# Propagation Through Conical Crossings: An Asymptotic Semigroup

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**Abstract**

We consider the standard model problem for a conical intersection of electronic surfaces in molecular dynamics. Our main result is the construction of a semigroup in order to approximate the Wigner function associated with the solution of the Schrödinger equation at leading order in the semiclassical parameter. The semigroup stems from an underlying Markov process that combines deterministic transport along classical trajectories within the electronic surfaces and random jumps between the surfaces near the crossing. Our semigroup can be viewed as a rigorous mathematical counterpart of so-called trajectory surface hopping algorithms, which are of major importance in molecular simulations in chemical physics. The key point of our analysis, the incorporation of the nonadiabatic transitions, is based on the Landau-Zener type formula of Fermanian-Kammerer and Gérard [10] for the propagation of two-scale Wigner measures through conical crossings. © 2005 Wiley Periodicals, Inc.

## 1 Introduction

We consider the time-dependent Schrödinger equation

$$\begin{aligned} i\varepsilon\partial_t\psi^\varepsilon(q,t) &= \left(-\frac{\varepsilon^2}{2}\Delta_q + V(q)\right)\psi^\varepsilon(q,t),\\ \psi^\varepsilon(q,0) &= \psi_0^\varepsilon(q) \in L^2(\mathbb{R}^2,\mathbb{C}^2), \end{aligned} \tag{1.1}$$

with matrix-valued potential

$$V(q) = \begin{pmatrix} q_1 & q_2 \\ q_2 & -q_1 \end{pmatrix}, \quad q = (q_1, q_2) \in \mathbb{R}^2,$$

and small semiclassical parameter $\varepsilon > 0$. The eigenvalues of the matrix $V(q)$ are $E^\pm(q) = \pm|q|$ and meet at $q = 0$. Their joint graph shows two intersecting cones explaining the notion of a conical crossing. It is well known that away from the crossing region and for small $\varepsilon$ the system (1.1) approximately decouples into two

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scalar equations. We denote by $\chi^{\pm}(q)$ smooth eigenfunctions of $V(q)$ corresponding to the eigenvalues $E^{\pm}(q)$ and decompose the solution of (1.1) as

$$\psi(q,t) = \phi^{+}(q,t)\chi^{+}(q) + \phi^{-}(q,t)\chi^{-}(q).$$

Then, the scalar components $\phi^{\pm}(t) \in L^2(\mathbb{R}^2, \mathbb{C})$ approximately satisfy the effective equations of motion

$$\begin{aligned} \mathrm{i}\varepsilon\partial_t\phi^{+}(q,t) &= \left(-\frac{\varepsilon^2}{2}\Delta_q^{A^+} + E^{+}(q)\right)\phi^{+}(q,t), \\ \mathrm{i}\varepsilon\partial_t\phi^{-}(q,t) &= \left(-\frac{\varepsilon^2}{2}\Delta_q^{A^-} + E^{-}(q)\right)\phi^{-}(q,t), \end{aligned} \tag{1.2}$$

as long as $\psi(q,t)$ is mostly supported away from the crossing $q = 0$. Here,

$$-\Delta_q^{A^{\pm}} = (-\mathrm{i}\nabla_q - A^{\pm}(q))^2, \qquad A^{\pm}(q) = \mathrm{i}\langle \chi^{\pm}(q), \nabla_q \chi^{\pm}(q)\rangle_{\mathbb{C}^2},$$

is the Laplacian of the covariant derivative with respect to the Berry connection $A^{\pm}(q)$. This form of adiabatic decoupling is at the heart of time-dependent Born-Oppenheimer approximation. The smaller the adiabatic parameter $\varepsilon > 0$, the better the decoupling. Near the crossing point, however, this decoupling breaks down no matter how small $\varepsilon$ is, and the main concern of our work is an approximate description of solutions to (1.1), which come near or pass through $q = 0$.

To make this more precise, we recall that the solutions $\phi^{\pm}(q,t)$ of the decoupled system (1.2) behave semiclassically; i.e., they can approximately be described by means of the classical flows $\Phi^t_{\pm} : \mathbb{R}^4 \to \mathbb{R}^4$ of

$$\dot q(t) = p(t), \quad \dot p(t) = \mp\frac{q(t)}{|q(t)|}, \qquad q(0) = q_0, \quad p(0) = p_0, \tag{1.3}$$

which stem from the Hamiltonian functions $\lambda^{\pm}(q,p) = \frac{1}{2}|p|^2 + E^{\pm}(q)$. One possible way of formulating such a semiclassical limit uses Wigner transforms. The Wigner transform

$$\begin{aligned} w^{\varepsilon}_{\pm}(t)(q,p) &:= w^{\varepsilon}(\phi^{\pm}(t))(q,p) \\ &= (2\pi)^{-2}\int_{\mathbb{R}^2} \phi^{\pm}\left(t, q - \frac{\varepsilon}{2}x\right)\overline{\phi^{\pm}}\left(t, q + \frac{\varepsilon}{2}x\right)\mathrm{e}^{\mathrm{i}x\cdot p}\,\mathrm{d}x, \end{aligned}$$

is a real-valued function on phase space $\mathbb{R}^4$, encoding position and momentum distribution of the scalar-valued wave function $\phi^{\pm}(t)$. Basic properties and an alternative definition as a distribution are discussed in Section 2. The Wigner functions $w^{\varepsilon}_{\pm}(t)$ are approximately transported by the respective classical flows,

$$\begin{pmatrix} w^{\varepsilon}_{+}(t) \\ w^{\varepsilon}_{-}(t) \end{pmatrix} \approx \underbrace{\begin{pmatrix} \mathcal{L}^t_{+} & 0 \\ 0 & \mathcal{L}^t_{-} \end{pmatrix}}_{=:\mathcal{L}^t_0} \begin{pmatrix} w^{\varepsilon}_{+}(0) \\ w^{\varepsilon}_{-}(0) \end{pmatrix} := \begin{pmatrix} w^{\varepsilon}_{+}(0)\circ\Phi^{-t}_{+} \\ w^{\varepsilon}_{-}(0)\circ\Phi^{-t}_{-} \end{pmatrix}. \tag{1.4}$$

Our main result is an extension of (1.4) to the case where the solution of (1.1) comes close to the crossing. While a semiclassical description is still appropriate near the crossing, the adiabatic decoupling breaks down. Therefore, the group $\mathcal{L}_0^t$ in (1.4) must be supplemented with off-diagonal terms describing an exchange of mass between $w_+^\varepsilon$ and $w_-^\varepsilon$. In Section 2, we give (1.4) a more precise meaning and extend it to the crossing region by constructing an $\varepsilon$-dependent semigroup $\mathcal{L}_\varepsilon^t$. The semigroup $\mathcal{L}_\varepsilon^t$ is the forward semigroup of a Markov process based on a family of random trajectories. The random trajectories are just the deterministic solutions of (1.3), which jump from one band to the other with a certain probability whenever their distance to the crossing attains a local minimum. The jump probabilities are obtained from the solution of the classical, purely time-dependent Landau-Zener problem.

The breakdown of the adiabatic approximation near conical crossings of eigenvalue bands has generated a lot of research in the context of molecular dynamics as well as in solid state physics; see, for example, the review article [30] or the monograph [5]. The mathematical results on the propagation through crossings can be organized into two groups. The first is the semiclassical propagation of coherent states. In his pioneering monograph [15], Hagedorn classified 11 possible types of eigenvalue crossings of minimal multiplicity in molecular dynamics and constructed Gaussian wave packets whose centers pass exactly through the crossing. The 11 types have crossing manifolds of codimension one, two, three, or five in the nucleonic configuration space, the conical crossing being the codimension two crossing. The second group includes approaches within the framework of microlocal analysis. In [10], Fermanian-Kammerer and Gérard derived Landau-Zener type formulae for the two-scale Wigner measure passing through conical crossings. An analogous result for codimension three crossings was given by the same authors in [11]. A central role in the proof of their transition formulae is played by microlocal normal forms for the time-dependent operator near the crossing manifold. More precise normal forms have also been found by Colin de Verdière [6].

The proof of our result as given in this paper heavily relies on the results obtained by Fermanian-Kammerer and Gérard in [10]. The key idea is to lift the Landau-Zener type formula for the two-scale Wigner measure established in [10] to a semigroup acting on the Wigner function. The main novelty of our result is that it yields an approximate description of the solution to the Schrödinger equation (1.1) combining the following three properties. Firstly, since the effective semigroup acts on the Wigner function, we obtain an effective description for finite values of $\varepsilon > 0$. Secondly, we allow for general initial conditions. Thirdly, the scale $\sqrt{\varepsilon}$ associated with the nonadiabatic transitions enters the semigroup just via the transition rates and does not require the introduction of additional variables. As a consequence, our description directly translates into an algorithm for numerical simulations in concrete applications. In contrast, the previous mathematical results have one or more of these points as desiderata. While Hagedorn constructs approximate solutions to (1.1) and as such obtains very detailed information, his construction is restricted to special initial states, namely semiclassical Gaussian wave packets with center passing exactly through the crossing. The approach of Fermanian-Kammerer and Gérard comes with the difficulty that the two-scale Wigner measure is an object defined only in the semiclassical limit $\varepsilon \to 0$ and intrinsically associated with an involutive manifold in the cotangent space of space-time $T^*(\mathbb{R}_t \times \mathbb{R}^2_q)$. Moreover, the two-scale Wigner measures depend on an additional variable introduced to control the $\sqrt{\varepsilon}$-concentration of the wave function with respect to this manifold.

We postpone a more detailed discussion of the applicability of our method and its connection to the trajectory surface-hopping algorithms of chemical physics to a forthcoming publication [18]. There we present, in particular, an implementation of the algorithm based on our semigroup as well as numerical experiments comparing true numerical solutions of the Schrödinger equation (1.1) with solutions obtained by applying our semigroup to the Wigner function of the initial data.

The plan of this paper is as follows: In Section 2 we introduce the semigroup $\mathcal{L}^t_\varepsilon$, which transports the diagonal elements of the Wigner transform through the crossing region, and in Section 3 we formulate our main result. Section 4 together with the appendix provides a self-contained discussion of the two-scale analysis of the problem, which allows us, in particular, to incorporate the Landau-Zener type formulae of [10] in the proof of the main result in Section 5.

We end the introduction with some remarks on the origin of the model problem (1.1) in molecular dynamics. If the electronic part of the full molecular Hamiltonian has a pair of eigenvalue surfaces that intersect each other but are globally isolated from the remainder of the electronic spectrum, then the results of [28] allow for a uniform reduction of the full molecular problem to a two-band model of the form

$$\begin{aligned} i\varepsilon\partial_t \psi(q,t) &= -\frac{\varepsilon^2}{2}\Delta_q \psi(q,t) + \widetilde{V}(q)\psi(q,t), \\ \psi(q,0) &= \psi_0(q) \in L^2(\mathbb{R}^n, \mathbb{C}^2), \end{aligned}$$

where the semiclassical parameter $\varepsilon = \sqrt{m_\mathrm{e}/m_\mathrm{n}}$ is given through the mass ratio between the light electrons and the heavy nuclei. The potential $\widetilde{V}(q)$ is a hermitian $2 \times 2$ matrix with eigenvalues intersecting on a submanifold of the nucleonic configuration space $\mathbb{R}^n$. For time-reversal invariant systems, $\widetilde{V}(q)$ is real symmetric. Generically, for such matrices the crossing manifold is a submanifold of codimension two. Following [15], one first subtracts the trace of the matrix. Then, a (locally) linear change of coordinates moves the crossing into the submanifold $\{q \in \mathbb{R}^n \mid q_1 = q_2 = 0\}$. Taylor expansion around the point $q = 0$ provides the generic form

$$\widetilde{V}(q) = \begin{pmatrix} \alpha \cdot q & \beta \cdot q \\ \beta \cdot q & -\alpha \cdot q \end{pmatrix} + \mathcal{O}(|q^2|), \quad q \in \mathbb{R}^n,$$

with linearly independent vectors $\alpha, \beta \in \mathbb{R}^n$. The Taylor expansion is justified if one is interested only in the behavior of the solutions near the crossing. An appropriate rotation eliminates all but the first two components of $\alpha$ and $\beta$ and thus leaves us with linearly independent vectors $a, b \in \mathbb{R}^2$ and

$$\begin{pmatrix} a \cdot \tilde{q} & b \cdot \tilde{q} \\ b \cdot \tilde{q} & -a \cdot \tilde{q} \end{pmatrix}, \quad \mathbb{R}^2 \ni \tilde{q} = (e_1 \cdot q, e_2 \cdot q),$$

which is the potential of our model problem if $a = e_1 = (1,0)^{\mathsf{T}}$ and $b = e_2 = (0,1)^{\mathsf{T}}$.

## 2 An Asymptotic Semigroup for the Wigner Function

A straightforward adaption of the Faris-Lavine theorem to the case of matrix-valued operators [17] shows the essential self-adjointness of the Hamiltonian

$$H^\varepsilon = -\frac{\varepsilon^2}{2}\Delta_q + V(q) = -\frac{\varepsilon^2}{2}\Delta_q + \begin{pmatrix} q_1 & q_2 \\ q_2 & -q_1 \end{pmatrix} \tag{2.1}$$

on $C_c^\infty(\mathbb{R}^2, \mathbb{C}^2)$. By the spectral theorem, the Schrödinger equation (1.1) has a unique global solution $\psi^\varepsilon(\cdot) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$. We are interested in the leading-order asymptotics of this solution for small values of the semiclassical parameter $\varepsilon$. Up to a global phase factor, a wave function $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ can be uniquely represented by its Wigner function $W^\varepsilon(\psi) \in L^2(\mathbb{R}^4, \mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2))$ given through

$$W^\varepsilon(\psi)(q,p) = (2\pi)^{-2} \int_{\mathbb{R}^2} \psi\left(q - \frac{\varepsilon}{2}x\right) \otimes \overline{\psi}\left(q + \frac{\varepsilon}{2}x\right) \mathrm{e}^{\mathrm{i}x\cdot p}\, \mathrm{d}x.$$

For vector-valued wave functions $\psi$, the Wigner function $W^\varepsilon(\psi)$ takes values in the space of self-adjoint $2 \times 2$ matrices $\mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2)$. Moreover, the Wigner transformation

$$W^\varepsilon : L^2(\mathbb{R}^2, \mathbb{C}^2) \to L^2(\mathbb{R}^4, \mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2)), \quad \psi \mapsto W^\varepsilon(\psi),$$

is bounded, and we also have $W^\varepsilon(\psi) \in C_0(\mathbb{R}^4, \mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2))$. One is tempted to think of the trace of a Wigner function as a probability density on phase space. However, in general, $W^\varepsilon(\psi)(q,p)$ may have negative eigenvalues. The analytical power of the Wigner function stems from a direct relation to expectation values with respect to certain Weyl quantized observables. A convenient symbol class is

$$S_0^0(1) = C_b^\infty(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)),$$

consisting of smooth functions with values in the space of $2 \times 2$ matrices $\mathcal{L}(\mathbb{C}^2)$, which are bounded together with all their derivatives. By the Calderon-Vaillancourt theorem, the Weyl quantization of an observable $a \in S_0^0(1)$ is a continuous operator on $L^2(\mathbb{R}^2, \mathbb{C}^2)$ with

$$\|a(q, -\mathrm{i}\varepsilon\nabla_q)\|_{\mathcal{L}(L^2)} \le \text{const} \sum_{|\alpha| \le 5} \|\partial^\alpha a\|_\infty =: c_4(a) \quad \text{for all } \varepsilon > 0,$$

where the index in $c_4$ reminds us of the four dimensions of phase space. Thus, for wave functions $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ and observables $a \in S^0_0(1)$ we have

$$|\langle \psi, a(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)}| \le c_4(a)\|\psi\|^2_{L^2(\mathbb{R}^2)},$$

and the mapping $a \mapsto \langle \psi, a(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle$ is a continuous linear functional on the space $C^\infty_{\mathrm{b}}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$. For Schwartz functions $a \in \mathcal{S}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$ an explicit calculation yields

$$\langle \psi, a(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} = \int_{\mathbb{R}^4} \mathrm{tr}\,(a(q, p)\, W^\varepsilon(\psi)(q, p))\, \mathrm{d}q\, \mathrm{d}p. \tag{2.2}$$

Hence, we can view the Wigner function $W^\varepsilon(\psi)$ as a continuous functional on any subspace of admissible observables $\mathcal{B} \subset S^0_0(1)$,

$$\mathcal{B} \to \mathbb{C}, \quad a \mapsto \langle W^\varepsilon(\psi), a\rangle_{\mathcal{B}',\mathcal{B}}.$$

In the following, various test function spaces $\mathcal{B}$ will appear. The dual pairing $\langle W^\varepsilon(\psi), a\rangle_{\mathcal{B}',\mathcal{B}}$ will always be well-defined by either the left- or the right-hand side of (2.2).

