

# OBSERVABLE ERROR BOUNDS OF THE TIME-SPLITTING SCHEME FOR QUANTUM-CLASSICAL MOLECULAR DYNAMICS\*

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**Abstract.** Quantum-classical molecular dynamics, as a partial classical limit of the full quantum Schrödinger equation, is a widely used framework for quantum molecular dynamics. The underlying equations are nonlinear in nature, containing a quantum part (representing the electrons) and a classical part (standing for the nuclei). An accurate simulation of the wave function typically requires a time step comparable to the rescaled Planck constant  $\hbar$ , resulting in a formidable cost when  $\hbar \ll 1$ . We prove an additive observable error bound of Schwartz observables for the proposed time-splitting schemes based on semiclassical analysis, which decreases as  $\hbar$  becomes smaller. Furthermore, we establish a uniform-in- $\hbar$  observable error bound, which allows for an  $\mathcal{O}(1)$  time step to accurately capture the physical observable regardless of the size of  $\hbar$ . Numerical results verify our estimates.

**Key words.** quantum-classical molecular dynamics, observable error bounds, Weyl quantization, semiclassical analysis

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**1. Introduction.** Molecular dynamics has been a long-standing topic in computational chemistry, physics, and biology. The ultimate goal is to solve the many-body Schrödinger equation governed by the molecular Hamiltonian describing both electrons and nuclei, direct simulation of which is known to be a formidable challenge due to the curse of dimensionality. Mixed quantum-classical approaches are thus proposed to tackle this issue. These methods treat the nuclei and electrons separately, while a mathematically certain underlying tensor product ansatz is typically assumed in deriving the coupled system. Such an approximation fully exploits the multiscale structure, that is, the scale separation between the fast-varying electrons and the slowly evolved nuclei. Some mixed approaches remain fully on the quantum level, such as time-dependent self-consistent field methods [9, 37, 38, 45, 54] and other time-dependent mean-field approximations [2, 35, 51, 52, 62, 64], while others combine together with classical or semiclassical levels, such as the Born–Oppenheimer (or adiabatic) approximation, the mean-field type treatments [49, 50], and trajectory based surface hopping algorithms [25, 26, 27, 28, 40, 43, 59, 60, 61]. In the latter approaches, the nuclear degrees of freedom are described classically by a Newtonian flow with the forces acting on the nuclei as feedback of the electronic structures calculated “on-the-fly” (for detailed reviews, see, e.g., [8, 47, 48, 60]).

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We focus on the quantum-classical molecular dynamics (QCMD) as proposed and justified in [11], also sometimes referred to as the Ehrenfest molecular dynamics [11, 12, 22, 60] whose equations of motion are given by a Newtonian flow (for the nuclei) coupled with a Schrödinger equation (for the electrons):

$$(1.1) \quad i\hbar \partial_t \psi^h = -\frac{\hbar^2}{2} \Delta_x \psi^h + V(x, y(t)) \psi^h := H_t^h \psi^h, \quad \psi^h(0, x) = \psi_0^h(x),$$

$$(1.2) \quad \ddot{y}(t) = -\nabla_y V_e^E(y(t)), \quad y(0) = y_0, \quad y'(0) = v_0,$$

where  $\psi^h(t, x) \in C(\mathbb{R}_t^+, \mathbb{R}_x^d)$  is the wave function of the electrons,  $\hbar$  is the rescaled Planck constant,  $y(t) \in C(\mathbb{R}_t^+)$  is the position of the nucleus,  $V$  is a given potential of the whole system, and

$$V_e^E = \int_{\mathbb{R}^d} V(x, y) |\psi^h(t, x)|^2 dx$$

is often called the Ehrenfest potential, given by the quantum dynamics of the electrons, which vividly explains the spirit of the “on-the-fly” simulation.

QCMD is typically seen as an extension of the Born–Oppenheimer approximation to the time-dependent situation that serves as a popular tool in chemical and engineering applications (see, e.g., textbooks [7, 48, 62]). The basic assumption is that the masses between particles differ significantly, which leads to the heuristics that the heavier particle can be modeled classically while the lighter ones remain quantum. Interestingly, such heuristics can be justified on a rigorous level. Known as an approximation of full many-body quantum dynamics, it can be derived mathematically as a partial classical limit of the full molecular Schrödinger equation by combining the separation of the full wave function and short wave asymptotics [11].

The numerical difficulties associated with this type of semiclassical Schrödinger equation lie in the oscillations of order  $1/h$  in both time and spatial discretizations [5], which need to be resolved for an accurate simulation of the wave function. However, it has been observed and formally demonstrated in [5], using the Wigner analysis, that for the linear Schrödinger equation with scalar potentials under suitable conditions, a time-splitting spectral method can still capture the correct *physical observables*—not the wave function—when the time step is much larger than  $\hbar$ , although spatially one still needs to resolve  $\hbar$ . Such interesting observations have been recently justified on a rigorous level by breakthrough results via different strategies: [31] investigates the time-splitting algorithm for the von Neumann equation by measuring the Wasserstein distance between the Husimi functions of the approximate and the exact quantum density operators, and achieves an observable error bound of  $\mathcal{O}(\Delta t^2 + \hbar^{1/2})$  for the second-order splitting; and, more recently, a direct observable expectation comparison is considered in [39], resulting in a tighter bound of  $\mathcal{O}(\Delta t^2 + \hbar^2)$ .

For nonlinear Schrödinger equations, nevertheless, one still needs to resolve  $\hbar$  temporally in general, as was numerically demonstrated in [6]. However, for the QCMD and other Ehrenfest models, numerical evidence presented in [24, 36] suggests the existence of a temporal meshing strategy independent of  $\hbar$  for the physical observables associated with these nonlinear Schrödinger type systems. Though heuristic arguments based on the Wigner transform have been provided in both works, no rigorous error analysis was carried out for such nonlinear problems, which is the focus of this work.

**Contribution:** At the continuous level, though the semiclassical limit of the Ehrenfest dynamics has been rigorously proved in [36] in the weak-\* sense, the distance between the Ehrenfest dynamics and its semiclassical limit has not been understood.

As the *first contribution*, we establish the distance of the observable expectations between such two dynamics and show that the error is  $\mathcal{O}(h^2)$ . This Egorov-type estimate also holds when the momentum part or the potential part in the electron dynamics is absent, which serves as a building block for the numerical analysis with the time-splitting scheme.

As the *second contribution* of this paper, we prove the observable error bounds of this momentum-potential type operator splitting scheme, as proposed in [24, 36], for the QCMD model, which extends the linear result in [39] to a weakly *nonlinear* Schrödinger system. Inspired by [39], the proof utilizes the observable error bounds of the QCMD model on the continuous level. Different from the observable estimates obtained from the wave function that exhibits  $h$  scaling in the denominator, this new observable error bound, taking advantage of its semiclassical limit, removes  $h$  in the denominator and exhibits an additive scaling instead. Moreover, combining two types of estimates, we present a *uniform* (in  $h$ ) observable error bound of the second-order time-splitting scheme for the QCMD, which serves as our *third contribution*.

**Organization:** The rest of this paper is organized as follows. In section 2 we introduce some preliminaries of the semiclassical descriptions for the QCMD model, including the Wigner transform and Husimi functions, and two important lemmas that are used in later proofs. We then derive the observable error bounds between the QCMD and its semiclassical limit in section 3. Section 4 discusses the time-splitting scheme for the QCMD and analyzes the numerical errors of the observable resulting from the time-splitting strategy. Finally, we provide the numerical evidence supporting our analytical results in section 5.

