3022.
- [15] E. GINDENSPERGER, C. MEIER, AND J. A. BESWICK, *Mixing quantum and classical dynamics using Bohmian trajectories*, J. Chem. Phys., 113 (2000), p. 9369.
- [16] M. GRUEBELE, *Quantum dynamics and control of vibrational dephasing*, J. Phys.: Condens. Matter, 16 (2004).
- [17] B. JÄGER, R. HELLMANN, E. BICH, AND E. VOGEL, *State-of-the-art ab initio potential energy curve for the krypton atom pair and thermophysical properties of dilute krypton gas*, J. Chem. Phys., 144 (2016), p. 114304.
- [18] S. JIN, C. SPARBER, AND Z. ZHOU, *On the classical limit of a time-dependent self-consistent field system: analysis and computation*, Kinet. Relat. Models, 10 (2017), pp. 263–298.
- [19] R. KAPRAL AND G. CICCOTTI, *Mixed quantum-classical dynamics*, J. Chem. Phys., 110 (1999), p. 8919.
- [20] T. KATO, *On the adiabatic theorem of quantum mechanics*, J. Phys. Soc. Japan, 5 (1950), pp. 435–439.
- [21] C. LASSER AND C. LUBICH, *Computing quantum dynamics in the semiclassical regime*, Acta Numer., 29 (2020), pp. 229–401.
- [22] A. M. LEVINE, M. SHAPIRO, AND E. POLLAK, *Hamiltonian theory for vibrational dephasing rates of small molecules in liquids*, J. Chem. Phys., 88 (1988), p. 1959.

- [23] C. LUBICH, *On variational approximations in quantum molecular dynamics*, Math. Comp., 74 (2005), pp. 765–779.
- [24] ———, *From quantum to classical molecular dynamics: reduced models and numerical analysis*, Zurich Lectures in Advanced Mathematics, European Mathematical Society (EMS), Zürich, 2008.
- [25] C. C. MARTENS AND J.-Y. FANG, *Semiclassical-limit molecular dynamics on multiple electronic surfaces*, J. Chem. Phys., 106 (1994), p. 4918.
- [26] A. MARTINEZ AND V. SORDONI, *Twisted pseudodifferential calculus and application to the quantum evolution of molecules*, Mem. Amer. Math. Soc., 200 (2009), pp. vi+82.
- [27] M. REED AND B. SIMON, *Methods of modern mathematical physics. II. Fourier analysis, self-adjointness*, Academic Press [Harcourt Brace Jovanovich Publishers], New York, 1975.
- [28] S. RÖMER AND I. BURGHARDT, *Towards a variational formulation of mixed quantum-classical molecular dynamics*, Molecular Physics, 111 (2013), pp. 3618–3624.
- [29] L. L. SALCEDO, *Statistical consistency of quantum-classical hybrids*, Phys. Rev. A, 85 (2012), p. 022127.
- [30] W. STRUYVE, *Semi-classical approximations based on Bohmian mechanics*, Int. J. Mod. Phys. A, 35 (2020), p. 2050070.
- [31] D. R. TERNO, *Inconsistency of quantum-classical dynamics, and what it implies*, Found. Phys., 36 (2006), p. 102.
- [32] S. TEUFEL, *Adiabatic perturbation theory in quantum dynamics*, vol. 1821 of Lecture Notes in Mathematics, Springer-Verlag, Berlin, 2003.
- [33] H. WEI, R. J. LE ROY, R. WHEATLEY, AND W. J. MEATH, *A reliable new three-dimensional potential energy surface for H<sub>2</sub>-Kr*, J. Chem. Phys., 122 (2005), p. 084321.
- [34] U. WEISS, *Quantum dissipative systems, 4th Ed.*, World Scientific, Singapore, 2012.
- [35] M. ZWORSKI, *Semiclassical analysis*, vol. 138 of Graduate Studies in Mathematics, American Mathematical Society, Providence, RI, 2012.

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