## 2.1 Propagation Away from the Crossing

Roughly speaking, as long as the solution $\psi^\varepsilon(t)$ of the Schrödinger system (1.1) is mostly supported away from the crossing $\{q = 0\}$, its leading-order asymptotics can be characterized conveniently in terms of classical transport equations for the diagonal elements of its Wigner function $W^\varepsilon(\psi^\varepsilon(t))$. For a more precise statement, we need to fix some notation. Let $h(q, p) = \frac{1}{2}|p|^2 + V(q)$ denote the symbol of the operator $H^\varepsilon$ in (2.1). Let $\lambda^\pm(q, p) = \frac{1}{2}|p|^2 + E^\pm(q)$ be the classical Hamiltonian function corresponding to the eigenvalue $E^\pm(q) = \pm|q|$ of $V(q)$. We denote by $\Pi^\pm(q) \in \mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2)$ the orthogonal spectral projection for $E^\pm(q)$, and observe that $\Pi^\pm \in C^\infty(\mathbb{R}^2 \setminus \{0\}, \mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2))$. Since the eigenspaces are one-dimensional, the diagonal components of a Wigner function are conveniently written as

$$\Pi^\pm W^\varepsilon(\psi)\Pi^\pm = \mathrm{tr}(W^\varepsilon(\psi)\Pi^\pm)\Pi^\pm =: w^\varepsilon_\pm(\psi)\Pi^\pm \in L^2(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)).$$

We first study the classical dynamics associated with the Hamilton functions $\lambda^+$ and $\lambda^-$, that is, the Hamiltonian systems (1.3). Away from the crossing manifold $\{q = 0\}$, the solution curves of these systems are well-defined and smooth. Due to the rotational symmetry of $E^\pm(q)$, we have two conserved quantities, energy $\lambda^\pm(q, p)$ and angular momentum

$$q \wedge p := q^\perp \cdot p = q_1 p_2 - q_2 p_1, \quad (q, p) \in \mathbb{R}^4.$$

Trajectories passing through the set $\{q = 0\}$ at some time $t_0$ must have zero angular momentum. As long as $p(t_0) \neq 0$, these trajectories have a unique continuous continuation through $\{q = 0\}$. Denoting the zero-energy shell by $(\lambda^\pm)^{-1}(0) :=$

$\{(q, p) \in \mathbb{R}^4 \mid \lambda^{\pm}(q, p) = 0\}$ and the hypersurface of zero angular momentum by $I = \{(q, p) \in \mathbb{R}^4 \mid q \wedge p = 0\}$, we define for $t \in \mathbb{R}$

$$\Phi^t_{\pm}(q_0, p_0) = (q^{\pm}(t), p^{\pm}(t)) \quad \text{for } (q_0, p_0) \notin (\lambda^{\pm})^{-1}(0) \cap I,$$
$$\Phi^t_{\pm}(q_0, p_0) = (q_0, p_0) \quad \text{for } (q_0, p_0) \in (\lambda^{\pm})^{-1}(0) \cap I.$$

We note, that $\{\Phi^t_{\pm}(q, p)\}_{t\in\mathbb{R}}$ forms a group for all $(q, p) \in \mathbb{R}^4$. Since $(\lambda^+)^{-1}(0) = \{(0, 0)\}$, the mapping $\mathbb{R}^4 \to \mathbb{R}^4$, $(q, p) \mapsto \Phi^t_+(q, p)$ is continuous for all $t \in \mathbb{R}$. For the dynamics associated with $\lambda^-$, however, we only have continuity of the mapping $(q, p) \mapsto \Phi^t_-(q, p)$ outside the codimension two set $(\lambda^-)^{-1}(0) \cap I = \{(q, p) \in \mathbb{R}^4 \mid q = \pm(|p|/2)\, p\}$. Nevertheless, for any wave function $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ the functions $w^{\varepsilon}_{\pm}(\psi) \circ \Phi^{-t}_{\pm}$ are well-defined in $L^2(\mathbb{R}^4, \mathbb{C})$.

Now, let $\psi^{\varepsilon}(t)$ be the solution to the Schrödinger equation (1.1). For the moment, we work on time intervals for which the solution is mostly supported away from the crossing. Such finite time intervals $[0, T]$ can be characterized by the existence of an open set $\{q = 0\} \subset U \subset \mathbb{R}^4$ containing the crossing manifold such that for all $t \in [0, T]$

$$\int_U |W^{\varepsilon}(\psi^{\varepsilon}(t))(q, p)| \mathrm{d}q\, \mathrm{d}p = \mathcal{O}(\varepsilon). \tag{2.3}$$

On such intervals $[0, T]$, one recovers the leading-order Born-Oppenheimer approximation, that is

$$\int_{\mathbb{R}^4} \left(w^{\varepsilon}_{\pm}(\psi^{\varepsilon}(t)) - w^{\varepsilon}_{\pm}(\psi_0) \circ \Phi^{-t}_{\pm}\right)(q, p) a(q, p) \mathrm{d}q\, \mathrm{d}p = \mathcal{O}(\varepsilon) \tag{2.4}$$

for all observables $a \in \mathcal{S}(\mathbb{R}^4, \mathbb{C})$ with $\operatorname{supp}(a) \cap \{q = 0\} = \varnothing$, uniformly in $t \in [0, T]$. For a stronger result, which implies the above approximation, we refer to theorem 4 in [28]. Equation (2.4) means that away from the crossing the diagonal elements of $W^{\varepsilon}(\psi^{\varepsilon}(t))$ are approximately transported like classical densities on phase space. This motivates the definition of a Born-Oppenheimer function

$$W^{\varepsilon}_{\mathrm{BO}}(t) := \left(w^{\varepsilon}_{+}(\psi_0) \circ \Phi^{-t}_{+}\right) \Pi^{+} + \left(w^{\varepsilon}_{-}(\psi_0) \circ \Phi^{-t}_{-}\right) \Pi^{-} \in L^2(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$$

for $t \in \mathbb{R}$. Rephrasing the preceding remarks, we have for all finite time intervals $[0, T]$ satisfying (2.3) and for all diagonal observables $a \in \mathcal{S}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$ with $[a(q, p), V(q)] = 0$ and $\operatorname{supp}(a) \cap \{q = 0\} = \varnothing$

$$\int_{\mathbb{R}^4} \operatorname{tr}\left(\left(W^{\varepsilon}(\psi^{\varepsilon}(t)) - W^{\varepsilon}_{\mathrm{BO}}(t)\right)(q, p) a(q, p)\right) \mathrm{d}q\, \mathrm{d}p = \mathcal{O}(\varepsilon) \tag{2.5}$$

uniformly for $t \in [0, T]$. This is one way to formulate the leading-order time-dependent Born-Oppenheimer approximation: away from the crossing, where the eigenvalue bands are separated by a gap, one has adiabatic decoupling of the associated subspaces and within the decoupled subspaces semiclassical behavior of the solutions of (1.1).

## 2.2 Propagation near the Crossing

It is expected that near the crossing at $q = 0$ certain solutions $\psi^\varepsilon(t)$ of (1.1) exhibit transitions between the subspaces $\operatorname{Ran}\Pi_+$ and $\operatorname{Ran}\Pi_-$ even in the limit $\varepsilon \to 0$. The goal of our analysis is to modify the transport equation (1.4) by taking transfer between the diagonal components $w_+^\varepsilon(\psi)$ and $w_-^\varepsilon(\psi)$ into account.

Following Remark 5.2 in [25], we observe that the Hamiltonian $-\frac{\varepsilon^2}{2}\Delta_q + V(q)$ is unitarily equivalent to the semiclassical Weyl quantization of

$$\frac{1}{2}|p|^2 \; + \; |p|^{-1} \begin{pmatrix} q \cdot p & q \wedge p \\ q \wedge p & -q \cdot p \end{pmatrix}. \tag{2.6}$$

*Remark* 2.1. This unitary equivalence is achieved by $\varepsilon$-Fourier transformation, a change to polar coordinates $(r, \phi)$, conjugation by the $\phi/2$-angle rotation matrix, and the observation that the Weyl quantization of the tempered distributions $\sigma(q, p) = |p|^{-1}(q \cdot p)$ and $\tau(q, p) = |p|^{-1}(q \wedge p)$ reads in Fourier transformed polar coordinates as

$$\sigma(q, -\mathrm{i}\varepsilon\nabla_q) \simeq -\mathrm{i}\varepsilon\partial_r - \mathrm{i}\varepsilon\,\frac{1}{2r}, \qquad \tau(q, -i\varepsilon\nabla_q) \simeq -\mathrm{i}\varepsilon\,\frac{1}{r}\,\partial_\phi.$$

We note that the Weyl operator of the symbol in (2.6) is the first step for an orbital decomposition of the Hamiltonian $H^\varepsilon$; see [25] and for a related result also [1]. The symbol in (2.6) carries two key signatures of the classical dynamics: the angular momentum $q \wedge p$, which is preserved by the Hamiltonian flows $\Phi_\pm^t$, and the function $q \cdot p$, which characterizes the hypersurface

$$S = \{(q, p) \in \mathbb{R}^4 \mid q \cdot p = 0\}$$

containing the points in phase space, at which the classical trajectories attain their minimal distance to the crossing $q = 0$; cf. Figure 2.1.

The heuristic picture underlying our result is to replace $(q, p)$ in (2.6) by classical trajectories $(q(t), p(t))$ related to the classical flows $\Phi_\pm^t$ and to solve the purely time-adiabatic problem

$$\mathrm{i}\varepsilon\partial_t\phi(t) = |p(t)|^{-1}\begin{pmatrix} q(t)\cdot p(t) & q(t)\wedge p(t) \\ q(t)\wedge p(t) & -q(t)\cdot p(t)\end{pmatrix}\phi(t), \quad \phi(t) \in \mathbb{C}^2. \tag{2.7}$$

Since the transitions happen only in the region where a trajectory has minimal distance to the crossing, we linearize the flows around $S$. The linearizations of the classical flows $\Phi_\pm^t$ at a point $(q_*, p_*) \in S$ are

$$q^\pm(t) = q_* + tp_* + \mathcal{O}(t^2) \quad \text{and} \quad p^\pm(t) = p_* \mp t\,\frac{q_*}{|q_*|} + \mathcal{O}(t^2). \tag{2.8}$$

The system (2.7) becomes

$$\mathrm{i}\,\underbrace{\frac{\varepsilon}{|p_*|}}_{=:\widetilde{\varepsilon}}\,\partial_t\,\phi(t) = \begin{pmatrix} t & \frac{q_*\wedge p_*}{|p_*|^2} \\ \frac{q_*\wedge p_*}{|p_*|^2} & -t \end{pmatrix}\phi(t) =: \begin{pmatrix} t & \delta \\ \delta & -t\end{pmatrix}\phi(t), \tag{2.9}$$

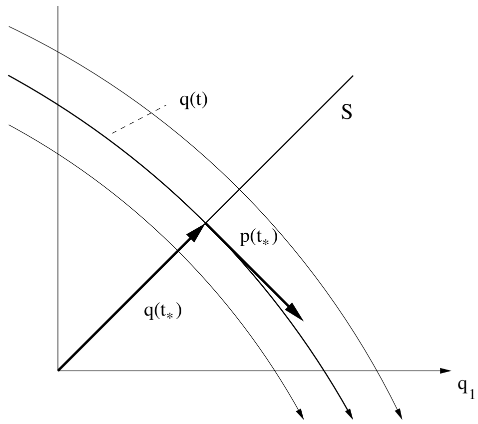


FIGURE 2.1. We see the projections of three neighboring trajectories $(q(t), p(t))$ onto configuration space $\mathbb{R}^2_q$. The crossing manifold $\{q = 0\}$ is therefore projected onto the origin. The trajectories attain their minimal distance to the crossing at the time $t_*$ when $q(t_*) \cdot p(t_*) = 0$. The points in phase space where $q \cdot p = 0$ build up the jump manifold $S$.

where we used that $|q_*|/|p_*|^2 \ll 1$ near the crossing. We note that this last expression does not depend on whether we employ $\Phi^t_+$ or $\Phi^t_-$. However, (2.9) is nothing but the famous Landau-Zener problem [31]. It is well known that for

$$\begin{pmatrix} |\phi^+(-\infty)|^2 \\ |\phi^-(-\infty)|^2 \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \text{or} \quad \begin{pmatrix} |\phi^+(-\infty)|^2 \\ |\phi^-(-\infty)|^2 \end{pmatrix} = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

the solution $\phi(t)$ of (2.9) satisfies

$$\begin{pmatrix} |\phi^+(\infty)|^2 \\ |\phi^-(\infty)|^2 \end{pmatrix} = \begin{pmatrix} 1 - T^\varepsilon(q_*, p_*) & T^\varepsilon(q_*, p_*) \\ T^\varepsilon(q_*, p_*) & 1 - T^\varepsilon(q_*, p_*) \end{pmatrix} \begin{pmatrix} |\phi^+(-\infty)|^2 \\ |\phi^-(-\infty)|^2 \end{pmatrix}$$

with

$$\begin{aligned} T^\varepsilon(q_*, p_*) := \exp\left(-\frac{\pi\,\delta^2}{\widetilde{\varepsilon}}\right) &= \exp\left(-\frac{\pi}{\varepsilon}\frac{(q_* \wedge p_*)^2}{|p_*|^3}\right) \\ &= \exp\left(-\frac{\pi}{\varepsilon}\frac{|q_*|^2}{|p_*|}\right). \end{aligned} \tag{2.10}$$

The components of the solution $\phi$ are $\phi^\pm(\pm\infty)$, when $\phi$ is decomposed into the eigenvectors of the Landau-Zener matrix for large positive and negative times $t \to \pm\infty$, respectively. For a concise review on Landau-Zener type problems we refer to [16]. The subsequent analysis will indeed show that the heuristic picture of classical transport in combination with the transition probability (2.10) yields a correct description of the leading-order dynamics.