**2. Preliminaries.** In this section we first introduce the tools to describe the semiclassical limit of QCMD and then some preliminary lemmas used in later proofs.

**2.1. The Wigner transform and the semiclassical limit.** We revisit the Wigner transform [30, 41, 63] and the semiclassical limit of the QCMD (or the Ehrenfest dynamics) shown in [36]. Let us first recall that the  $h$ -scaled Wigner transform [30, 41, 46] associated with any continuously parametrized family  $\psi^h \equiv \{\psi^h\}_{0 \leq h \leq 1} \in L^2(\mathbb{R}^d)$  is given by

$$w^h[\psi^h](x, \xi) = \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \psi^h\left(x - \frac{h}{2}y\right) \overline{\psi^h}\left(x + \frac{h}{2}y\right) e^{i\xi \cdot y} dy.$$

By Plancherel's theorem and a change of variables one easily finds

$$\|w^h[\psi^h]\|_{L^2(\mathbb{R}^{2d})} = \frac{1}{(2\pi)^{d/2} h^d} \|\psi^h\|_{L^2(\mathbb{R}^d)}^2.$$

After Wigner transforming the Schrödinger equation, one finds that  $w^h(t, x, \xi) := w^h[\psi^h](t, x, \xi)$  satisfies the following nonlocal kinetic equation (see, e.g., [41]),

$$\begin{aligned} \partial_t w^h + \xi \cdot \nabla_x w^h + \Theta^h[V^h] w^h &= 0, \quad w^h(0, x, \xi) = w^h[\psi_0^h], \\ \ddot{y}(t) &= - \int_{\mathbb{R}} \nabla_y V(x, y(t)) w^h(t, x, \xi) dx d\xi, \quad y(0) = y_0, \quad y'(0) = v_0, \end{aligned}$$

where  $\Theta^h[V^h]$  is the pseudodifferential operator

$$(2.1) \quad \begin{aligned} (\Theta^h[V^h] w^h)(t, x, \xi) &= \frac{i}{h(2\pi)^d} \int_{\mathbb{R}^d} \left( V^h\left(x + \frac{h}{2}z, y(t)\right) \right. \\ &\quad \left. - V^h\left(x - \frac{h}{2}z, y(t)\right) \right) \widehat{w}^h(t, x, z) e^{iz \cdot \xi} dz \end{aligned}$$

with  $\widehat{w}^h$  denoting the Fourier transformation of  $w^h$  with respect to the second variable only. It was shown in [36] that one can pass the limit  $h \rightarrow 0_+$  and get its semiclassical limit consisting of a Liouville equation coupled with a Newtonian flow,

$$(2.2) \quad \begin{aligned} \partial_t \mu + \xi \cdot \nabla_x \mu - \nabla_x V(x, y(t)) \cdot \nabla_\xi \mu &= 0, \\ \ddot{y}(t) &= - \int_{\mathbb{R}^d} \nabla_y V(x, y(t)) \mu(t, x, \xi) dx d\xi, \end{aligned}$$

where  $w^h \xrightarrow{h \rightarrow 0_+} \mu$  and the convergence is in the weak-\* sense. Because the measure converges in the weak-\* topology, a direct evaluation of the distance between  $w^h$  and its limit  $\mu$  becomes a nontrivial task. A framework based on the Wasserstein distance has been proposed for the *linear* von Neumann equation in [31].

**2.2. The Weyl quantization and Husimi functions.** An alternate way to view the connection between quantum dynamics and its classical analog is through the Weyl quantization. For a Schwartz function  $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ ,  $a = a(x, \xi)$ , we define its Weyl quantization [67] to be the operator  $\text{op}(a)$  acting on  $\psi \in L^2(\mathbb{R}^d)$  by the formula

$$(2.3) \quad \text{op}(a)\psi = \frac{1}{(2\pi h)^d} \int_{\mathbb{R}^{2d}} a\left(\frac{x+y}{2}, \xi\right) e^{i\xi \cdot (x-y)/h} \psi(y) d\xi dy,$$

so that the expectation values of an operator  $A = \text{op}(a)$  can be written in terms of the Wigner transform as

$$(2.4) \quad \langle A \rangle_\psi = \langle \psi | A | \psi \rangle = \int_{\mathbb{R}^{2d}} a(x, \xi) w^h[\psi](x, \xi) dx d\xi.$$

One defines the Husimi function  $\sigma^h[\psi] : \mathbb{R}^{2d} \rightarrow [0, \infty)$  as

$$(2.5) \quad \sigma^h[\psi](x, \xi) = (w^h[g_{(0,0)}] * w^h[\psi])(z) = \frac{1}{(2\pi h)^d} |\langle g_{(x,\xi)} | \psi \rangle|^2,$$

where  $g_{(x,\xi)}(\cdot)$  is the Gaussian wave packet (or the coherent state)

$$g_{(x,\xi)}(y) = \frac{1}{(\pi h)^{d/4}} \exp\left(-\frac{1}{2h}|y-x|^2 + \frac{i}{h}\xi \cdot (y-x)\right).$$

We remark that the Husimi function of  $\psi$  with  $\|\psi\| = 1$  is in fact a nonnegative probability density on the phase space, and is the modulus squared of the so-called Fourier–Bros–Iagolnitzer transform acting on  $\psi$ . The expectation of  $A = \text{op}(a)$  can be described by the Husimi function,

$$\langle A \rangle_\psi = \int_{\mathbb{R}^{2d}} \left(a - \frac{h}{4}\Delta a\right)(x, \xi) \sigma^h[\psi](x, \xi) dx d\xi + \mathcal{O}(h^2),$$

as detailed in Lemma 2.2.

**2.3. Important lemmas.** We review two lemmas to be used in the proof of the paper. The first lemma is the stationary phase [67], which is used in the proof of Theorem 3.1.

**LEMMA 2.1** (stationary phase lemma). *Assume that  $a \in C_c^\infty(\mathbb{R}^{2d})$ . Then for each positive integer  $N$ , we have*

$$\int_{\mathbb{R}^{2d}} e^{\frac{i}{h}\langle x, \xi \rangle} a(x, \xi) dx d\xi = (2\pi h)^d \left( \sum_{k=0}^{N-1} \frac{h^k}{k!} \left( \frac{\langle \nabla_x, \nabla_\xi \rangle}{i} \right)^k a(0, 0) + \mathcal{O}(h^N) \right)$$

as  $h \rightarrow 0$ , where  $\langle \cdot, \cdot \rangle$  stands for the usual inner product.

The second lemma describes the approximation error of the observable expectations in terms of the Husimi function [39, Theorem 6.11], which is used in the proof of Corollary 3.3.

**LEMMA 2.2** (expectations in terms of Husimi functions). *Assume the function  $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$  that defines the observable  $A = \text{op}(a)$  is smooth and bounded together with all its derivatives. Then, for all  $\psi \in L^2(\mathbb{R})$ , we have*

$$\left| \langle A \rangle_\psi - \int_{\mathbb{R}^{2d}} \left( a(x, \xi) - \frac{h}{4} \Delta a(x, \xi) \right) \sigma^h[\psi](x, \xi) dx d\xi \right| \leq C h^2 \|\psi\|_{L^2(\mathbb{R}^d)}^2, \quad 0 < t \leq T,$$

where  $C \geq 0$  depends only on the derivative bounds of  $a$ .