To incorporate the $\varepsilon$-dependent transition probability (2.10) into the transport of the Wigner function, we first append to phase space a label $j \in \{-1, 1\}$ indicating whether the description refers to Ran $\Pi^-$ or Ran $\Pi^+$. We introduce a Markov process defined by the random trajectories

$$\mathcal{J}_\varepsilon^{(q,p,j)} : [0, \infty) \to \mathbb{R}^4 \times \{-1, 1\},$$

where $\mathcal{J}_\varepsilon^{(q,p,j)}(t) = (\Phi_j^t(q, p), j)$ as long as $q(t) \cdot p(t) \neq 0$. Whenever the deterministic flow $\Phi_j^t(q, p)$ hits the manifold $S$, a jump occurs with probability $T^\varepsilon(q(t), p(t))$; that is, $j$ changes to $-j$ with probability $T^\varepsilon(q(t), p(t))$. After the jump the trajectory follows again the deterministic flow depending on $j$ until the trajectory again hits $S$. At the jump hypersurface $S$, the trajectories are chosen right continuous. On the submanifold $S_{\mathrm{cl}} = \{(q, p) \in S \mid |p|^2 = |q|\}$ of closed circular orbits of $\Phi_+^t$ the trajectories do not jump.

*Remark* 2.2. We emphasize that the underlying physics is of course not one of instantaneously jumping particles. Indeed, for (2.9) it is known that the transition occurs smoothly within an $\sqrt{\varepsilon}$-neighborhood of $t = 0$; cf. [2, 3, 4, 21].

In each finite time interval $[0, T] \subset [0, \infty)$, each path $(q, p, j) \to \mathcal{J}_\varepsilon^{(q,p,j)}(t)$ has only a finite number of jumps and remains in a bounded region of phase space. Moreover, the paths $(q, p, j) \to \mathcal{J}_\varepsilon^{(q,p,j)}(t)$ are smooth away from $S$, that is, on $(\mathbb{R}^4 \setminus S) \times \{-1, 1\}$. Hence, the random trajectories $\mathcal{J}_\varepsilon^{(q,p,j)}$ define a Markov process

$$\{\mathbb{P}^{(q,p,j)} \mid (q, p, j) \in \mathbb{R}^4 \times \{-1, 1\}\}$$

on $\mathbb{R}^4 \times \{-1, 1\}$; see, for example, III-§1 in [8]. With the transition function of a Markov process one associates a backward and a forward semigroup, which act on function spaces and spaces of set functions, respectively; cf. [8] or [20]. We define the corresponding Markov (backwards) semigroup on the following space of functions:

DEFINITION 2.3 A compactly supported function $f \in C_{\mathrm{c}}((\mathbb{R}^4 \setminus S) \times \{-1, 1\}, \mathbb{C})$ belongs to the space $\mathcal{C}$ if it satisfies the following boundary conditions at the jump manifold:

$$\begin{aligned}\lim_{\delta \to +0} f\left(q - \delta p, p + \delta j \frac{q}{|q|}, j\right) &= T^\varepsilon(q, p) \lim_{\delta \to +0} f\left(q + \delta p, p + \delta j \frac{q}{|q|}, -j\right)\\ &= T^\varepsilon(q, p) f(q, p, -j)\end{aligned}$$

and

$$\begin{aligned}&\lim_{\delta \to +0} f\left(q - \delta p, p + \delta j \frac{q}{|q|}, j\right)\\ &\quad = (1 - T^\varepsilon(q, p)) \lim_{\delta \to +0} f\left(q + \delta p, p - \delta j \frac{q}{|q|}, j\right)\\ &\quad = (1 - T^\varepsilon(q, p)) f(q, p, j)\end{aligned}$$

for all $(q, p) \in S \setminus S_{\mathrm{cl}}$.

*Remark* 2.4. The limits in the preceding definition are taken along the linearization of the unique trajectory of (1.3) passing through a point in $S \setminus S_{\mathrm{cl}}$ when hitting the jump manifold $S$; see also (2.8).

By construction, the backwards semigroup acting on functions $f \in \mathcal{C}$

$$\mathcal{L}_\varepsilon^t : \mathcal{C} \to \mathcal{C}, \quad (\mathcal{L}_\varepsilon^t f)(q,p,j) := \mathbb{E}^{(q,p,j)} f(\mathcal{J}_\varepsilon^{(q,p,j)}(t)),$$

leaves invariant the space $\mathcal{C}$. We write continuous, compactly supported matrix-valued functions $a \in C_{\mathrm{c}}(\mathbb{R}^4 \setminus S, \mathcal{L}(\mathbb{C}^2))$ as

$$a = a^+ \Pi^+ + a^- \Pi^- + \Pi^+ a\, \Pi^- + \Pi^- a\, \Pi^+$$

with $a^\pm := \mathrm{tr}(a\, \Pi^\pm)$. We denote by $\mathcal{C}_{\mathrm{diag}}$ the space of diagonal test functions $a \in C_{\mathrm{c}}(\mathbb{R}^4 \setminus S, \mathcal{L}(\mathbb{C}^2))$ such that $a = a^+\Pi^+ + a^-\Pi^-$ with $a^+, a^- \in \mathcal{C}$, and set for such $a \in \mathcal{C}_{\mathrm{diag}}$

$$\mathcal{L}_{\varepsilon,\pm}^t a := \left(\mathcal{L}_\varepsilon^t (a^+, a^-)\right)^\pm, \quad \mathcal{L}_\varepsilon^t a := \left(\mathcal{L}_{\varepsilon,+}^t a\right)\Pi^+ + \left(\mathcal{L}_{\varepsilon,-}^t a\right)\Pi^-.$$

With this definition the semigroup $\mathcal{L}_\varepsilon^t$ acts invariantly on $\mathcal{C}_{\mathrm{diag}}$, and we can now define its action on Wigner functions by duality.

DEFINITION 2.5 Let $W^\varepsilon(\psi)$ be the Wigner function of a function $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$. We define $\mathcal{L}_\varepsilon^t\, W^\varepsilon(\psi)$ as the linear functional

$$\mathcal{L}_\varepsilon^t W^\varepsilon(\psi) : \mathcal{C}_{\mathrm{diag}} \to \mathbb{C}, \quad a \mapsto \int_{\mathbb{R}^4} \mathrm{tr}\left(W^\varepsilon(\psi)(q,p)\left(\mathcal{L}_\varepsilon^t a\right)(q,p)\right) \mathrm{d}q\, \mathrm{d}p.$$

Since $W^\varepsilon(\psi) \in C_0(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$ and $\mathcal{L}_\varepsilon^t a \in \mathcal{C}_{\mathrm{diag}}$, we clearly have $\mathcal{L}_\varepsilon^t W^\varepsilon(\psi) \in C(\mathbb{R}^4 \setminus S, \mathcal{L}(\mathbb{C}^2))$. Moreover, $S \subset \mathbb{R}^4$ has zero Lebesgue measure. Hence,

$$\mathcal{L}_\varepsilon^t\, W^\varepsilon(\psi) \in L^1_{\mathrm{loc}}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)).$$

Analogously to the Born-Oppenheimer function $W_{\mathrm{BO}}^\varepsilon(t)$, we name

$$W_{\mathrm{LZ}}^\varepsilon(t) := \mathcal{L}_\varepsilon^t W^\varepsilon(\psi_0) \in L^1_{\mathrm{loc}}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)), \quad t \in [0,\infty),$$

the Landau-Zener function. The Landau-Zener function $W_{\mathrm{LZ}}^\varepsilon(t)$ incorporates classical transport and $\varepsilon$-dependent nonadiabatic transitions near the crossing. Our main result, Theorem 3.2, states that $W_{\mathrm{LZ}}^\varepsilon(t)$ approximates the Wigner function $W^\varepsilon(\psi^\varepsilon(t))$ of the solution to the Schrödinger equation (1.1) in the limit $\varepsilon \to 0$.

*Remark* 2.6. The heuristic argument yielding the Landau-Zener formula (2.10) also applies to the generic potential discussed in the introduction

$$V(q) = \begin{pmatrix} a \cdot q & b \cdot q \\ b \cdot q & -a \cdot q \end{pmatrix}.$$

If we denote by $M = (a^\mathsf{T}, b^\mathsf{T})$ the $2 \times 2$ matrix with row vectors $a^\mathsf{T}, b^\mathsf{T} \in \mathbb{R}^2$, then the jump manifold is given by $\{(q,p) \in \mathbb{R}^4 \mid Mq \cdot Mp = 0\}$, and the transition probability reads as

$$T_\varepsilon(q_*, p_*) = \exp\left(-\frac{\pi}{\varepsilon}\frac{(Mq_* \wedge Mp_*)^2}{|Mp_*|^3}\right).$$

However, our proof only works for the case $a = (1, 0)^\mathsf{T}$, $b = (0, 1)^\mathsf{T}$, since it relies on conservation of angular momentum $q \wedge p$.

## 3 Main Result

The nonadiabatic transfer of mass between $\operatorname{Ran}\Pi^+$ and $\operatorname{Ran}\Pi^-$ in the crossing region is realized in the semigroup $\mathcal{L}_\varepsilon^t$ by jumping at the manifold $S$ with the Landau-Zener transition probability $T^\varepsilon$. Clearly, $\mathcal{L}_\varepsilon^t$ does not correctly resolve the dynamics directly at the manifold $S$, but it gives an approximate description of the total nonadiabatic transfer when the solution has passed by. Hence, the Landau-Zener function $W_{\mathrm{LZ}}^\varepsilon(t)$ can only be a sensible approximation to the true Wigner function $W^\varepsilon(\psi^\varepsilon(t))$ away from $S$. Therefore we restrict ourselves to test functions supported away from $S$, and we also have to assume that the initial data have negligible mass near the jump manifold $S$.

DEFINITION 3.1 A sequence of wave functions $(\psi^\varepsilon)_{\varepsilon>0}$ in $L^2(\mathbb{R}^2, \mathbb{C}^2)$ is said to have negligible mass near the jump manifold $S$ if there exists $\delta > 0$ such that

$$\lim_{\varepsilon\to 0} \int_{S_\delta} |W^\varepsilon(\psi^\varepsilon)(q, p)|\, \mathrm{d}q\, \mathrm{d}p = 0$$

with $S_\delta = \{(q, p) \in \mathbb{R}^4 \mid |q \cdot p| \le \delta\}$ the closed $\delta$-tube around $S$.

Initial data with negligible mass near $S$ are, for example, associated with semiclassical Gaussian wave packets

$$(2\pi\varepsilon)^{-1/2} \exp\left(-\frac{1}{2\varepsilon}|q - q_0|^2 + \frac{\mathrm{i}}{\varepsilon} p_0 \cdot q\right)$$

with center $(q_0, p_0) \in \mathbb{R}^4$, $|q_0 \cdot p_0| \ne 0$, or WKB type states $f(q)\mathrm{e}^{\mathrm{i}S(q)/\varepsilon}$ with amplitude $f \in L^2(\mathbb{R}^2, \mathbb{R})$ and phase $S \in C^1(\mathbb{R}^2, \mathbb{R})$ such that $|q \cdot \nabla_q S(q)| \ge \delta$ on $\operatorname{supp}(f)$.

Though incorporating nonadiabatic transitions, the semigroup $\mathcal{L}_\varepsilon^t$ still gives a semiclassical description of the dynamics. Hence, we do not obtain information about the off-diagonal terms of the Wigner function, which are highly oscillatory and vanish when averaged over time; see Lemma 4.9. By choosing observables that are diagonal with respect to the potential $V(q)$, we conveniently suppress the uncontrolled off-diagonal parts of $W^\varepsilon(\psi^\varepsilon(t))$. This restriction to the diagonal components, however, prohibits the resolution of possible interferences between parts of the wave function originating from different levels. Such interferences might occur if classical trajectories arrive with the same momentum at the same time at the jump manifold on the upper and the lower band. A simple condition ruling out such a scenario is the choice of initial data just associated with $\operatorname{Ran}\Pi^+$, that is, $\psi_0^\varepsilon(q) = \phi_0^\varepsilon(q)\,\chi^+(q)$ with $\phi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C})$. In this case, all trajectories associated with the flow $\Phi_-^t$ originate from trajectories of the flow $\Phi_+^t$ having passed the jump manifold $S$. Since such trajectories $(q^-(t), p^-(t))$ do not come back to $S$, there are no interferences.

The last issue to be addressed before formulating our main result is rather technical. Since we must allow for $\varepsilon$-dependent initial data, we have to make sure that the family of initial wave functions $(\psi_0^\varepsilon)_{\varepsilon>0}$ behaves properly as $\varepsilon \to 0$. It turns out that the appropriate condition is that the sequence of two-scale Wigner functionals $(W_2^\varepsilon(\psi_0^\varepsilon))_{\varepsilon>0}$ converges to a two-scale Wigner measure $\rho_0$. We postpone the definition and discussion of two-scale Wigner functionals and measures to the following section. However, we note that this assumption is satisfied by all standard families of initial wave functions $(\psi_0^\varepsilon)_{\varepsilon>0}$ like semiclassical wave packets and semiclassical WKB states and also by initial conditions not depending on $\varepsilon$ at all. Moreover, the assumption can be dropped completely if one is willing to work with subsequences of the initial sequence $(\psi_0^\varepsilon)_{\varepsilon>0}$.

THEOREM 3.2 *Let* $(\psi_0^\varepsilon)_{\varepsilon>0}$ *be a bounded sequence in* $L^2(\mathbb{R}^2, \mathbb{C}^2)$ *associated with* $\operatorname{Ran}\Pi^+$, *that is, with* $w_-^\varepsilon(\psi_0^\varepsilon) = 0$, *with negligible mass near the jump manifold* $S$. *Assume that the sequence of two-scale Wigner functionals* $(W_2^\varepsilon(\psi_0^\varepsilon))_{\varepsilon>0}$ *has a weak-star limit* $\rho_0$ *as defined in Definition* 4.6.