**3. Error between its limit in observables.** We quantify the distance between the QCMD and its semiclassical limit by a direct investigation on the observable expectations. When the Hamiltonian is linear and time independent, the observable bounds (also called Egorov's theorem) is a consequence of the semiclassical expansion and commutator estimates. On an intuitive level, one has

$$\partial_t \left( e^{iHt/h} A e^{-iHt/h} \right) = e^{iHt/h} \frac{i}{h} [H, A] e^{-iHt/h},$$

and the commutator follows the semiclassical expansion (see, e.g., [67, Theorem 4.12] and [39, Proposition 6.2])

$$(3.1) \quad [H, A] = \frac{h}{i} \text{op}(\{\mathcal{H}, a\}) + \mathcal{O}(h^3)$$

for any  $H$  and  $A$  that are Weyl quantizations of the phase space functions (symbols)  $\mathcal{H}(x, p)$  and  $a(x, p)$ , respectively, where  $[H, A] = HA - AH$  is the commutator of the operators  $H$  and  $A$ , and  $\{\mathcal{H}, a\} = \nabla_p \mathcal{H} \cdot \nabla_x a - \nabla_x \mathcal{H} \cdot \nabla_p a$  is the Poisson bracket of the corresponding functions  $\mathcal{H}$  and  $a$ . Note that here we follow the notation of [67] that is of the opposite sign compared to that in [39]. More precisely, for the linear case (see, e.g., [39, Proposition 6.3] and [67, Section 11]), we can estimate in terms of the operators,

$$\begin{aligned} e^{iHt/h} A e^{-iHt/h} - \text{op}(a \circ \Xi^t) &= \int_0^t \partial_s \left( e^{iHs/h} \text{op}(a \circ \Xi^{t-s}) e^{-iHs/h} \right) ds \\ &= \int_0^t e^{iHs/h} \left( \frac{i}{h} [\mathcal{H}, \text{op}(a \circ \Xi^{t-s})] \right. \\ &\quad \left. - \text{op}(\{\mathcal{H}, a \circ \Xi^{t-s}\}) \right) e^{-iHs/h} ds, \end{aligned}$$

where we use  $\Xi^t$  to denote the flow governed by the Hamiltonian  $\mathcal{H}$  at time  $t$ . Thanks to (3.1), the difference between  $e^{iHt/h} A e^{-iHt/h}$  and its semiclassical limit  $\text{op}(a \circ \Xi^t)$  in the operator norm from  $L^2$  to  $L^2$  is bounded by  $\mathcal{O}(h^2)$ . However, when  $H$  depends on the wave function itself, it is not sufficient to deal with linear operators. Our goal here is also to perform the semiclassical expansion, but due to the time dependence in the potential and nonlinearity of the problem, we consider such an expansion in terms of the wave function (that is, the solution), instead of the operators. One certainly does not hope such results hold for arbitrary nonlinearity. Fortunately, here the observable expectations still exhibit the semiclassical behavior thanks to the structure of the QCMD system.

In order to discuss the classical counterpart of the QCMD system, we consider the following ODE with its flow map denoted as  $\Phi^t$ ,

$$(3.2) \quad \begin{cases} \dot{x} = \xi, \\ \dot{\xi} = -\nabla_x V(x, y), \\ \dot{y} = v, \\ \dot{v} = -\nabla_y V(x, y), \end{cases}$$

which can be viewed as the Lagrangian description of the semiclassical limit (2.2). In other words,  $\Phi^t$  is governed by the total Hamiltonian  $\mathcal{H}_0 := \frac{|\xi|^2}{2} + \frac{|v|^2}{2} + V(x, y)$ . We are now ready to state and prove the Egorov-type theorem for the QCMD.

**THEOREM 3.1** (Egorov's theorem for the QCMD). *Let  $a(x, \xi)$  be a Schwartz function that defines the observable  $A = \text{op}(a)$ , and the potential function  $V$  be smooth with its derivatives of order  $\geq 2$  all bounded. Then there exists some constant  $C \geq 0$  depending on the derivative bounds of  $a$  and independent of  $h$  and  $\psi_0$  so that*

$$|\langle A \rangle_{\psi^h(t, \cdot)} - \langle \text{op}(a \circ \Phi^t) \rangle_{\psi_0^h(\cdot)}| \leq Cth^2.$$

*Proof.* In this proof, we drop the superscript  $h$  in the wave function  $\psi^h$  for notational simplicity. One starts by looking at the integrands of the expectations,

$$\begin{aligned} \bar{\psi}(t, x) \text{op}(a) \psi(t, x) - \bar{\psi}_0(x) \text{op}(a \circ \Phi^t) \psi_0(x) &= \int_0^t \frac{d}{ds} (\bar{\psi}(s) \text{op}(a \circ \Phi^{t-s}) \psi(s, x)) ds \\ &= \int_0^t \partial_s \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) \psi(s, x) + \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) \partial_s \psi(s, x) \\ &\quad - \bar{\psi}(s, x) \partial_t \text{op}(a \circ \Phi^{t-s}) \psi(s, x) ds \\ &= \int_0^t \frac{i\hbar}{2} (-\Delta_x \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) \psi(s, x) + \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) \Delta_x \psi(s, x)) ds \\ &\quad + \int_0^t \frac{i}{\hbar} (V(x, y(s)) \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) \psi(s, x) \\ &\quad - \bar{\psi}(s, x) \text{op}(a \circ \Phi^{t-s}) V(x, y(s)) \psi(s, x)) ds \\ &\quad + \int_0^t \bar{\psi}(s, x) \text{op}(\{a \circ \Phi^{t-s}, \mathcal{H}_0\}) \psi(s, x) ds := \int_0^t (\text{I}) + (\text{II}) + (\text{III}) ds, \end{aligned}$$

where we use (1.1) and the fact that  $\partial_t \text{op}(a \circ \Phi^{t-s}) = -\text{op}(\{a \circ \Phi^{t-s}, \mathcal{H}_0\})$ . Denote  $b = a \circ \Phi^{t-s}$ , one then computes the terms one by one. Starting from the momentum terms, we have

$$\Delta_x \bar{\psi}(s, x) \text{op}(b) \psi(s, x) = (2\pi\hbar)^{-d} \int_{\mathbb{R}^{2d}} \Delta_x \bar{\psi}(s, x) b\left(\frac{x+q}{2}, p\right) e^{ip \cdot (x-q)/\hbar} \psi(s, q) dq dp$$

and it follows from the definition of Weyl quantization and integration by parts that

$$\begin{aligned} &\bar{\psi}(s, x) \text{op}(b) \Delta_x \psi(s, x) \\ &= (2\pi\hbar)^{-d} \int_{\mathbb{R}^{2d}} \bar{\psi}(s, x) b\left(\frac{x+q}{2}, p\right) e^{ip \cdot (x-q)/\hbar} \Delta_q \psi(s, q) dq dp \\ &= (2\pi\hbar)^{-d} \int_{\mathbb{R}^{2d}} \bar{\psi}(s, x) \Delta_q \left( b\left(\frac{x+q}{2}, p\right) e^{ip \cdot (x-q)/\hbar} \right) \psi(s, q) dq dp. \end{aligned}$$

Following the chain rule by changing the differentiation variable from  $q$  to  $x$ , one has

$$\begin{aligned} & \bar{\psi}(s, x) \operatorname{op}(b) \Delta_x \psi(s, x) \\ &= (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} \bar{\psi}(s, x) \Delta_x \left( b \left( \frac{x+q}{2}, p \right) e^{ip \cdot (x-q)/h} \right) \psi(s, q) dq dp \\ & \quad + (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} \frac{2i}{h} \bar{\psi}(s, x) \nabla_q b \left( \frac{x+q}{2}, p \right) \cdot p e^{ip \cdot (x-q)/h} \psi(s, q) dq dp, \end{aligned}$$

where the fact that  $\nabla_q b \left( \frac{x+q}{2}, p \right) = \nabla_x b \left( \frac{x+q}{2}, p \right)$  is also used. Therefore, one has

$$\begin{aligned} \int (\text{I}) dx &= -\frac{ih}{2} (2\pi h)^{-d} \int_{\mathbb{R}^{3d}} \Delta_x \bar{\psi}(s, x) b \left( \frac{x+q}{2}, p \right) e^{ip \cdot (x-q)/h} \psi(s, q) dq dp dx \\ & \quad + \frac{ih}{2} (2\pi h)^{-d} \int_{\mathbb{R}^{3d}} \bar{\psi}(s, x) \Delta_x \left( b \left( \frac{x+q}{2}, p \right) e^{ip \cdot (x-q)/h} \right) \psi(s, q) dq dp dx \\ & \quad + (2\pi h)^{-d} \int_{\mathbb{R}^{3d}} \bar{\psi}(s, x) \nabla_q b \left( \frac{x+q}{2}, p \right) \cdot p e^{ip \cdot (x-q)/h} \psi(s, q) dq dp dx \\ &= \int_{\mathbb{R}^d} \bar{\psi}(s, x) \operatorname{op}(\nabla_x b \cdot p) \psi(s, x) dx, \end{aligned}$$

where the first two terms cancel thanks to the integration by parts in  $x$ . Note that there is no approximation error in the momentum terms and this is essentially because the Laplacian is a quadratic observable.

Next we compute the potential terms. Since  $V(x, y)$  is not necessarily quadratic, one expects some approximation errors entering the calculations:

$$\begin{aligned} (\text{II}) &= \frac{i}{h} (V(x, y(s)) \bar{\psi}(s, x) \operatorname{op}(b) \psi(s, x) - \bar{\psi}(s, x) \operatorname{op}(b) V(x, y(s)) \psi(s, x)) \\ &= \frac{i}{h} (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} b \left( \frac{x+q}{2}, p \right) (V(x, y(s)) \\ & \quad - V(q, y(s))) e^{ip \cdot (x-q)/h} \bar{\psi}(s, x) \psi(s, q) dq dp \\ &= -\frac{i}{h} (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} b \left( x - \frac{q}{2}, p \right) (V(x, y(s)) \\ & \quad - V(x - q, y(s))) e^{ip \cdot q/h} \bar{\psi}(s, x) \psi(s, x - q) dq dp. \end{aligned}$$

Apply Lemma 2.1, one has

$$\begin{aligned} (3.3) \quad -(\text{II}) &= \bar{\psi}(s, x) \langle \nabla_q, \nabla_p \rangle \left( b \left( x - \frac{q}{2}, p \right) (V(x, y(s)) - V(x - q, y(s))) \psi(s, x - q) \right) \Big|_{q=p=0} \\ & \quad + \bar{\psi}(s, x) \frac{h}{2i} \langle \nabla_q, \nabla_p \rangle^2 \left( b \left( x - \frac{q}{2}, p \right) (V(x, y(s)) \right. \\ & \quad \left. - V(x - q, y(s))) \psi(s, x - q) \right) \Big|_{q=p=0} + \mathcal{O}(h^2) \\ &= \bar{\psi}(s, x) \nabla_p b(x, 0) \cdot \nabla_x V(x, y(s)) \psi(s, x) + \mathcal{O}(h^2) \\ & \quad + \bar{\psi}(s, x) \frac{h}{2i} \langle \nabla_q, \nabla_p \rangle^2 \left( b \left( x - \frac{q}{2}, p \right) (V(x, y(s)) \right. \\ & \quad \left. - V(x - q, y(s))) \psi(s, x - q) \right) \Big|_{q=p=0}. \end{aligned}$$

On the other hand,

$$\begin{aligned}
& \text{op}(\nabla_p b \cdot \nabla_x V) \psi(s, x) \\
&= (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} \nabla_p b \left( \frac{x+q}{2}, p \right) \nabla_x V \left( \frac{x+q}{2}, y(s) \right) e^{ip \cdot (x-q)/h} \psi(s, q) dq dp \\
&= (2\pi h)^{-d} \int_{\mathbb{R}^{2d}} \nabla_p b \left( x - \frac{q}{2}, p \right) \nabla_x V \left( x - \frac{q}{2}, y(s) \right) e^{ip \cdot q/h} \psi(s, x-q) dq dp \\
&= \nabla_p b(x, 0) \nabla_x V(x, y(s)) \psi(s, x) \\
&\quad + \frac{h}{i} \nabla_q \cdot \left( \nabla_p \nabla_p b \left( x - \frac{q}{2}, p \right) \nabla_x V \left( x - \frac{q}{2}, y(s) \right) \psi(s, x-q) \right) |_{q=p=0} + \mathcal{O}(h^2),
\end{aligned}$$

where the stationary phase lemma is applied in the last line. One can check that  $\bar{\psi}(s, x) \text{op}(\nabla_p b \cdot \nabla_x V) \psi(s, x)$  and (II) share the same  $\mathcal{O}(1)$  and  $\mathcal{O}(h)$  terms and, hence,

$$(3.4) \quad (\text{II}) = -\bar{\psi}(s, x) \text{op}(\nabla_p b \cdot \nabla_x V) \psi(s, x) + \mathcal{O}(h^2).$$

Combining (3.3) and (3.4), we have

$$\begin{aligned}
& \int_{\mathbb{R}} (\text{I}) + (\text{II}) + (\text{III}) dx \\
&= \int_{\mathbb{R}^d} \bar{\psi}(s, x) (\text{op}(\nabla_x b \cdot p) - \text{op}(\nabla_p b \cdot \nabla_x V) + \text{op}(\{b, \mathcal{H}_0\})) \psi(s, x) dx + \mathcal{O}(h^2) \\
&= \mathcal{O}(h^2).
\end{aligned}$$

Integrating both sides, we get the desired result.  $\square$

*Remark 3.2.*

1. It follows from the proof that a similar observable bound also holds for the Schrödinger equation with only the kinetic part or potential part. In particular, the  $\mathcal{O}(h^2)$  error only comes from the potential part and there is no asymptotic error in the kinetic part.
2. Note that the proof also works with minor modifications for the linear time-dependent Hamiltonian

$$-\frac{h^2}{2} \Delta + V(t, x),$$

provided that  $V$  is smooth with all derivatives bounded, as well as the controlled Hamiltonian in quantum control problems [1, 44, 56, 66],

$$-\frac{h^2}{2} u_1(t) \Delta + u_2(t) V(x),$$

where  $u_1, u_2 \in [0, 1]$  are the control functions and  $V(x)$  smooth with all derivatives bounded. But note that for these linear cases, it can be more convenient to directly work with operators. Specifically, the Hamiltonian corresponds to the Weyl quantization of time-dependent symbols, and Egorov's theorem in such cases can be found in, e.g., [67, section 11].

3. The proof only works for Schwartz functions  $a(x, \xi)$ . An extension to a more general symbol class  $S_\delta(m)$  (see [67, Chapter 11]) may be possible and is left for future study.

Theorem 3.1, together with (2.4) and Lemma 2.2 provides the Egorov-type results in terms of the Wigner and Husimi functions.