*Then, for all* $T > 0$ *the solution* $\psi^\varepsilon(t)$ *of the Schrödinger equation* (1.1) *with initial data* $\psi^\varepsilon(0) = \psi_0^\varepsilon$ *satisfies*

$$\lim_{\varepsilon\to 0} \sup_{t\in[0,T]} \int_{\mathbb{R}^4} \operatorname{tr}\Big(\big(W^\varepsilon(\psi^\varepsilon(t)) - W_{\mathrm{LZ}}^\varepsilon(t)\big)(q,p)a(q,p)\Big)\mathrm{d}q\,\mathrm{d}p = 0 \tag{3.1}$$

*for all observables* $a \in C_{\mathrm{c}}^\infty(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$ *with*

$$\operatorname{supp}(a) \subset \mathbb{R}^4 \setminus S \quad \textit{and} \quad [a(q,p), V(q)] = 0 \ \textit{for } (q,p) \in \mathbb{R}^4.$$

*Remark* 3.3. We emphasize that Theorem 3.2 extends the Born-Oppenheimer approximation in a nontrivial way. The transition probabilities $T^\varepsilon(q,p)$ incorporated into the semigroup $\mathcal{L}_\varepsilon^t$ result in leading-order nonadiabatic transitions for a large class of initial data. All initial wave functions with phase space support in an $\sqrt{\varepsilon}$-neighborhood of the zero angular momentum hypersurface $\{(q,p) \in \mathbb{R}^4 \mid q \wedge p = 0\}$ exhibit order-one transitions.

## 4 Two-Scale Wigner Functionals and Measures

In this section we provide a self-contained discussion of the necessary two-scale analysis required for our proof. Two-scale Wigner measures are measures on an extended phase space $\mathbb{R}_{q,p}^{2d} \times \mathbb{R}_\eta$, using the extra variable $\eta \in \mathbb{R}$ to resolve concentration effects on certain submanifolds of phase space on the finer scale $\sqrt{\varepsilon}$. They have been introduced by Fermanian-Kammerer [9] and Miller [24]. In this section, we review and extend a number of notions and results from [10], which we then will use in the proof of Theorem 3.2. In particular, we pursue three issues. Firstly, we present a self-contained construction of two-scale measures, which relies only on the Calderon-Vaillancourt theorem and a Gårding-type inequality. Secondly, the two-scale Wigner measures used in [10] are measures on an extended phase space of space-time $T^*(\mathbb{R}_t \times \mathbb{R}^2_q) \times \mathbb{R}_\eta = \mathbb{R}^7$. Here, we provide a detailed discussion of the necessary tools to incorporate their Landau-Zener type formula into a description that is pointwise in time. Thirdly, the space of observables used in [10] consists of functions that are constant for large values of the additional coordinate $\eta$. That space is not invariant under multiplication by the two-scale transition rate $\exp(-\pi\,\eta^2/|p|^3)$, and we have to enlarge the space of admissible observables to obtain a well-defined description of the dynamics by means of a semigroup. To proceed in a transparent way, we quickly fix the symbol classes we are working with and recapitulate a suitable definition of Wigner measures.

## 4.1 Symbol Classes and Wigner Measures

With the notation of chapter 7 in [7], we denote by

$$S(m) = \big\{a \in C^\infty(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2)) \mid \\ \forall \alpha \in \mathbb{N}_0^{2n},\ \exists C_\alpha > 0,\ \forall x \in \mathbb{R}^{2n} : |\partial^\alpha a(x)| \le C_\alpha m(x)\big\}.$$

The function $m : \mathbb{R}^{2n} \to [0, \infty]$ is an order function; that is, there exist positive constants $C_m > 0$ and $N_m > 0$ such that

$$\forall x, y \in \mathbb{R}^{2n} : m(x) \le C_m \langle x - y\rangle^{N_m} m(y),$$

where $\langle x\rangle = (1 + |x|^2)^{1/2}$. The space $S(m)$ is a Fréchet space. Let $k \in \mathbb{R}$ and $\delta \in [0, \frac{1}{2}]$. The space $S^k_\delta(m)$ consists of functions $a : \mathbb{R}^{2n} \times ]0, 1], (x, \varepsilon) \mapsto a(x; \varepsilon)$, that satisfy the following two conditions. Firstly, $a(\cdot\,; \varepsilon) \in S(m)$ for all $\varepsilon \in ]0, 1]$, and secondly,

$$\forall \alpha \in \mathbb{N}_0^{2n},\ \exists C_\alpha > 0,\ \forall (x, \varepsilon) \in \mathbb{R}^{2n} \times ]0, 1] : |\partial^\alpha a(x; \varepsilon)| \le C_\alpha m(x) \varepsilon^{-\delta|\alpha| - k}.$$

For us, the two extreme cases $\delta = 0$ and $\delta = \frac{1}{2}$ are the relevant parameters. We note that $S^0_{1/2}(m)$ is a symbol class within which the semiclassical Moyal product $\sharp_\varepsilon$ does not have an asymptotic expansion. However, Moyal multiplication of symbols in $S^0_{1/2}(m)$ with symbols in $S^0_0(m)$ and vice versa is unproblematic, as the following lemma illustrates:

LEMMA 4.1 *For all order functions $m_1$ and $m_2$, the bilinear map*

$$\mathcal{S}(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2)) \times \mathcal{S}(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2)) \to \mathcal{S}(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2)),$$

$$(a \sharp_\varepsilon b)(q, p) := \left(\exp\left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right) a(q, p) b(q', p')\right)\Big|_{q = q', p = p'}$$

*extends continuously to a map* $S^0_0(m_1) \times S^0_{1/2}(m_2) \to S^0_{1/2}(m_1 m_2)$ *and has an asymptotic expansion in* $S^0_{1/2}(m_1 m_2)$

$$\begin{aligned}(a \sharp_\varepsilon b)(q,p) &\sim \sum_{j=0}^{\infty} \frac{1}{j!}\left(\left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right)^j a(q,p)b(q',p')\right)\bigg|_{q=q',p=p'}\\ &=: \sum_{j=0}^{\infty} c_j,\end{aligned}$$

*meaning that* $c_j \in S^{-j/2}_{1/2}(m_1 m_2)$ *for all* $j \in \mathbb{N}_0$ *and that*

$$\left(a \sharp_\varepsilon b - \sum_{j=0}^{N} c_j\right) \in S^{-(N+1)/2}_{1/2}(m_1 m_2) \tag{4.1}$$

*for all* $N \in \mathbb{N}_0$.

The proof of Lemma 4.1 follows standard arguments and we postpone it to Appendix A.1. In Section 2, we have already mentioned that the Wigner function $W^\varepsilon(\psi)$ of a wave function $\psi \in L^2(\mathbb{R}^n, \mathbb{C}^2)$ is a tempered distribution with

$$\left|\langle W^\varepsilon(\psi), a\rangle_{\mathcal{S}',\mathcal{S}}\right| \le c_4(a)\|\psi\|^2_{L^2(\mathbb{R}^n)}$$

for all $a \in \mathcal{S}(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2))$ uniformly in $\varepsilon > 0$. Hence, for bounded sequences $(\psi^\varepsilon)_{\varepsilon>0}$ in $L^2(\mathbb{R}^n, \mathbb{C}^2)$, an application of the Banach-Alaoglu theorem (see [27, theorem 3.17] gives existence of a subsequence $(W^{\varepsilon_k}(\psi^{\varepsilon_k}))_{\varepsilon_k>0}$ that converges with respect to the weak-star topology in $\mathcal{S}'(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2))$. We denote the weak-star limit points of such subsequences by $\mu$. The positivity of $\mu$ is provided by the semiclassical sharp Gårding inequality. In the matrix-valued case, the sharp Gårding inequality was first proven in [19]. Its semiclassical version states that for nonnegative $0 \le a \in S^0_0(1)$, that is, for symbols $a \in C^\infty_{\mathrm{b}}(\mathbb{R}^{2n}, \mathcal{L}(\mathbb{C}^2))$ with

$$\forall u \in \mathbb{C}^2,\ \forall (q,p) \in \mathbb{R}^{2n} : \langle u, a(q,p)u\rangle_{\mathbb{C}^2} \ge 0,$$

there is a positive constant $C = C(a) > 0$ such that for all $\varepsilon > 0$ and all $\psi \in L^2(\mathbb{R}^n, \mathbb{C}^2)$

$$\langle \psi, a(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^n)} \ge -C\varepsilon\|\psi\|^2_{L^2(\mathbb{R}^n)}.$$

Thus, a weak-star limit point $\mu$ of $(W^\varepsilon(\psi^\varepsilon))_{\varepsilon>0}$ is a positive distribution and therefore a positive matrix-valued Radon measure on phase space $\mathbb{R}^{2n}$ called Wigner measure. For an alternative construction of matrix-valued Wigner measures using smooth square roots and composition of pseudodifferential operators, we refer to [14].

## 4.2 Two-Scale Wigner Functionals

We want to analyze concentration effects with respect to a submanifold in phase space

$$I_g := \{(q,p) \in \mathbb{R}^4 \mid g(q,p) = 0\}.$$

For the Schrödinger equation (1.1), we will choose $g(q,p) = q \wedge p$, which is angular momentum, a conserved quantity under the associated Hamiltonian dynamics. We recall that $q \wedge p$ also appeared explicitly in the Landau-Zener transition rate (2.10). This rate indicates that only trajectories within a $\sqrt{\varepsilon}$-neighborhood of $I_g$ in phase space, i.e., in a set

$$\{(q,p) \in \mathbb{R}^4 \mid |q \wedge p| \leq \mathrm{const}\sqrt{\varepsilon}\}$$

experience order-one transition probabilities when coming close to the crossing. The Wigner measure, however, does not resolve this $\sqrt{\varepsilon}$-neighborhood, and a more detailed two-scale analysis becomes necessary. For the general statements about two-scale Wigner functionals and measures, we only assume that $g \in C^\infty(\mathbb{R}^4, \mathbb{R})$ is a smooth, polynomially bounded function; that is, for all $\beta \in \mathbb{N}_0^4$ there is a positive constant $C = C(\beta) > 0$ and a natural number $M = M(\beta) \in \mathbb{N}_0$ such that

$$\forall (q,p) \in \mathbb{R}^4 : |\partial^\beta g(q,p)| \leq C\langle (q,p)\rangle^M.$$

The function $g$ provides us with a notion of (signed) distance to the manifold $I_g$ through $d((q,p), I_g) = g(q,p)$. In the following, the variable $\eta \in \overline{\mathbb{R}}$ measures this distance scaled with $\sqrt{\varepsilon}$, i.e., $\eta(q,p) = g(q,p)/\sqrt{\varepsilon}$. Since we are interested in the limit $\varepsilon \to 0$, the variable $\eta$ is viewed as an element of the one-point compactification $\overline{\mathbb{R}}$ of $\mathbb{R}$. We will use observables depending on $(q,p)$ and on $\eta$ to test the Wigner transform near $I_g$ with respect to the $\sqrt{\varepsilon}$ scale. For $a \in C_{\mathrm{b}}^\infty(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2))$ let

(P) $\forall \alpha \in \mathbb{N}_0, \beta \in \mathbb{N}_0^5, \|\langle (q,p)\rangle^\alpha \partial_{q,p,\eta}^\beta a(q,p,\eta)\|_\infty < \infty,$
$\forall \alpha \in \mathbb{N}_0^4, \beta \in \mathbb{N}, \exists a \in C_{\mathrm{b}}^\infty(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)), \lim_{|\eta|\to\infty} \|\partial_{q,p}^\alpha (a(\cdot,\eta) - a_\infty)\|_\infty = 0, \lim_{|\eta|\to\infty} \|\partial_{q,p}^\alpha \partial_\eta^\beta a(\cdot,\eta)\|_\infty = 0.$

We define the relevant test function space as

$$\mathcal{A} := \left\{a \in C_{\mathrm{b}}^\infty(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2)) \mid a \text{ satisfies property (P)}\right\}$$

and equip it with the topology, which is induced by the family of seminorms

$$\|\langle (q,p)\rangle^\alpha \partial^\beta a(q,p,\eta)\|_\infty, \quad \alpha \in \mathbb{N}_0,\ \beta \in \mathbb{N}_0^5. \tag{4.2}$$

We note that $\mathcal{A}$ is a Fréchet space with the Heine-Borel property; that is, closed and bounded sets are compact. Therefore, $\mathcal{A}$ is a Montel space. In the dual $\mathcal{A}'$ of such spaces, every weak-star convergent sequence is strongly convergent, meaning that for a sequence $(l_n)_{n\in\mathbb{N}}$ in $\mathcal{A}'$

$$\forall a \in \mathcal{A} : \lim_{n\to\infty} l_n(a) = l(a) \Longrightarrow \forall \text{ bounded } B \subset \mathcal{A} : \lim_{n\to\infty} \sup_{a\in B} |l_n(a) - l(a)| = 0;$$

see, for example, [29, prop. 34.6]. We will use this strong convergence property later on.

Let $M_5 := \max_{|\beta|\leq 5} |M(\beta)|$. For $a \in \mathcal{A}$, we denote by

$$s_5(a) := \sum_{\substack{|\alpha|\leq M_5,\\ |\beta|\leq 5}} \|\langle(q,p)\rangle^{\alpha}\partial^{\beta}a(q,p,\eta)\|_{\infty},$$

the finite sum over Schwarz norms, which are of the form (4.2). For observables $a \in \mathcal{A}$, the scaled function

$$(q,p) \mapsto a_{\varepsilon}(q,p) := a\left(q, p, \frac{g(q,p)}{\sqrt{\varepsilon}}\right)$$

lies in the symbol class $S^0_{1/2}(1)$, and we observe that $c_4(a_\varepsilon)$ cannot be bounded by $s_5(a)$ uniformly in $\varepsilon > 0$. Therefore, as in the proof of the Calderon-Vaillancourt theorem for symbol classes $S^0_\delta(1)$ with $\delta \in [0, \frac{1}{2}]$ (see, e.g., [7, theorem 7.11]), we use the unitary scaling

$$S^\varepsilon : L^2(\mathbb{R}^2, \mathbb{C}^2) \to L^2(\mathbb{R}^2, \mathbb{C}^2), \quad \psi(q) \mapsto (S^\varepsilon\psi)(q) := \sqrt{\varepsilon}\,\psi(\sqrt{\varepsilon}\,q),$$

and the alternatively scaled symbol

$$(q,p) \mapsto a_{\varepsilon,2}(q,p) := a\left(\sqrt{\varepsilon}\,q, \sqrt{\varepsilon}\,p, \frac{g(\sqrt{\varepsilon}\,q, \sqrt{\varepsilon}\,p)}{\sqrt{\varepsilon}}\right),$$

which belongs to the symbol class $S^0_0(1)$.