COROLLARY 3.3 (Egorov's theorem in the Wigner functions and Husimi functions). *Under the conditions of Theorem 3.1, the observable expectations in terms of the Wigner and Husimi functions satisfy*

$$(3.5) \quad \left| \langle A \rangle_{\psi^h(t, \cdot)} - \int_{\mathbb{R}^{2d}} a \circ \Phi^t(x, \xi) w^h[\psi_0^h](x, \xi) dx d\xi \right| \leq Cth^2,$$

$$(3.6) \quad \left| \langle A \rangle_{\psi^h(t, \cdot)} - \int_{\mathbb{R}^{2d}} \left( a - \frac{h}{4} \Delta a \right) \circ \Phi^t(x, \xi) \sigma^h[\psi_0^h](x, \xi) dx d\xi \right| \leq Cth^2,$$

respectively, where  $C \geq 0$  is some constant depending on the derivatives of  $a$  and  $V$  but independent of  $h$  and  $\psi_0^h$ .

#### 4. Numerical schemes and observable errors.

**4.1. Time-splitting schemes.** The time-splitting algorithm is proposed as follows: From time  $t = t_n = n\Delta t$  to  $t = t_{n+1} = (n+1)\Delta t$ , with  $\Delta t$  given, the QCMD is split into two subsystems. One contains the kinetic contributions

$$(4.1) \quad \begin{cases} i\hbar \partial_t \psi^h = -\frac{\hbar^2}{2} \Delta_x \psi^h, \\ \dot{y} = v, \\ \dot{v} = 0, \end{cases}$$

whose unitary propagation of the wave function from time  $t = a$  to  $t = b$  is denoted as  $U_T(a, b)$ , and the numerical flow in this step is denoted as  $\Phi_T^{b-a}$  from time  $t = a$  to time  $t = b$ . The other is associated with the potential contributions

$$(4.2) \quad \begin{cases} i\hbar \partial_t \psi^h = V(x, y(t)) \psi^h, \\ \dot{y} = 0, \\ \dot{v} = -\int_{\mathbb{R}^d} \nabla_y V(x, y) |\psi^h(t, x)|^2 dx, \end{cases}$$

and we denote the unitary evolution of the wave function following this potential state from time  $t = a$  to  $t = b$  as  $U_V(a, b)$ , and the numerical flow in this step is denoted as  $\Phi_V^{b-a}$  from time  $t = a$  to time  $t = b$ . Note that in the evolution  $U_V(a, b)$ ,  $|\psi^h(t, x)|^2$  does not change with respect to time. This observation is crucially important in the consideration of its classical counterpart, which makes the resulting ODE an autonomous one. A first-order Lie splitting then corresponds to evolving (4.1) from time  $t_n$  to  $t_{n+1}$ , followed by the evolution of (4.2) from time  $t_n$  to  $t_{n+1}$ . Here, we focus on the discussion of the Strang splitting given as

$$(4.3) \quad \psi_{n+1}^h = U_V(t_n + \Delta t/2, t_{n+1}) U_T(t_n, t_{n+1}) U_V(t_n, t_n + \Delta t/2) \psi_n^h,$$

$$(4.4) \quad (y^{n+1}, v^{n+1}) = \Sigma_V^{\Delta t/2} \circ \Sigma_T^{\Delta t} \circ \Sigma_V^{\Delta t/2} (y^n, v^n) := \Sigma_{SV}^{\Delta t} (y^n, v^n),$$

where the scheme for the Newtonian part  $(y, v)$  is in fact the Störmer–Verlet integrator (denoted as SV), which will be detailed in section 4.2. To be more precise, the second-order time-splitting from time  $t_n$  to  $t_{n+1}$  reads

$$\begin{aligned} \psi_*^h &= e^{-i\Delta t V(x, y^n)/(2\hbar)} \psi_n^h, \quad y^* = y^n, \quad v^* = v^n - \frac{\Delta t}{2} \int \nabla_y V(x, y^n) |\psi_n^h(x)|^2 dx, \\ \psi_{**}^h &= e^{i\hbar \Delta t \Delta_x/2} \psi_*^h, \quad y^{**} = y^* + v^* \Delta t, \quad v^{**} = v^*, \\ \psi_{n+1}^h &= e^{-i\Delta t V(x, y^{**})/(2\hbar)} \psi_{**}^h, \quad y^{n+1} = y^{**}, \\ v^{n+1} &= v^{**} - \frac{\Delta t}{2} \int \nabla_y V(x, y^{**}) |\psi_{**}^h(x)|^2 dx, \end{aligned}$$

where one can use Fourier spectral methods to implement the exponentiation of the Laplacian and trapezoidal rules for the numerical quadrature. We focus on the time-splitting strategy (in the semidiscrete setup) and a detailed discussion on the spatial discretization is rather standard (see, e.g., [5, 6, 24]) and beyond the scope of this paper. Nevertheless, we point out that when employing the spectral discretization spatially, one gets an unconditionally stable scheme because the mass of the system can be shown to be conserved, following the strategies as proposed in [5, 24], which we do not detail here.

**4.2. Observable errors of the Strang splitting.** In this section, we aim to provide a direct error bound of the observable for the Strang splitting. The strategy is to convert the error estimate of the microscopic solver into an estimate of three parts: the errors of the macroscopic numerical solver, and the asymptotic errors of both the continuous and discrete multiscale limit, as is commonly used in the error analysis of the asymptotic preserving schemes (see, e.g., [29, 33, 42]) and asymptotically compatible schemes [19, 57]. Note that the corresponding classical counterparts of (4.1) and (4.2) are given as

$$(4.5) \quad \begin{cases} \dot{x} = \xi, \\ \dot{\xi} = 0, \\ \dot{y} = v, \\ \dot{v} = 0, \end{cases} \quad \text{and} \quad \begin{cases} \dot{x} = 0, \\ \dot{\xi} = -\nabla_x V(x, y), \\ \dot{y} = 0, \\ \dot{v} = -\nabla_y V(x, y), \end{cases}$$

respectively, and denote the corresponding flow map from time  $t = a$  to time  $t = b$  as  $\Phi_T^{b-a}$  and  $\Phi_V^{b-a}$ . We further define

$$\Phi_{SV}^{\Delta t} := \Phi_V^{\Delta t/2} \circ \Phi_T^{\Delta t} \circ \Phi_V^{\Delta t/2}.$$

Therefore, the Strang splitting of the QCMD as proposed in (4.3) corresponds to the second-order SV integrator of the Hamiltonian system governed by the Hamiltonian  $h_{\text{total}} = \frac{|\xi|^2}{2} + \frac{|v|^2}{2} + V(x, y)$ , namely,

$$\begin{aligned} x^* &= x^n, \quad \xi^* = \xi^n - \frac{\Delta t}{2} \nabla_x V(x^n, y^n), \quad y^* = y^n, \quad v^* = v^n - \frac{\Delta t}{2} \nabla_y V(x^n, y^n), \\ x^{**} &= x^* + \Delta t \xi^*, \quad \xi^{**} = \xi^*, \quad y^{**} = y^* + v^* \Delta t, \quad v^{**} = v^*, \quad x^{n+1} = x^{**}, \\ \xi^{n+1} &= \xi^{**} - \frac{\Delta t}{2} \nabla_x V(x^{**}, y^{**}), \quad y^{n+1} = y^{**}, \quad v^{n+1} = v^{**} - \frac{\Delta t}{2} \nabla_y V(x^{**}, y^{**}). \end{aligned}$$

We provide two cases as proofs for our main theorem. One is in the simplified case where the initial wave function is a complex Gaussian and in this proof we only use the Wigner function; the second proof is for the general case, where when the Wigner function loses its nonnegativity, we are forced to turn to the Husimi functions. Both proofs employ the observable estimate of the QCMD Theorem 3.1 and its consequence Corollary 3.3, which bridge the quantum observables with a classical Hamiltonian flow, and the numerical errors of the SV integrator of this classical counterpart.

**THEOREM 4.1** (observable convergence of the Strang splitting). *Let the observable  $A = \text{op}(a)$  be the Weyl quantization of a Schwartz function  $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ , and the potential function  $V$  be smooth with its derivatives of order  $\geq 2$  all bounded. Then the global error in the expectation value of  $A$  can be estimated as*

$$(4.6) \quad |\langle \psi_n^h | A | \psi_n^h \rangle - \langle \psi^h(T) | A | \psi^h(T) \rangle| \leq C_T (\Delta t^2 + h^2)$$

for some  $C_T$  depending on  $t_n = T$  and the derivatives of  $a$  and  $V$ .

*Proof of the case with an initial complex Gaussian.* From  $t_n$  to  $t_{n+1}$ , because of the construction of the scheme

$$\begin{aligned}\psi_{n+1}^h &= U_V(t_n + \Delta t/2, t_{n+1}) U_T(t_n, t_{n+1}) U_V(t_n, t_n + \Delta t/2) \psi_n^h \\ &= U_V(t_n + \Delta t/2, t_{n+1}) U_T(t_n, t_{n+1}) \psi_*^h = U_V(t_n + \Delta t/2, t_{n+1}) \psi_{**}^h\end{aligned}$$

and Theorem 3.1, one has

$$\begin{aligned}\langle \psi_{n+1}^h | A | \psi_{n+1}^h \rangle &= \langle A \rangle_{U_V(t_n + \Delta t/2, t_{n+1}) \psi_{**}^h} \\ &= \langle \text{op}(a \circ \Phi_V^{\Delta t/2}) \rangle_{\psi_{**}^h} + \mathcal{O}(\Delta t h^2) \\ &= \langle \text{op}(a \circ \Phi_V^{\Delta t/2} \circ \Phi_T^{\Delta t}) \rangle_{\psi_*^h} + \mathcal{O}(\Delta t h^2) \\ &= \langle \text{op}(a \circ \Phi_V^{\Delta t/2} \circ \Phi_T^{\Delta t} \circ \Phi_V^{\Delta t/2}) \rangle_{\psi_n^h} + \mathcal{O}(\Delta t h^2) \\ &= \langle \text{op}(a \circ \Phi_{SV}^{\Delta t}) \rangle_{\psi_n^h} + \mathcal{O}(\Delta t h^2).\end{aligned}$$

One can repeat this procedure for each time interval and obtain

$$\begin{aligned}\langle \psi_n^h | A | \psi_n^h \rangle &= \left\langle \text{op} \left( a \circ (\Phi_{SV}^{\Delta t})^n \right) \right\rangle_{\psi_0^h} + \mathcal{O}(T h^2) \\ (4.7) \quad &= \int_{\mathbb{R}^{2d}} \left( a \circ (\Phi_{SV}^{\Delta t})^n \right) (q, p) w^h[\psi_0^h](q, p) dq dp + \mathcal{O}(h^2),\end{aligned}$$

where in the last line we used the relationship of the Wigner function and Weyl quantization. Note that since  $\psi_0^h$  is a complex Gaussian, its Wigner function is nonnegative, i.e.,  $w^h[\psi_0^h] \geq 0$  and

$$\int_{\mathbb{R}^{2d}} w^h[\psi_0^h] dq dp = \|\psi_0^h\|^2 = 1.$$

On the other hand, applying Corollary 3.3 to the exact solution  $\psi^h(t_n)$ , we have

$$(4.8) \quad \langle \psi^h(t_n) | A | \psi^h(t_n) \rangle = \int_{\mathbb{R}^{2d}} (a \circ \Phi^{t_n}) (q, p) w^h[\psi_0^h](q, p) dq dp + \mathcal{O}(h^2),$$

and hence it suffices to compare the difference in the  $a$ 's following the two different classical flows. The error bounds of the SV integrator of the classical Hamiltonian system read

$$\|a \circ \Phi^{t_n} - a \circ (\Phi_{SV}^{\Delta t})^n\|_{L^\infty(\mathbb{R}^{2d})} \leq C \Delta t^2$$

for some constant  $C$  depending on  $t_n = T$ . As an immediate consequence, one has

$$\int_{\mathbb{R}^{2d}} \left( a \circ (\Phi_{SV}^{\Delta t})^n - a \circ \Phi^{t_n} \right) (q, p) w^h[\psi_0^h](q, p) dq dp \leq C \Delta t^2.$$

Combining (4.7) and (4.8), we arrived at the desired result.  $\square$

For general initial data, one no longer has the nonnegativity of the Wigner function  $w^h[\psi_0^h] \geq 0$ , but fortunately, the Husimi function is still nonnegative. Therefore, we perform the proof using the Husimi function instead.

*Proof of the general case.* Similar to the proof using the Wigner function, we have

$$\begin{aligned}\langle \psi_n^h | A | \psi_n^h \rangle &= \left\langle \text{op} \left( a \circ (\Phi_{SV}^{\Delta t})^n \right) \right\rangle_{\psi_0^h} + \mathcal{O}(T h^2) \\ &= \int_{\mathbb{R}^{2d}} \left( a \circ (\Phi_{SV}^{\Delta t})^n - \frac{h}{4} \Delta a \circ (\Phi_{SV}^{\Delta t})^n \right) (q, p) \sigma^h[\psi_0^h](q, p) dq dp + \mathcal{O}(h^2),\end{aligned}$$

where in the last line we used Lemma 2.2 to represent the expectation in the Husimi function. On the other hand, thanks to Corollary 3.3, we also have a similar result for the exact solution,

$$\langle \psi^h(t_n) | A | \psi^h(t_n) \rangle = \int_{\mathbb{R}^{2d}} \left( a \circ \Phi^{t_n} - \frac{h}{4} \Delta a \circ \Phi^{t_n} \right) (q, p) \sigma^h[\psi_0^h](q, p) dq dp + \mathcal{O}(h^2).$$

Notice that  $\sigma^h[\psi_0]$  is a nonnegative function and satisfies

$$\int_{\mathbb{R}^{2d}} \sigma^h[\psi_0^h] dq dp = \|\psi_0^h\|^2 = 1,$$

and hence besides the comparison of  $a$  following the flows  $\Phi^{t_n}$  and  $(\Phi_{SV}^{\Delta t})^n$ , we also need to compare that of  $\Delta a$ . Fortunately, the error bounds of the SV integrator [32] of the classical Hamiltonian system satisfy

$$|\Phi^{t_n}(q, p) - (\Phi_{SV}^{\Delta t})^n(q, p)| \leq C\Delta t^2, \quad |D\Phi^{t_n}(q, p) - D(\Phi_{SV}^{\Delta t})^n(q, p)| \leq C\Delta t^2$$

for some constant  $C$  depending on  $t_n = T$  and independent for  $n$  and  $\Delta t$  thanks to the symplecticity of the integrator. Thus,

$$\|a \circ \Phi^{t_n} - a \circ (\Phi_{SV}^{\Delta t})^n\|_{L^\infty(\mathbb{R}^{2d})} \leq C\Delta t^2,$$

and

$$\begin{aligned} & \|\Delta a \circ \Phi^{t_n} - \Delta a \circ (\Phi_{SV}^{\Delta t})^n\|_{L^\infty(\mathbb{R}^{2d})} \\ & \leq \|D(\Delta a)\|_{L^\infty(\mathbb{R}^{2d})} \|D\Phi^{t_n} - D(\Phi_{SV}^{\Delta t})^n\|_{L^\infty(\mathbb{R}^{2d})} \leq C\Delta t^2. \end{aligned}$$