LEMMA 4.2 *Let $a \in \mathcal{A}$ and $\phi, \psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$. Then,*

$$\left\langle \phi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\right\rangle_{L^2(\mathbb{R}^2)} = \left\langle S^\varepsilon\phi, a_{\varepsilon,2}(q, -\mathrm{i}\nabla_q)S^\varepsilon\psi\right\rangle_{L^2(\mathbb{R}^2)}. \tag{4.3}$$

PROOF: Since $a_\varepsilon$ and $a_{\varepsilon,2}$ are Schwartz functions, we just have to carry out a calculation. We have for $\phi, \psi \in \mathcal{S}(\mathbb{R}^2, \mathbb{C}^2)$

$$\langle \phi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} = (2\pi\varepsilon)^{-2}\int_{\mathbb{R}^6} \overline{\phi}(q)\mathrm{e}^{\mathrm{i}(q-q')\cdot p/\varepsilon} a_\varepsilon\left(\frac{q+q'}{2}, p\right)\psi(q')\mathrm{d}q'\,\mathrm{d}p\,\mathrm{d}q.$$

Substituting $q = \sqrt{\varepsilon}x$, $q' = \sqrt{\varepsilon}x'$, and $p = \sqrt{\varepsilon}\,\xi$, we obtain

$$\begin{aligned}
&\langle \phi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\,\psi\rangle_{L^2(\mathbb{R}^2)} \\
&\quad = \varepsilon(2\pi)^{-2}\int_{\mathbb{R}^6} \overline{\phi}(\sqrt{\varepsilon}x)\mathrm{e}^{\mathrm{i}(x-x')\cdot\xi} a\left(\sqrt{\varepsilon}\,\frac{x+x'}{2}, \sqrt{\varepsilon}\xi, g\frac{\left(\sqrt{\varepsilon}\,\frac{x+x'}{2}, \sqrt{\varepsilon}\xi\right)}{\sqrt{\varepsilon}}\right)\cdots \\
&\qquad\qquad \psi(\sqrt{\varepsilon}\,x')\mathrm{d}x'\,\mathrm{d}\xi\,\mathrm{d}x \\
&\quad = \left\langle S^\varepsilon\phi, a_{\varepsilon,2}(q, -\mathrm{i}\nabla_q)S^\varepsilon\psi\right\rangle_{L^2(\mathbb{R}^2)}.
\end{aligned}$$

Since $\|a_{\varepsilon,2}(q, -\mathrm{i}\nabla_q)\|_{\mathcal{L}(L^2)} \leq c_4(a_{\varepsilon,2}) < \infty$, we can conclude (4.3) also for $\phi, \psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ by density. $\square$

For $a \in \mathcal{A}$ we have $c_4(a_{\varepsilon,2}) \leq \text{const}\, s_5(a)$ uniformly in $\varepsilon > 0$ and thus, with Lemma 4.2,

$$\|a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\|_{\mathcal{L}(L^2)} \leq \text{const}\, s_5(a) < \infty. \tag{4.4}$$

As a consequence,

$$\mathcal{A} \to \mathbb{C}, \quad a \mapsto \langle \psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\, \psi \rangle_{L^2(\mathbb{R}^2)},$$

defines a continuous linear functional on $\mathcal{A}$, called a two-scale Wigner functional $W_2^\varepsilon(\psi)$ of $\psi$; see also [12, def. 1]. We note that by identity (4.3) the duality pairing between $W_2^\varepsilon(\psi)$ and $a$ can also be expressed as

$$\left\langle W_2^\varepsilon(\psi), a \right\rangle_{\mathcal{A}',\mathcal{A}} = \int_{\mathbb{R}^4} \operatorname{tr}\left( W^\varepsilon(\psi)(q,p)\, a\left(q, p, \frac{g(q,p)}{\sqrt{\varepsilon}}\right)\right) \mathrm{d}q\, \mathrm{d}p.$$

Therefore, since $W^\varepsilon(\psi) \in C_0(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$, the two-scale Wigner functional $W_2^\varepsilon(\psi)$ can be viewed as the distribution

$$W^\varepsilon(\psi)(q,p)\,\delta\left(\eta - \frac{g(q,p)}{\sqrt{\varepsilon}}\right).$$

The above representation of the two-scale functional $W_2^\varepsilon(\psi)$ also illustrates its dependence on the function $g$ chosen to parametrize the distance to the submanifold $I_g$. In general, the two-scale functional $W_2^\varepsilon(\psi)$ inherits from the Wigner function $W^\varepsilon(\psi)$ the nonpositivity. However, when passing to the semiclassical limit $\varepsilon \to 0$, the following Gårding-type inequality guarantees positivity of the limit points:

PROPOSITION 4.3 *For each nonnegative symbol* $0 \leq a \in \mathcal{A}$ *there is a function* $c : [0, 1[ \to [0, 1[$ *with* $c(\varepsilon) \to 0$ *monotonically as* $\varepsilon \to 0$ *such that for all wave functions* $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$

$$\langle \psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi \rangle_{L^2(\mathbb{R}^2)} \geq -\, c(\varepsilon)\|\psi\|^2_{L^2(\mathbb{R}^2)}.$$

Proposition 4.3 is proven in Appendix A.2. The key observation for the proof is that for symbols in $\mathcal{A} \subset S^0_{1/2}(1)$ the Moyal product has, at least on the formal level, an asymptotic expansion. This is due to a cancellation of "bad terms" of order $1/\sqrt{\varepsilon}$. Our proof uses this fact by approximating the nonnegative symbol $a_\varepsilon$ by a Moyal square $p_\varepsilon \sharp_\varepsilon p_\varepsilon$ of a polynomial $p$ for which the formal expansion agrees with the Moyal product.

*Remark* 4.4. The above Gårding-type inequality, which can be proven by symbolic calculus, is enough for our purpose, the self-contained construction of two-scale Wigner measures. However, by Fourier integral operator techniques, one can improve to a bona fide sharp Gårding inequality: Let $g \in C^\infty(\mathbb{R}^4, \mathbb{R})$ be such that

$\nabla g(q,p) \neq 0$ for all $(q,p) \in I_g$. For each nonnegative $0 \leq a \in \mathcal{A}$ there is a constant $C > 0$ such that for all $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$

$$\langle \psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} \geq -C\sqrt{\varepsilon}\|\psi\|^2_{L^2(\mathbb{R}^2)}.$$

The proof of this stronger inequality is also indicated in Appendix A.2.

## 4.3 Two-Scale Wigner Measures

The Calderon-Vaillancourt theorem and the previous Gårding-type inequality are all we need to study the semiclassical limit of the two-scale Wigner functional $W_2^\varepsilon(\psi^\varepsilon)$ for bounded sequences $(\psi^\varepsilon)_{\varepsilon>0}$ in $L^2(\mathbb{R}^2, \mathbb{C}^2)$.

PROPOSITION 4.5 *Let $(\psi^\varepsilon)_{\varepsilon>0}$ be a bounded sequence in $L^2(\mathbb{R}^2, \mathbb{C}^2)$.*

(i) *$(W_2^\varepsilon(\psi^\varepsilon))_{\varepsilon>0}$ has weak-star limit points $\rho$ in $\mathcal{A}'$. All such limit points $\rho$ are bounded, positive, matrix-valued Radon measures on $\mathbb{R}^4 \times \overline{\mathbb{R}}$.*

(ii) *Let $(W_2^\varepsilon(\psi^\varepsilon))_{\varepsilon>0}$ converge to $\rho$ with respect to the weak-star topology on $\mathcal{A}'$. Then, the sequence $(W^\varepsilon(\psi^\varepsilon))_{\varepsilon>0}$ converges to a Wigner measure $\mu$ in $\mathcal{S}'(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$, and there exists a bounded positive Radon measure $\nu$ on $I_g \times \overline{\mathbb{R}}$ such that*

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} a(q,p,\eta)\rho(\mathrm{d}p,\mathrm{d}q,\mathrm{d}\eta) = \int_{\mathbb{R}^4\setminus I_g} a(q,p,\infty)\mu(\mathrm{d}q,\mathrm{d}p) + \int_{I_g\times\overline{\mathbb{R}}} a(q,p,\eta)\nu(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)$$

*for all $a \in C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$, and we have $\int_{\overline{\mathbb{R}}} \nu(\,\cdot\,, \mathrm{d}\eta) = \mu|_{I_g}$.*

DEFINITION 4.6 The measures $\rho$ introduced in Proposition 4.5 are called two-scale Wigner measures of the bounded sequence $(\psi^\varepsilon)_{\varepsilon>0}$ in $L^2(\mathbb{R}^2, \mathbb{C}^2)$ with respect to the submanifold $I_g$.

Proposition 4.5 is the analogue of theorem 1 in [10]. There, admissible observables are required to be constant with respect to $\eta$ for large $\eta$. That property, however, prevents the definition of a semigroup comparable to $\mathcal{L}_\varepsilon^t$ acting on two-scale observables. Thus, we provide a self-contained proof for the construction with observables in $\mathcal{A}$ in Appendix A.3, which in contrast to the proof of [10] avoids the use of Fourier integral operators.

The measures $\rho$ and $\nu$ depend on the function $g(q,p)$ chosen to describe the submanifold $I_g$. If $\widetilde{g} \in C^\infty(\mathbb{R}^4, \mathbb{R})$ is another function with $I_g = \{\widetilde{g} = 0\}$ sharing the same growth properties as $g$, then for $a \in \mathcal{A}$ the scaled function

$$\widetilde{a}_\varepsilon(q,p) := a\left(q, p, \frac{\widetilde{g}(q,p)}{\sqrt{\varepsilon}}\right)$$

is in $C_{\mathrm{b}}^\infty(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$. Moreover, there exists $f \in C^\infty(\mathbb{R}^4, \mathbb{R})$ with $f(q,p) \neq 0$ for all $(q,p)$ such that $\widetilde{g}(q,p) = f(q,p)g(q,p)$, and setting $a_f(q,p,\eta) :=$

$a(q, p, f(q, p)\eta)$ we clearly have $\widetilde{a}_\varepsilon = (a_f)_\varepsilon$. Thus, repeating the corresponding two-scale construction and denoting the resulting measures by $\widetilde{\rho}$ and $\widetilde{\nu}$, we obtain

$$\rho(q, p, f^{-1}(q, p)\eta) = \widetilde{\rho}(q, p, \eta), \qquad \nu(q, p, f^{-1}(q, p)\eta) = \widetilde{\nu}(q, p, \eta).$$

## 4.4 Propagation of Two-Scale Wigner Functionals

Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ be a solution of the Schrödinger equation (1.1) with initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ and $g(q, p) = q \wedge p$. The two-scale Wigner functional inherits the solution's continuous time dependence, that is,

$$W_2^\varepsilon(\psi^\varepsilon(t)) \in C(\mathbb{R}, \mathcal{A}'),$$

where continuity is understood with respect to the strong dual topology on $\mathcal{A}'$. Indeed, for bounded subsets $B \subset \mathcal{A}$, that is $\sup_{a\in B} \|\langle (q, p)\rangle^\beta \partial^\gamma a\|_\infty < \infty$ for all $\beta \in \mathbb{N}_0$ and $\gamma \in \mathbb{N}_0^5$, we have for $t, t' \in \mathbb{R}$

$$\sup_{a\in B} \left|\left\langle W_2^\varepsilon(\psi^\varepsilon(t)) - W_2^\varepsilon(\psi^\varepsilon(t')), a\right\rangle_{\mathcal{A}',\mathcal{A}}\right| \leq \\ \sup_{a\in B} s_5(a) \|\psi^\varepsilon(t) - \psi^\varepsilon(t')\|_{L^2(\mathbb{R}^2)} \big(\|\psi^\varepsilon(t)\|_{L^2(\mathbb{R}^2)} + \|\psi^\varepsilon(t')\|_{L^2(\mathbb{R}^2)}\big),$$

and thus the asserted continuity with respect to time. However, passing to the limit $\varepsilon \to 0$, we are confronted with the possibility that different points of time $t$ could require different subsequences $(\varepsilon_k(t))_{k\in\mathbb{N}}$ for convergence to a two-scale measure. In that case, neither continuity with respect to time nor other properties of the two-scale Wigner functional would carry over to the two-scale measures. This difficulty is dealt with by restricting the analysis to diagonal observables.

PROPOSITION 4.7 *Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ be the solution of the Schrödinger equation* (1.1) *with initial data $(\psi_0^\varepsilon)_{\varepsilon>0}$ bounded in $L^2(\mathbb{R}^2, \mathbb{C}^2)$ such that the two-scale Wigner functions $(W_2^\varepsilon(\psi_0^\varepsilon))_{\varepsilon>0}$ converge to a two-scale measure $\rho_0$ in $\mathcal{A}'$.*

(i) *Then, for every $T > 0$ there is a subsequence $(\varepsilon_k)_{k\in\mathbb{N}}$ such that*

$$\lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\right\rangle_{\mathcal{A}',\mathcal{A}} \quad \textit{and} \quad \lim_{k\to\infty} \left\langle W^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\right\rangle_{\mathcal{S}',\mathcal{S}}$$

*exist uniformly in $t \in [0, T]$ for all $a \in \mathcal{A}$ and $a \in \mathcal{S}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$, respectively, with vanishing commutator $[a, V] = 0$ and* $\operatorname{supp}(a) \cap \{q = 0\} = \varnothing$.

(ii) *For scalar-valued $a$ with the same properties, the limits*

$$\lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}} =: \left\langle \rho_t^\pm, a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}}$$

*and*

$$\lim_{k\to\infty} \left\langle W^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\Pi^\pm\right\rangle_{\mathcal{S}',\mathcal{S}} =: \left\langle \mu_t^\pm, a\Pi^\pm\right\rangle_{\mathcal{S}',\mathcal{S}}$$

*define positive, bounded, scalar-valued Radon measures $\rho_t^\pm$ and $\mu_t^\pm$ on $(\mathbb{R}^4 \setminus \{q = 0\}) \times \overline{\mathbb{R}}$ and $\mathbb{R}^4 \setminus \{q = 0\}$, respectively, for all $t \in [0, T]$.*

(iii) *For scalar-valued observables a with the same properties, we have convergence of the full sequence*

$$\lim_{\varepsilon\to 0}\left\langle W_2^\varepsilon(\psi^\varepsilon(t)) - W_2^\varepsilon(\psi_0^\varepsilon)\circ\Phi_\pm^{-t}, a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}} = \lim_{\varepsilon\to 0}\left\langle W^\varepsilon(\psi^\varepsilon(t)) - W^\varepsilon(\psi_0^\varepsilon)\circ\Phi_\pm^{-t}, a\Pi^\pm\right\rangle_{\mathcal{S}',\mathcal{S}} = 0$$

*uniformly on time intervals* $[0, T]$ *such that for all* $t \in [0, T]$

$$\bigcup_{j\in\{\pm\}}\left\{\Phi_j^t(q, p) \mid \exists\eta\in\overline{\mathbb{R}} : (q, p, \eta)\in\operatorname{supp}(\rho_0)\right\}\cap\{q = 0\} = \varnothing.$$

*Remark* 4.8. Without incorporating nonadiabatic transitions, convergence of the full sequence is only obtained on time intervals where the leading-order dynamics can be described purely by classical transport. However, the uniform convergence of subsequences on arbitrary time intervals $[0, T]$ will be extended later on to convergence of the full sequence in the proof of Theorem 3.2.