Therefore,

$$|\langle \psi^h(t_n) | A | \psi^h(t_n) \rangle - \langle \psi_n^h | A | \psi_n^h \rangle| \leq C_T(\Delta t^2 + h^2),$$

as desired.  $\square$

Note that Theorem 4.1 is not a uniform bound due to the presence of  $h$  on the right-hand side, but it drastically improves the result using a direct estimate of the wave function  $\Delta t^2/h^2$  or  $\Delta t^2/h$  depending on the regularity of the initial condition (see, e.g., [5, 20, 21]). Nevertheless, we remark that it is possible to achieve an estimate uniform in  $h$  combining the two kind of error bounds, namely, the error can be chosen as the minimum value between them,

$$(4.9) \quad \min_{0 < h \leq 1} \left\{ \Delta t^2 + h^2, \frac{\Delta t^2}{h} \right\},$$

and in the worst case scenario when  $h = \mathcal{O}(\Delta t^{2/3})$ , one has the error bound of  $\mathcal{O}(\Delta t^{4/3})$ . We summarize the result in the following corollary.

**COROLLARY 4.2** (a uniform observable estimate of the Strang splitting). *Let the observable  $A = \text{op}(a)$  be the Weyl quantization of a Schwartz function  $a : \mathbb{R}^{2d} \rightarrow \mathbb{R}$ , and the initial wave function  $\psi_0^h(x)$  is of the WKB type with bounded amplitude and phase. Under the assumptions of Theorem 4.1, we have the uniform-in- $h$  estimate of the global error in the expectation value of  $A$  as*

$$|\langle \psi_n^h | A | \psi_n^h \rangle - \langle \psi^h(T) | A | \psi^h(T) \rangle| \leq C_T \Delta t^{4/3}$$

for some  $C_T$  depending on  $t_n = T$ , the functions  $a$  and  $V$ , and the initial wave packet  $\psi_0^h$ .

*Remark 4.3.*

1. We remark that the additional assumption on the initial wave function comes from the condition of the result for the wave function error estimate. To be specific, for the WKB type initial condition  $\psi_0^h$ , one has  $h^j \|\psi_0\|_{H^j} \leq M_j$  with some constant  $M_j$  for  $j = 1, 2$ , which is the assumption of the initial condition for the vector norm bound of such semiclassical problems [20, 21].
2. More importantly, we remark that the convergence rate of  $4/3$  is not sharp. The optimal rate is unknown in the literature even for the linear case. We conjecture that the optimal uniform-in- $h$  rate is  $\Delta t^2$  at least for the linear case, and the improvement of the estimate is left for future work.
3. We also clarify that these results are for the time-splitting (Trotter) errors with spatial degrees of freedom kept continuous. The spatial discretization error is not characterized here. In the numerical examples, we implement the spatial discretization via the Fourier-based pseudospectral discretization with  $\Delta x = \mathcal{O}(h)$ , which has high accuracy (spectral accuracy). To prove a uniform error bound with spatial discretization is a more delicate issue, and there is so far no rigorous result in the literature even for the linear case, which remains as an important open question in this field. The description for the nonlinear case is beyond the scope of this paper. Nevertheless, it is worth mentioning that we have recently discovered an approach directly analyzing the observable error bounds in the spatially discrete setting via discrete microlocal analysis [13, 18, 23], which is a work in progress by one of the authors [14].

**5. Numerical results.** In this section, we shall report on a few numerical examples, which illustrate our theoretical results, in particular, the observable error bounds presented in Theorem 4.1 and the errors of the wave function depicted in (4.9).

To this end, we choose an interaction potential of the form  $V(x, y) = \sin(x^2 + y^2)$ , and the initial condition as

$$(5.1) \quad \psi_0^h = \mathcal{Z} \exp(-12.5(x+1)^2 + i50(x+1)),$$

where  $\mathcal{Z}$  is the normalization factor such that  $\|\psi_0^h\|_{L^2} = 1$ . The computational domain is  $[-\pi, \pi]$  with  $\Delta x = 2\pi h/32$ . The numerical solutions are computed by the time-splitting methods till  $T = 0.5$  and the trapezoidal rules are applied spatially for the quadrature that are known to be spectrally accurate for periodic functions. We present the absolute error of the wave functions  $\psi^h$  at  $t_n = T$  which are computed by

$$(\Delta x)^{1/2} \|\psi_n^h - \psi^h(t_n)\|_2 \approx \|\psi_n^h(\cdot) - \psi^h(t_n, \cdot)\|_{L^2},$$

and for the observable expectations are computed using absolute values between the reference and numerical values as defined on the left-hand side of (4.6). We use the numerical results computed by a very small step size  $\Delta t = 10^{-5}$  as the reference solution.

Besides the wave function, we consider the expectation  $\langle A \rangle_{\psi^h(t, \cdot)}$  of the following observables:

- The position operator  $\hat{x}$ , defined as multiplication by  $x$ , namely,  $\hat{x}\psi = x\psi$ .
- The momentum operator  $\hat{p} = -i\hbar \nabla_x$ .

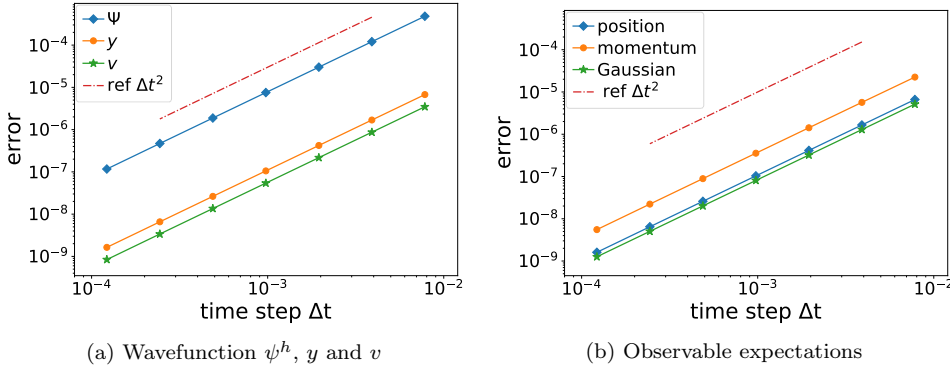


FIG. 1. Errors versus various time steps. Left: Log-log plot of the errors of the wave functions  $\psi$ ,  $y$ , and  $v$ . Right: Log-log plot of the errors of the position, momentum, and Gaussian observable expectations. All errors are of second-order convergence in time.

- The Gaussian observable (labeled as “Gaussian” in the figures) defined as the multiplication by  $e^{-4x^2}$ .
- The Schwartz observable (labeled as “xGaussian” in the figures) defined as the multiplication by  $xe^{-4x^2}$ .
- Kinetic operator  $\frac{1}{2}\hat{p}^2 = -\frac{1}{2}h^2\Delta_x$ .