PROOF: We write $a = \Pi^+ a\Pi^+ + \Pi^- a\Pi^-$ and study

$$\left\langle W_2^\varepsilon(\psi^\varepsilon(t)), \Pi^\pm a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}} = \left\langle\psi^\varepsilon(t), (\Pi^\pm a_\varepsilon\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\,\psi^\varepsilon(t)\right\rangle_{L^2(\mathbb{R}^2)}.$$

The assertions for the one-scale Wigner transform will follow immediately from the corresponding statements for the two-scale transform. As a first step, we establish the claimed uniform convergence with respect to time $t$. Cutting off the singularity of the projectors $\Pi^\pm$ at the crossing manifold $\{q = 0\}$, we choose a function $\phi\in C_{\mathrm{b}}^\infty(\mathbb{R}^2, [0, 1])$ such that $\phi = 1$ on $\{q\in\mathbb{R}^2 \mid \exists(p,\eta)\in\mathbb{R}^3 : (q, p, \eta)\in\operatorname{supp}(a)\}$ and $\phi(0) = 0$. We then have by Lemma 4.1

$$\Pi^\pm a_\varepsilon\Pi^\pm - (\phi^2\Pi^\pm)\,\sharp_\varepsilon\, a_\varepsilon\,\sharp_\varepsilon\,(\phi^2\Pi^\pm)\in S_{1/2}^{-1/2}(1)$$

and therefore

$$\left\langle W_2^\varepsilon(\psi^\varepsilon(t)), \Pi^\pm a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}} = \Big\langle(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t), a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t)\Big\rangle_{L^2} + \mathcal{O}(\sqrt{\varepsilon})$$

We choose initial data $\psi_0^\varepsilon$ in $D(H^\varepsilon)$. We observe that the first summand on the right-hand side of the previous equation defines a continuously differentiable function $f_{\psi_0^\varepsilon}^\varepsilon : \mathbb{R}\to\mathbb{C}$,

$$f_{\psi_0^\varepsilon}^\varepsilon(t) := \left\langle(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t), a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t)\right\rangle_{L^2}.$$

We have for the derivative

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}f_{\psi_0^\varepsilon}^\varepsilon(t) = {}&(\mathrm{i}\varepsilon)^{-1}\Big\langle(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)H^\varepsilon\psi^\varepsilon(t),\\ &\qquad a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t)\Big\rangle_{L^2}\\ &-(\mathrm{i}\varepsilon)^{-1}\Big\langle(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t),\\ &\qquad a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)(\phi^2\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)H^\varepsilon\psi^\varepsilon(t)\Big\rangle_{L^2}.\end{aligned}$$

We want to show that

$$\sup_{\varepsilon>0} \left\| \frac{\mathrm{d}}{\mathrm{d}t} f^{\varepsilon}_{\psi_0^\varepsilon}(\cdot) \right\|_\infty < \infty$$

to apply the Arzela-Ascoli theorem. Since $a_\varepsilon$ decays superpolynomially in $(q, p)$, semiclassical calculus gives

$$a_\varepsilon^* \sharp_\varepsilon (\phi^2 \Pi^\pm) \sharp_\varepsilon h - a_\varepsilon^* \sharp_\varepsilon (\phi\lambda^\pm) \sharp_\varepsilon (\phi \Pi^\pm) \in S^{-1}_{1/2}(1).$$

Thus, it remains to prove a uniform bound in $\varepsilon$ and $t$ for

$$\begin{aligned} &(\mathrm{i}\varepsilon)^{-1}\big\langle (\phi\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t), \\ &\qquad [\phi\lambda^\pm, a_\varepsilon]_{\sharp_\varepsilon}(q, -\mathrm{i}\varepsilon\nabla_q)(\phi\Pi^\pm)(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon(t)\big\rangle_{L^2}. \end{aligned} \tag{4.5}$$

However, $[\phi\lambda^\pm, a_\varepsilon]_{\sharp_\varepsilon} \in S^{-1}_{1/2}(1)$, since $[\phi\lambda^\pm, a_\varepsilon] = 0$ and

$$\{\phi\lambda^\pm, a_\varepsilon\} = \{\lambda^\pm, a_\varepsilon\} = \big(\nabla_p \lambda^\pm\big)\big(\nabla_q a\big)_\varepsilon - \big(\nabla_q\lambda^\pm\big)\big(\nabla_p a\big)_\varepsilon \in S^0_{1/2}(1),$$

where the last identity uses that $\{\lambda^\pm, q \wedge p\} = 0$ on $\mathbb{R}^4 \setminus \{q = 0\}$. Choosing general initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ and $\psi \in D(H^\varepsilon)$, we clearly have for $s, t \in \mathbb{R}$

$$\big|f^\varepsilon_{\psi_0^\varepsilon}(s) - f^\varepsilon_{\psi_0^\varepsilon}(t)\big| \le \big|f^\varepsilon_{\psi_0^\varepsilon}(s) - f^\varepsilon_{\psi}(s)\big| + \big|f^\varepsilon_{\psi}(s) - f^\varepsilon_{\psi}(t)\big| + \big|f^\varepsilon_{\psi}(t) - f^\varepsilon_{\psi_0^\varepsilon}(t)\big|.$$

Denoting the strongly continuous one-parameter group of $H^\varepsilon$ by $(U^\varepsilon(t))_{t\in\mathbb{R}}$, we obtain for the first term on the right-hand side of the above inequality (and analogously for the third one)

$$\begin{aligned} &\big|f^\varepsilon_{\psi_0^\varepsilon}(s) - f^\varepsilon_{\psi}(s)\big| \\ &\quad \le \big|\big\langle \psi_0^\varepsilon - \psi, U^\varepsilon(-s)\big(\Pi^\pm a_\varepsilon \Pi^\pm\big)\big(q, -\mathrm{i}\varepsilon\nabla_q\big)U^\varepsilon(s)\psi_0^\varepsilon\big\rangle_{L^2(\mathbb{R}^2)}\big| \\ &\qquad + \big|\big\langle \psi, U^\varepsilon(-s)\big(\Pi^\pm a_\varepsilon \Pi^\pm\big)\big(q, -\mathrm{i}\varepsilon\nabla_q\big)U^\varepsilon(s)\big(\psi - \psi_0^\varepsilon\big)\big\rangle_{L^2(\mathbb{R}^2)}\big| \\ &\quad \le \mathrm{const}\big\|\psi_0^\varepsilon - \psi\big\|_{L^2(\mathbb{R}^2)}\big(\|\psi_0^\varepsilon\|_{L^2(\mathbb{R}^2)} + \|\psi\|_{L^2(\mathbb{R}^2)}\big), \end{aligned}$$

while for the second term we have by the bound on the first derivative

$$\big|f^\varepsilon_{\psi}(s) - f^\varepsilon_{\psi}(t)\big| \le \mathrm{const}\,|s - t|.$$

Thus, regardless of the choice of initial data, the sequence $(f^\varepsilon_{\psi_0^\varepsilon})_{\varepsilon>0}$ is pointwise bounded and equicontinuous. By the Arzela-Ascoli theorem, we then have uniform convergence of a subsequence on compact subsets of $\mathbb{R}$, which shows the claimed uniform convergence on intervals $[0, T]$ for all $T > 0$.

Second, we prove that the two-scale limits define positive, bounded, scalar-valued Radon measures $\rho_t^\pm$ for all $t \in [0, T]$. Clearly, the limits define linear forms on the space of functions in $\mathcal{A}$ with support away from $\{q = 0\}$. By the standard arguments, which have already been invoked in the proof of Proposition 4.5, they extend to linear forms on compactly supported continuous functions on $\mathbb{R}^4 \times \overline{\mathbb{R}}$ with support away from $\{q = 0\}$. Such functions, however, are dense with respect to the sup norm in $C_c((\mathbb{R}^4 \setminus \{q = 0\}) \times \overline{\mathbb{R}}, \mathbb{C})$, and we obtain the measures $\rho_t^\pm$ on $(\mathbb{R}^4 \setminus \{q = 0\}) \times \overline{\mathbb{R}}$.

Third, we show the asserted transport properties. Omitting the subscript $\psi_0^\varepsilon$ of the function $f^\varepsilon_{\psi_0^\varepsilon}$ for notational simplicity, we have for scalar-valued observables $a \in \mathcal{A}$ with support away from $\{q = 0\}$

$$\lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\Pi^{\pm}\right\rangle_{\mathcal{A}',\mathcal{A}} = \lim_{k\to\infty} f^{\varepsilon_k}(t)$$

uniformly in $t \in [0, T]$. As already noted, the above uniform limit defines a measure $\rho_t^\pm$ on $(\mathbb{R}^4 \setminus \{q = 0\}) \times \overline{\mathbb{R}}$ for all $t \in [0, T]$. For initial data $\psi_0^\varepsilon \in D(H^\varepsilon)$, the function $t \mapsto f^\varepsilon(t)$ is continuously differentiable with a first-order derivative, whose leading-order term in $\varepsilon$ is given by the commutator expression in equation (4.5). Thus,

$$\begin{aligned}
\lim_{k\to\infty} \frac{\mathrm{d}}{\mathrm{d}t} f^{\varepsilon_k}(t) = \lim_{k\to\infty} \big\langle &(\phi\Pi^\pm)(q, -\mathrm{i}\varepsilon_k\nabla_q)\psi^{\varepsilon_k}(t),\\
&(\{\lambda^\pm, a\})_{\varepsilon_k}(q, -\mathrm{i}\varepsilon_k\nabla_q)(\phi\Pi^\pm)(q, -\mathrm{i}\varepsilon_k\nabla_q)\psi^{\varepsilon_k}(t)\big\rangle_{L^2}\\
&= \lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), \{\lambda^\pm, a\}\,\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}}\\
&= \int \{\lambda^\pm, a\}(q, p, \eta)\,\rho_t^\pm(\mathrm{d}q, \mathrm{d}p, \mathrm{d}\eta).
\end{aligned}$$

On the other hand, by the uniform convergence of $(f^{\varepsilon_k}(t))_{k\in\mathbb{N}}$,

$$\begin{aligned}
\lim_{k\to\infty} \frac{\mathrm{d}}{\mathrm{d}t} f^{\varepsilon_k}(t) = \frac{\mathrm{d}}{\mathrm{d}t} \lim_{k\to\infty} f^{\varepsilon_k}(t) &= \frac{\mathrm{d}}{\mathrm{d}t} \lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}}\\
&= \frac{\mathrm{d}}{\mathrm{d}t} \int a(q, p, \eta)\,\rho_t^\pm(\mathrm{d}q, \mathrm{d}p, \mathrm{d}\eta),
\end{aligned}$$

which implies

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_t^\pm = -\{\lambda^\pm, \rho_t\}$$

for $t \in [0, T]$ such that

$$\bigcup_{j\in\{\pm\}} \left\{\Phi_j^t(q, p) \mid \exists\, \eta \in \overline{\mathbb{R}} : (q, p, \eta) \in \operatorname{supp}(\rho_0)\right\} \cap \{q = 0\} = \varnothing,$$

or, equivalently, $\rho_t^\pm(q, p, \eta) = \rho_0^\pm(\Phi_\pm^t(q, p), \eta)$, or

$$\lim_{k\to\infty} \left\langle W^{\varepsilon_k}(\psi^{\varepsilon_k}(t)) - W^{\varepsilon_k}(\psi_0^{\varepsilon_k}) \circ \Phi_\pm^{-t}, a\Pi^\pm\right\rangle_{\mathcal{A}',\mathcal{A}} = 0.$$

The assumption on the measure $\rho_0$ guarantees that $(\langle W_2^\varepsilon(\psi_0^\varepsilon), a\Pi^\pm\rangle)_{\varepsilon>0}$ converges to measures $\rho_0^\pm$ without extraction of subsequences. Thus, every convergent subsequence of $(\langle W_2^\varepsilon(\psi^\varepsilon(t)), a\Pi^\pm\rangle)_{\varepsilon>0}$ converges to the same limit point, and therefore the whole sequence itself has to converge. Observing that

$$\begin{aligned}
&L^2(\mathbb{R}^2, \mathbb{C}^2) \times L^2(\mathbb{R}^2, \mathbb{C}^2) \to \mathbb{C},\\
&(f, g) \mapsto \left\langle U^\varepsilon(t) f, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\, U^\varepsilon(t) g\right\rangle_{L^2(\mathbb{R}^2)},
\end{aligned}$$

is a bounded bilinear form, we conclude the proof of the transport equation also for the case of general initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ by a density argument. $\square$

Proposition 4.7 also shows for the Wigner measures $\mu_t^\pm$ that $\mu_t^\pm = \mu_0^\pm \circ \Phi_\pm^{-t}$ on time intervals $[0, T]$ such that for all $t \in [0, T]$

$$\bigcup_{j \in \{\pm\}} \left\{ \Phi_j^t(q, p) \mid (q, p) \in \operatorname{supp}(\rho_0) \right\} \cap \{q = 0\} = \varnothing.$$

Since $\Phi_\pm^t$ leaves $I = \{q \wedge p = 0\}$ invariant,

$$\mu_t^\pm\big|_{\mathbb{R}^4 \setminus I} = (\mu_0^\pm \circ \Phi_\pm^{-t})\big|_{\mathbb{R}^4 \setminus I} \tag{4.6}$$

for all times $t \in \mathbb{R}$. While the diagonal components of a two-scale Wigner functional approximately satisfy classical transport equations, its off-diagonal elements vanish when taking time averages. For a similar statement in a slightly different context, see [23].