To demonstrate Theorem 4.1, we present the numerical results of varying both  $\Delta t$  and  $h$ . In the first test, we fix  $h = 0.04$  and take the step sizes  $\Delta t = 2^{-6}, 2^{-7}, \dots, 2^{-11}$ . The numerical errors of the wave function  $\psi^h$  and the macroscopic quantities  $y$  and  $v$  are plotted in Figure 1(a), while Figure 1(b) plots the errors in the expectations of the position, momentum, and Gaussian observables. It can be seen that all errors are of second-order convergence in time, which confirms the scaling in  $\Delta t$  as shown in (4.6). However, the errors in the wave function are of a magnitude larger than those of the observable expectations and the macroscopic quantities  $y$  and  $v$ . This implies that to capture the correct physical observables, one may choose a larger step size compared to the wave function.

We then vary the size of the semiclassical parameter  $h$ . In this test, we fix  $\Delta t = 0.001$  and take  $h = 2^{-4}, 2^{-5}, \dots, 2^{-10}$ . The errors of the observable expectations are recorded in Figure 2. It can be seen in Figure 2(a) that the errors of the Gaussian and Schwartz observables are of second order in  $h$ , which agrees with (4.6). Besides the Schwartz observables, we also test other non-Schwartz observables such as the position, momentum, and kinetic operators. Note that, although, in these cases, our main theorem can no longer be applied, we still observe additive error bounds in  $h$ . In particular, the errors of the expectations for the position and momentum operator scale as  $\mathcal{O}(h)$  as shown in Figure 2(b) and that for the position operator scales as  $\mathcal{O}(h^2)$ . It is worth pointing out that we define the Weyl quantization (2.3) for Schwartz functions  $a$  and the lemma on expectation Lemma 2.2 also requires  $a$  to be Schwartz, and hence a proof of rigorous observable error bounds for such non-Schwartz observables or polynomials of  $\hat{x}$  and  $\hat{p}$  is beyond the current framework, which is left as an interesting future direction.

Finally, we present the error of the wave function when varying  $h$ . Figure 3 plots the errors of the wave function  $\psi^h$  for  $h = 2^{-7}, 2^{-8}, \dots, 2^{-13}$ . It can be seen the scaling is  $h^{-1}$  as given in (4.9), which is used to generate the uniform error bound. This  $h^{-1}$  scaling is interesting in the following two perspectives. First, it helps to demonstrate the uniform error bounds. To be specific, note that the uniform-in- $h$  observable error

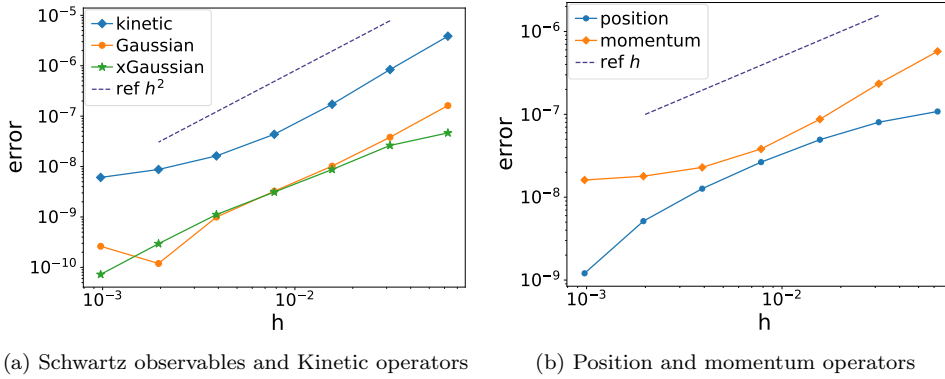


FIG. 2. Errors for various  $h$ . Left: Log-log plot of the errors of the expectations for the Schwartz observables and kinetic operator. Right: Log-log plot of the errors of the position and momentum operators. For Schwartz observables, the error scales as  $h^2$ , which matches our theoretical bounds as in (4.6). For non-Schwartz observables, a scaling of  $h^\alpha$  (with  $\alpha > 0$ ) is observed.

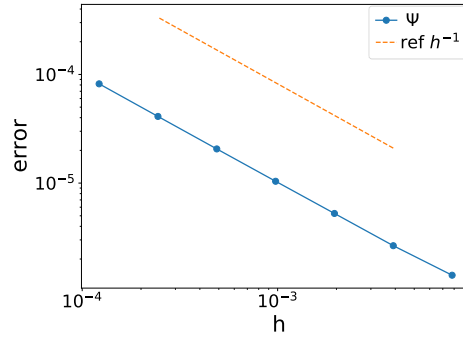


FIG. 3. Log-log plot of the errors of the wave function  $\psi^h$ .

bounds in Corollary 4.2 are not tight and hence difficult to be directly numerically verified. Here we verify both terms in (4.9) numerically instead. Furthermore, the error grows as  $h$  increases which is the opposite of the observable and this further illustrates that capturing the correct physical observables allows one to take an  $\mathcal{O}(1)$  time step, much larger than that of the wave function calculations.

**6. Conclusion and discussion.** In this work, we investigate the distance between the QCMD and its semiclassical limit by a careful estimate of the observable expectations. This observable results on the continuous level are then rewritten using both the Wigner and Husimi functions. We then discuss the time-splitting strategies for this quantum-classical mixed dynamics, and prove the observable error bounds of the Strang splitting using the continuous-level result. In particular, the observable error bounds present an additive scaling as  $\Delta t^2 + h^2$ , which drastically improves the estimate compared to a direct estimation via the errors in the wave function that scales as  $\Delta t^2/h$ . Finally, a uniform observable error bound is provided by combining both estimation strategies.

Possible future directions branch into three different streams. One is to go beyond the Strang splitting and consider higher-order time-splitting strategies such as Suzuki construction [55], Yoshida's triple jump [65], symmetric Zassenhaus splitting

[34], symmetric-conjugate composition [10], for this nonlinear system. The other is to investigate such observable estimates for other linear and nonlinear systems with their corresponding numerical schemes, such as the surface hopping methods with matrix potentials [15, 16, 25, 43], the time-dependent self-consistent field equations [9, 36, 37, 38, 45], and other mean-field systems. We point out that the QCMD preserves its total energy and, hence, in the semiclassical limit an autonomous Hamiltonian flow is arrived at. However, when the total energy is not preserved or the symbol  $\mathcal{H}_0$  of the total energy is time dependent, one gets a nonautonomous ODE system instead. Even though the Egorov's theorem Theorem 3.1 on the continuous level could still hold, the analysis for the time-splitting schemes may require a correspondence between the time-splitting strategy with some symplectic integrator for nonautonomous Hamiltonian system, which could bring some challenges to the analysis. Nevertheless, such nonautonomous cases would be an interesting future direction. The last is to consider non-Schwartz functions, such as polynomials of the position and momentum operators, which is left for future study.

Our paper focuses on the observable error bounds in terms of the time discretization. On a separate matter, to establish a uniform error bound of the observables in the spatially discrete setting is an important open problem even for the linear cases. Though beyond the scope of this paper, we have recently discovered a promising approach directly analyzing the observable error bounds with spatial discretizations via discrete microlocal analysis [13, 14, 18, 23], which is a work in progress by one of the authors. As for implementations of the high-dimensional PDE grids, recent advances of quantum algorithms for unbounded Hamiltonian simulation [1, 3, 4, 17, 53, 58] provide new prospects for efficient implementations and the combination of the observable error bounds and quantum implementations is an interesting future direction.

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