LEMMA 4.9 *Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ be the solution of the Schrödinger equation* (1.1) *with arbitrary initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$. Then, for all $a \in \mathcal{A}$ and all $t_1, t_2 \in \mathbb{R}$ there exists a positive constant $C = C(a, V, t_1, t_2) > 0$ depending on $a$, $V$, $t_1$, and $t_2$ such that for all $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$*

$$\left| \int_{t_1}^{t_2} \left\langle W_2^\varepsilon(\psi^\varepsilon(\tau)), [V, a] \right\rangle_{\mathcal{A}', \mathcal{A}} \mathrm{d}\tau \right| \le \sqrt{\varepsilon}\, C \|\psi_0^\varepsilon\|^2_{L^2(\mathbb{R}^2)}.$$

PROOF: Let $\psi_0^\varepsilon \in D(H^\varepsilon)$ and $a \in \mathcal{A}$. We have for all $\tau \in \mathbb{R}$

$$\mathrm{i}\varepsilon \frac{d}{\mathrm{d}\tau} \left\langle W_2^\varepsilon(\psi^\varepsilon(\tau)), a \right\rangle_{\mathcal{A}', \mathcal{A}} = \left\langle \psi^\varepsilon(\tau), \left[ H^\varepsilon, a_\varepsilon(q, -\mathrm{i}\varepsilon \nabla_q) \right] \psi^\varepsilon(\tau) \right\rangle_{L^2(\mathbb{R}^2)}.$$

Thus, we analyze the commutator

$$\left[ H^\varepsilon, a_\varepsilon(q, -\mathrm{i}\varepsilon \nabla_q) \right] = [h, a_\varepsilon]_{\sharp_\varepsilon}(q, -\mathrm{i}\varepsilon \nabla_q).$$

Since $a_\varepsilon$ is a Schwarz function, we have $a_\varepsilon \in S^0_{1/2}(\langle q \rangle^{-1} \langle p \rangle^{-2})$, and applying Lemma 4.1 we obtain $[h, a_\varepsilon]_{\sharp_\varepsilon} - [h, a_\varepsilon] =: \sqrt{\varepsilon}\, r^\varepsilon \in S^{-1/2}_{1/2}(1)$. Thus, with $[h, a_\varepsilon] = [V, a_\varepsilon]$,

$$\begin{aligned} \mathrm{i}\varepsilon \frac{d}{\mathrm{d}\tau} &\left\langle W_2^\varepsilon(\psi^\varepsilon(\tau)), a \right\rangle_{\mathcal{A}', \mathcal{A}} = \\ &\left\langle W_2^\varepsilon(\psi^\varepsilon(\tau)), [h, a] \right\rangle_{\mathcal{A}', \mathcal{A}} + \sqrt{\varepsilon} \left\langle \psi^\varepsilon(\tau), r^\varepsilon(q, -\mathrm{i}\varepsilon \nabla_q)\, \psi^\varepsilon(\tau) \right\rangle_{L^2(\mathbb{R}^2)}. \end{aligned} \tag{4.7}$$

Integration from $t_1$ to $t_2$ gives

$$\begin{aligned} \varepsilon \left| \int_{t_1}^{t_2} \frac{d}{\mathrm{d}\tau} \left\langle W_2^\varepsilon(\psi^\varepsilon(\tau)), a \right\rangle_{\mathcal{A}', \mathcal{A}} \mathrm{d}\tau \right| &= \varepsilon \left| \left\langle W_2^\varepsilon(\psi^\varepsilon(t_2)) - W_2^\varepsilon(\psi^\varepsilon(t_1)), a \right\rangle_{\mathcal{A}', \mathcal{A}} \right| \\ &\le \varepsilon s_5(a) \left\| \psi_0^\varepsilon \right\|^2_{L^2(\mathbb{R}^2)} \end{aligned}$$

and

$$\sqrt{\varepsilon}\left|\int_{t_1}^{t_2}\left\langle\psi^{\varepsilon}(\tau), r^{\varepsilon}(q,-\mathrm{i}\varepsilon\nabla_q)\psi^{\varepsilon}(\tau)\right\rangle_{L^2(\mathbb{R}^2)}\mathrm{d}\tau\right| \leq \sqrt{\varepsilon}\, c_4(r^{\varepsilon})|t_1-t_2|\left\|\psi_0^{\varepsilon}\right\|^2_{L^2(\mathbb{R}^2)},$$

which together with equation (4.7) yields the claimed bound for $\psi_0^{\varepsilon} \in D(H^{\varepsilon})$. A density argument concludes the proof also for general initial data $\psi_0^{\varepsilon} \in L^2(\mathbb{R}^2, \mathbb{C}^2)$. □

*Remark* 4.10. The proof of Lemma 4.9 applies to general matrix-valued Schrödinger equations with essentially self-adjoint Hamiltonian whose symbol is polynomially bounded, and to two-scaled Wigner functionals associated with more general submanifolds than the hypersurface of zero angular momentum $I = \{q \wedge p = 0\}$.

Purely off-diagonal symbols $a \in \mathcal{A}$ with $\operatorname{supp}(a) \cap \{q = 0\} = \varnothing$ can be written as $a = \Pi^+ a \Pi^- + \Pi^- a \Pi^+$, which implies $[V, a] = (\lambda^+ - \lambda^-)a$ and $a = [V, (\lambda^+ - \lambda^-)^{-1}a]$. Thus, we have for such off-diagonal observables

$$\left|\int_{t_1}^{t_2}\left\langle W_2^{\varepsilon}(\psi^{\varepsilon}(\tau)), a\right\rangle_{\mathcal{A}',\mathcal{A}}\,\mathrm{d}\tau\right| \leq \sqrt{\varepsilon}\, C\left\|\psi_0^{\varepsilon}\right\|^2_{L^2(\mathbb{R}^2)}.$$

## 4.5 Measures on $\mathbb{R}^2_{t,\tau} \times \mathbb{R}^4_{q,p} \times \overline{\mathbb{R}}_{\eta}$

We fix some time interval of interest $[0, T]$ with $T > 0$ and define a set of admissible observables on an extended phase space $[0,T]_t \times \mathbb{R}_\tau \times \mathbb{R}^4_{q,p}$ as

$$\mathcal{A}_{\mathrm{T}} := \left\{a \in C_{\mathrm{b}}^{\infty}(\mathbb{R}^7, \mathcal{L}(\mathbb{C}^2)) \mid a \text{ satisfies property } (\mathrm{P_T})\right\},$$

where

$$(\mathrm{P_T}) \qquad \operatorname{supp}(a) \subset [0,T] \times \mathbb{R}^6 \quad \text{and} \quad a(t,\tau,\cdot) \in \mathcal{A} \quad \text{for all } t, \tau \in \mathbb{R}.$$

For $a \in \mathcal{A}_{\mathrm{T}}$ we set $a_{\varepsilon}(t,q,\tau,p) =: a(t,q,\tau,p,(q \wedge p)/\sqrt{\varepsilon})$ and choose a cutoff function $\chi_{\mathrm{T}} \in C_{\mathrm{c}}^{\infty}(\mathbb{R}, \mathbb{R})$ such that $\chi_{\mathrm{T}}(t) = 1$ for $t \in [0, T]$. Then, we define for $\psi \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$

$$W^{\varepsilon}_{2,\mathrm{T}}(\psi) : \mathcal{A}_{\mathrm{T}} \to \mathbb{C}, \quad a \mapsto \langle \chi_{\mathrm{T}}\psi, a_{\varepsilon}(t,q,-\mathrm{i}\varepsilon\nabla_{t,q})\chi_{\mathrm{T}}\psi\rangle_{L^2(\mathbb{R}^3)},$$

which is a bounded linear functional by the rescaling identity (4.3) already used before. The alternative approach followed up in [10] applies to observables $a \in \mathcal{S}(\mathbb{R}^7, \mathcal{L}(\mathbb{C}^2))$ and treats $\psi \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ as a tempered distribution on $\mathbb{R}^3$. Then, $a_{\varepsilon} \in \mathcal{S}(\mathbb{R}^6, \mathcal{L}(\mathbb{C}^2))$, and the Weyl quantized operator is regularizing, that is,

$$a_{\varepsilon}(t,q,-\mathrm{i}\varepsilon\nabla_{t,q}) \in \mathcal{L}(\mathcal{S}'(\mathbb{R}^3,\mathbb{C}^2), \mathcal{S}(\mathbb{R}^3,\mathbb{C}^2));$$

see, for example, remark 2.5.6 in [22] or the proof of proposition II-56 in [26]. For symbols $a \in \mathcal{A}_{\mathrm{T}} \cap \mathcal{S}(\mathbb{R}^7, \mathcal{L}(\mathbb{C}^2))$ we have by Lemma 4.1

$$\chi_{\mathrm{T}} \sharp_{\varepsilon}\, a_{\varepsilon} \sharp_{\varepsilon}\, \chi_{\mathrm{T}} \sim a_{\varepsilon} \quad \text{in } S^0_{1/2}(1),$$

and therefore

$$a_{\varepsilon}(t,q,-\mathrm{i}\varepsilon\nabla_{t,q}) = \chi_{\mathrm{T}} a_{\varepsilon}(t,q,-\mathrm{i}\varepsilon\nabla_{t,q})\chi_{\mathrm{T}} \in \mathcal{L}(\mathcal{S}'(\mathbb{R}^3,\mathbb{C}^2), \mathcal{S}(\mathbb{R}^3,\mathbb{C}^2)).$$

Consequently,

$$\langle \chi_{\mathrm{T}}\psi, a_\varepsilon(t,q,-\mathrm{i}\varepsilon\nabla_{t,q})\chi_{\mathrm{T}}\psi\rangle_{L^2(\mathbb{R}^3)} = \langle \overline{\psi}, a_\varepsilon(t,q,-\mathrm{i}\varepsilon\nabla_{t,q})\psi\rangle_{\mathcal{S}',\mathcal{S}}.$$

For different cutoff functions $\chi_{\mathrm{T}}, \widetilde{\chi}_{\mathrm{T}} \in C_{\mathrm{c}}^\infty(\mathbb{R},\mathbb{R})$ with $\chi_{\mathrm{T}}(t) = \widetilde{\chi}_{\mathrm{T}}(t) = 1$ for $t \in [0,T]$, we have

$$\chi_{\mathrm{T}} \sharp_\varepsilon a_\varepsilon \sharp_\varepsilon \chi_{\mathrm{T}} \sim \widetilde{\chi}_{\mathrm{T}} \sharp_\varepsilon a_\varepsilon \sharp_\varepsilon \widetilde{\chi}_{\mathrm{T}} \quad \text{in } S^0_{1/2}(1),$$

and thus the independence of $W^\varepsilon_{2,\mathrm{T}}(\psi)$ from the choice of the cutoff function. Balancing the benefits of the two equivalent approaches of using a cutoff function in $L^2(\mathbb{R}^3)$ versus working with tempered distributions, we have preferred the natural setting of $L^2$ theory.

For sequences $(\psi^\varepsilon)_{\varepsilon>0}$ in $C(\mathbb{R}, L^2(\mathbb{R}^2,\mathbb{C}^2))$ with $\sup_{\varepsilon,t}\|\psi^\varepsilon(t)\|_{L^2(\mathbb{R}^2)} < \infty$, the Wigner transformed sequence $(W^\varepsilon_{2,\mathrm{T}}(\psi^\varepsilon))_{\varepsilon>0}$ has weak-star limit points $\rho_{\mathrm{T}}$ in $\mathcal{A}'_{\mathrm{T}}$, which are bounded, positive, matrix-valued Radon measures on $[0,T]\times\mathbb{R}^5\times\overline{\mathbb{R}}$. As before, we denote by $\nu_{\mathrm{T}}$ the restriction of a measure $\rho_{\mathrm{T}}$ to the set $\{(t,q,\tau,p,\eta) \in [0,T]\times\mathbb{R}^5\times\overline{\mathbb{R}} \mid (q,p)\in I\}$.

The following lemma addresses the localization of the measures $\rho_{\mathrm{T}}$. The analogous statement for semiclassical measures has been given in section 3 of [13].

LEMMA 4.11 *Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2,\mathbb{C}^2))$ be a solution of the Schrödinger equation* (1.1) *whose initial data $(\psi_0^\varepsilon)_{\varepsilon>0}$ form a bounded sequence in $L^2(\mathbb{R}^2,\mathbb{C}^2)$. Then, we have for the weak-star limit points $\rho_{\mathrm{T}} \in \mathcal{A}'_{\mathrm{T}}$ of $(W^\varepsilon_{2,\mathrm{T}}(\psi^\varepsilon))_{\varepsilon>0}$*

$$\operatorname{supp}(\rho_{\mathrm{T}}) \subset \left\{(t,\tau,q,p,\eta) \in [0,T]\times\mathbb{R}^5\times\overline{\mathbb{R}} \mid \tau + \frac{1}{2}|p|^2 = \pm|q|\right\}.$$

For the proof of Lemma 4.11, we refer to Appendix A.4. It remains to clarify the relation between two-scale measures on $\mathbb{R}^4_{q,p}\times\overline{\mathbb{R}}_\eta$ and their pendant on $\mathbb{R}^2_{t,\tau}\times\mathbb{R}^4_{q,p}\times\overline{\mathbb{R}}_\eta$.

LEMMA 4.12 *Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2,\mathbb{C}^2))$ be the solution of the Schrödinger equation* (1.1) *with initial data $(\psi_0^\varepsilon)_{\varepsilon>0}$ bounded in $L^2(\mathbb{R}^2,\mathbb{C}^2)$. Let $\rho_{\mathrm{T}}$ be a weak-star limit point of $(W^\varepsilon_{2,\mathrm{T}}(\psi^\varepsilon))_{\varepsilon>0}$, and let $\rho_t^\pm$ be the scalar measures introduced in Proposition* 4.7. *Then,*

$$\left\langle\rho_{\mathrm{T}}, \Pi^\pm a\Pi^\pm\right\rangle_{\mathcal{A}'_{\mathrm{T}},\mathcal{A}_{\mathrm{T}}} = \int\limits_{\mathbb{R}^6\times\overline{\mathbb{R}}} a^\pm(t,q,\tau,p,\eta)\rho_t^\pm(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\delta\left(\tau - \frac{1}{2}|p|^2 \mp |q|\right)\mathrm{d}t$$

*for all $a \in \mathcal{A}_{\mathrm{T}}$ with* $\operatorname{supp}(a) \subset [0,T]\times\mathbb{R}^6\setminus\{q=0\}$ *and* $a^\pm = \operatorname{tr}(a\Pi^\pm)$.

PROOF: Let $(\varepsilon_k)_{k\in\mathbb{N}}$ be a subsequence such that

$$W^{\varepsilon_k}_{2,\mathrm{T}}(\psi^{\varepsilon_k}) \overset{*}{\rightharpoonup} \rho_{\mathrm{T}}, \quad \operatorname{tr}\left(W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t))\Pi^\pm\right) \overset{*}{\rightharpoonup} \rho_t^\pm \text{ uniformly in } t \in [0,T].$$

Since $\rho_{\mathrm{T}}\,(\tau + \frac{1}{2}|p|^2 + V) = 0$, one has $\mathrm{supp}(\mathrm{tr}(\rho_{\mathrm{T}}\Pi^{\pm})) \subset \left\{\tau + \frac{1}{2}|p|^2 \pm |q| = 0\right\}$ and

$$\mathrm{tr}(\rho_{\mathrm{T}}(t,q,\tau,p,\eta)\,\Pi^{\pm}(q)) = \int_{\mathbb{R}} \mathrm{tr}(\rho_{\mathrm{T}}(t,q,\mathrm{d}\tau,p,\eta)\Pi^{\pm}(q))\delta\left(\tau + \frac{1}{2}|p|^2 \pm |q|\right)$$

as measures on $[0,T] \times (\mathbb{R}^2_q \setminus \{0\}) \times \mathbb{R}^3_{\tau,p} \times \overline{\mathbb{R}}_\eta$. Thus, it remains to show that

$$\rho_t^{\pm}(q,p,\eta) = \int_{\mathbb{R}} \mathrm{tr}\left(\rho_{\mathrm{T}}(t,q,\mathrm{d}\tau,p,\eta)\Pi^{\pm}(q)\right)$$

as measures on $[0,T] \times (\mathbb{R}^2_q \setminus \{0\}) \times \mathbb{R}^2_p \times \overline{\mathbb{R}}_\eta$. We have for $a = a(t,q,p,\eta) \in \mathcal{A}_{\mathrm{T}}$, which do not depend on $\tau$ and have support away from $\{q = 0\}$,

$$\begin{aligned}
&\int_{\mathbb{R}^6 \times \overline{\mathbb{R}}} \mathrm{tr}(a(t,q,p,\eta)\Pi^{\pm}(q))\rho_{\mathrm{T}}^{\pm}(\mathrm{d}t,\mathrm{d}q,\mathrm{d}\tau,\mathrm{d}p,\mathrm{d}\eta)\\
&\quad = \lim_{k\to\infty} \left\langle \chi_{\mathrm{T}}\psi^{\varepsilon_k}, (\Pi^{\pm}a_\varepsilon\Pi^{\pm})(t,q,-\mathrm{i}\varepsilon_k\nabla_q)\chi_{\mathrm{T}}\psi^{\varepsilon_k}\right\rangle_{L^2(\mathbb{R}^3,\mathbb{C}^2)}\\
&\quad = \lim_{k\to\infty} \int_{\mathbb{R}} |\chi_{\mathrm{T}}(t)|^2 \left\langle \psi^{\varepsilon_k}(t), (\Pi^{\pm}a_\varepsilon\Pi^{\pm})(t,q,-\mathrm{i}\varepsilon_k\nabla_q)\psi^{\varepsilon_k}(t)\right\rangle_{L^2(\mathbb{R}^2,\mathbb{C}^2)}\,\mathrm{d}t\\
&\quad = \int_{\mathbb{R}^5 \times \overline{\mathbb{R}}} \mathrm{tr}\left(a(t,q,p,\eta)\Pi^{\pm}\right)\rho_t^{\pm}(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\mathrm{d}t,
\end{aligned}$$

which concludes our proof. □

### 4.6 Measures near the Crossing: Results of Fermanian and Gérard

In the following, we summarize the part of the results of [10] that we will use for the proof of Theorem 3.2, tacitly using some of the simplifications worked out in [12]. Fermanian-Kammerer and Gérard introduce the involutive manifold

$$I_{\mathrm{FG}} := \{(t,\tau,q,p) \in \mathbb{R}^6 \mid q \wedge p = 0\},$$

which contains all the classical trajectories hitting the crossing $\{q = 0\}$, and a space of admissible observables

$$\begin{aligned}
\mathcal{A}_{\mathrm{FG}} := \Big\{ & a \in C^\infty(\mathbb{R}^7, \mathcal{L}(\mathbb{C}^2)) \mid \mathrm{supp}(a) \subset K \times \mathbb{R},\ K \subset \mathbb{R}^6 \setminus \{(t,\tau,0,0)\}\\
& \text{compact},\ \exists a_\infty \in C^\infty(\mathbb{R}^6 \times \{\pm 1\}, \mathcal{L}(\mathbb{C}^2)),\ \exists R > 0,\ \forall m \in \mathbb{R}^6,\\
& \forall |\eta| > R : a(m,\eta) = a_\infty(m,\mathrm{sgn}(\eta))\Big\}.
\end{aligned}$$

Theorem 1 of [10] shows that for a bounded sequence $(u^\varepsilon)_{\varepsilon>0}$ in $L^2(\mathbb{R}^3, \mathbb{C}^2)$ there exists a subsequence $(\varepsilon_k)_{k>0}$ of positive numbers and a positive Radon measure $\nu_{\mathrm{FG}}$ on $I_{\mathrm{FG}} \times \overline{\mathbb{R}}$ with values in $\mathcal{L}_{\mathrm{sa}}(\mathbb{C}^2)$ such that for all $a \in \mathcal{A}_{\mathrm{FG}}$

$$\lim_{\varepsilon_k \to 0} \int_{\mathbb{R}^6} \operatorname{tr}\left( W^{\varepsilon_k}(u^{\varepsilon_k})(t, \tau, q, p) a\left(t, \tau, q, p, \frac{q \wedge p}{\sqrt{\varepsilon_k}}\right)\right) \mathrm{d}t\, \mathrm{d}\tau\, \mathrm{d}q\, \mathrm{d}p =$$
$$\int_{I_{\mathrm{FG}} \times \overline{\mathbb{R}}} \operatorname{tr}(a(t, \tau, q, p, \eta) \nu_{\mathrm{FG}}(\mathrm{d}t, \mathrm{d}\tau, \mathrm{d}q, \mathrm{d}p, \mathrm{d}\eta))$$
$$+ \int_{\mathbb{R}^6 \setminus I_{\mathrm{FG}}} \operatorname{tr}(a_\infty(t, \tau, q, p, \operatorname{sgn}(q \wedge p)) \mu(\mathrm{d}t, \mathrm{d}\tau, \mathrm{d}q, \mathrm{d}p)),$$

where $(W^\varepsilon(u^\varepsilon))_{\varepsilon>0}$ and $\mu$ are Wigner transforms and a Wigner measure of $(u^\varepsilon)_{\varepsilon>0}$. Theorem 2$'$ of [10] associates with the solution $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ of the Schrödinger equation (1.1) a measure $\nu_{\mathrm{FG}}$ on $\mathbb{R}^6 \times \overline{\mathbb{R}}$, which decomposes as

$$\nu_{\mathrm{FG}} = \nu_{\mathrm{FG}}^+ \Pi^+ + \nu_{\mathrm{FG}}^- \Pi^-$$

with scalar measures $\nu_{\mathrm{FG}}^\pm$ supported in $J^{\pm,p} \cup J^{\pm,f}$. For the definition of the sets $J^{\pm,p}$, $J^{\pm,f}$ they choose a point $(t_0, \tau_0, 0, p_0, \eta_0)$ inside the crossing manifold

$$S_{\mathrm{FG}} := \left\{(t, \tau, 0, p, \eta) \in \mathbb{R}^6 \times \overline{\mathbb{R}} \mid t \in \mathbb{R},\ \tau = -\frac{1}{2}|p|^2, p \neq 0, \eta \in \overline{\mathbb{R}}\right\},$$

a neighborhood $(t_0, \tau_0, 0, p_0) \in U \subset \mathbb{R}^6$, and set

$$J^{\pm,p} := \left\{(t+s, \tau, \Phi_\pm^s(0,p), \eta) \in \mathbb{R}^6 \times \overline{\mathbb{R}} \mid (t, \tau, 0, p) \in U,\ s < 0 \text{ suff. small}\right\},$$
$$J^{\pm,f} := \left\{(t+s, \tau, \Phi_\pm^s(0,p), \eta) \in \mathbb{R}^6 \times \overline{\mathbb{R}} \mid (t, \tau, 0, p) \in U,\ s > 0 \text{ suff. small}\right\},$$

where $\Phi_\pm^t$ are the classical flows associated with the Hamiltonian systems (1.3). Outside the crossing set on $(J^{\pm,p} \cup J^{\pm,f}) \setminus S_{\mathrm{FG}}$, the measures $\nu_{\mathrm{FG}}^\pm$ satisfy transport equations

$$\nu_{\mathrm{FG}}^\pm(t, \tau, q, p, \eta) = \nu_{\mathrm{FG}}^\pm(0, \tau, \Phi_\pm^t(q,p), \eta);$$

see theorem 2$'$ of [10] or proposition 2 of [12]. Denoting restrictions of the measures $\nu_{\mathrm{FG}}^\pm$ to $J^{\pm,p} \cap S_{\mathrm{FG}}$ and $J^{\pm,f} \cap S_{\mathrm{FG}}$ by $\nu_{S_{\mathrm{FG}}}^{\pm,p}$ and $\nu_{S_{\mathrm{FG}}}^{\pm,f}$, respectively, theorem 3 of [10] shows the Landau-Zener type formula

$$\begin{pmatrix} \nu_{S_{\mathrm{FG}}}^{+,f} \\ \nu_{S_{\mathrm{FG}}}^{-,f} \end{pmatrix} = \begin{pmatrix} 1-T & T \\ T & 1-T \end{pmatrix} \begin{pmatrix} \nu_{S_{\mathrm{FG}}}^{+,p} \\ \nu_{S_{\mathrm{FG}}}^{-,p} \end{pmatrix} \tag{4.8}$$

with $T = T(p, \eta) = \exp(-\pi\eta^2/|p|^3)$ if $\nu_{S_{\mathrm{FG}}}^{+,p}$ and $\nu_{S_{\mathrm{FG}}}^{-,p}$ are mutually singular on $S_{\mathrm{FG}}$. A sufficient condition to meet this singularity requirement for positive times $t \geq 0$ is the choice of initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2, \mathbb{C}^2)$ satisfying $\Pi^- \psi_0^\varepsilon = 0$, since then $\nu_{S_{\mathrm{FG}}}^{-,p}|_{\{t \geq 0\}} \equiv 0$.

## 4.7 A Semigroup for Two-Scale Measures

In complete analogy to the definition of the $\varepsilon$-dependent semigroup $\mathcal{L}_\varepsilon^t$ for the diagonal components $(w_+^\varepsilon(\psi(t)), w_-^\varepsilon(\psi(t)))$ of the Wigner function, we define a semigroup for the two-scale Wigner measures $(\rho_t^+, \rho_t^-)$ and $(\nu_t^+, \nu_t^-)$ in what follows.

We introduce the right-continuous random trajectories

$$\mathcal{J}^{(q,p,\eta,j)} : [0,\infty) \to \mathbb{R}^4 \times \overline{\mathbb{R}}_\eta \times \{-1, 1\},$$

where $\mathcal{J}^{(q,p,\eta,j)}(t) = (\Phi_j^t(q,p), \eta, j)$ as long as $\Phi_j^t(q,p) \notin S$. Whenever the flow $\Phi_j^t(q,p)$ hits the jump manifold $S$, a jump from $j$ to $-j$ occurs with probability

$$T(p,\eta) = \exp\left(-\pi \frac{\eta^2}{|p|^3}\right).$$

The random trajectories $\mathcal{J}^{(q,p,\eta,j)}$ define a Markov process

$$\left\{\mathbb{P}^{(q,p,\eta,j)} \mid (q,p,\eta,j) \in \mathbb{R}^4 \times \overline{\mathbb{R}}_\eta \times \{-1,1\}\right\}.$$

The pendant $\mathcal{C}^2$ to the space of observables $\mathcal{C}$ is defined as follows:

DEFINITION 4.13 A continuous compactly supported function $f \in C_c((\mathbb{R}^4 \setminus S) \times \overline{\mathbb{R}} \times \{-1,1\}, \mathbb{C})$ belongs to to the space $\mathcal{C}^2$ if the following boundary conditions at $(S \setminus S_{cl}) \times \overline{\mathbb{R}} \times \{-1,1\}$ are satisfied:

$$\lim_{\delta\to+0} f(q-\delta p, p-\delta j q/|q|, \eta, j) = T(p,\eta) \lim_{\delta\to+0} f\left(q+\delta p, p-\delta j\frac{q}{|q|}, \eta, -j\right),$$

$$\lim_{\delta\to+0} f(q-\delta p, p-\delta j q/|q|, \eta, j) = (1-T(p,\eta)) \lim_{\delta\to+0} f\left(q+\delta p, p+\delta j\frac{q}{|q|}, \eta, j\right).$$

By construction, the semigroup

$$(\mathcal{T}^t f)(q,p,\eta,j) := \mathbb{E}^{(q,p,\eta,j)} f(\mathcal{J}^{(q,p,\eta,j)}(t)), \quad t \geq 0,$$

leaves the space $\mathcal{C}^2$ invariant, that is, $\mathcal{T}^t : \mathcal{C}^2 \to \mathcal{C}^2$ for all $t \geq 0$. We denote the space of functions $a \in C_c((\mathbb{R}^4 \setminus S) \times \mathbb{R}, \mathcal{L}(\mathbb{C}^2))$ such that $a = a^+\Pi^+ + a^-\Pi^-$ with $(a^+, a^-) \in \mathcal{C}^2$ by $\mathcal{C}^2_{\mathrm{diag}}$ and set for $a \in \mathcal{C}^2_{\mathrm{diag}}$

$$\mathcal{T}_\pm^t a := (\mathcal{T}^t(a^+, a^-))^\pm, \quad \mathcal{T}^t a := (\mathcal{T}_+^t a)\Pi^+ + (\mathcal{T}_-^t a)\Pi^-, \quad t \geq 0.$$

We note that $\mathcal{T}^t$ leaves the space $\mathcal{C}^2_{\mathrm{diag}}$ invariant. To work exclusively on the subspaces $\mathrm{Ran}(\Pi^\pm)$, we will also need

$$\mathcal{T}_\pm^t a := \mathcal{T}_\pm^t(a\Pi^\pm)$$

for scalar-valued $a \in C_c((\mathbb{R}^4 \setminus S) \times \overline{\mathbb{R}}, \mathbb{C})$. By duality, we define for matrix-valued Radon measures $\rho$ on $\mathbb{R}^4 \times \overline{\mathbb{R}}$ with $\operatorname{supp}(\rho) \cap (S \times \overline{\mathbb{R}}) = \varnothing$, the matrix-valued measure $\mathcal{T}^t \rho$ on $(\mathbb{R}^4 \setminus S) \times \overline{\mathbb{R}}$; that is, we set

$$\int_{(\mathbb{R}^4\setminus S)\times\overline{\mathbb{R}}} \operatorname{tr}\big(a(q,p,\eta)(\mathcal{T}^t\rho)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big) := \int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big((\mathcal{T}^t a)(q,p,\eta)\rho(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big)$$

for $a \in \mathcal{C}^2_{\mathrm{diag}}$. Having fixed our notation and definitions, we can formulate the key observation for the proof of Theorem 3.2.

LEMMA 4.14 *Let $\psi^\varepsilon(t) \in C(\mathbb{R}, L^2(\mathbb{R}^2, \mathbb{C}^2))$ be the solution of the Schrödinger equation* (1.1) *with initial data $(\psi_0^\varepsilon)_{\varepsilon>0}$ bounded in $L^2(\mathbb{R}^2, \mathbb{C}^2)$. Let $T > 0$ and $\rho_t^\pm$, $t \in [0, T]$, be the scalar measures on $(\mathbb{R}^4 \setminus \{q = 0\}) \times \overline{\mathbb{R}}$ introduced in Proposition* 4.7. *If*

$$\rho_0^- = 0 \quad \textit{and} \quad \operatorname{supp}(\rho_0^+) \cap (S \times \overline{\mathbb{R}}) = \varnothing,$$

*then the restrictions $\nu_t^\pm$ of the measures $\rho_t^\pm$ to $I \times \overline{\mathbb{R}}$ satisfy*

$$\int_{I\times\overline{\mathbb{R}}} a(q,p,\eta)\nu_t^\pm(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta) = \int_{I\times\overline{\mathbb{R}}} \big(\mathcal{T}_\pm^t a\big)(q,p,\eta)\nu_0^\pm(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)$$

*for all scalar-valued $a \in \mathcal{A}$ with* $\operatorname{supp}(a) \cap (S \times \overline{\mathbb{R}}) = \varnothing$ *and for all $t \in [0, T]$.*

PROOF: We have to work with measures on $\mathbb{R}^4 \times \overline{\mathbb{R}}$ and on $[0, T] \times \mathbb{R}^5 \times \overline{\mathbb{R}}$ in the following. For all such measures $m$, which have support away from the jump manifold $S$, we define the measure $\mathcal{T}_\pm^t m$ by

$$\int a(x)\big(\mathcal{T}_\pm^t m\big)(\mathrm{d}x) := \int \big(\mathcal{T}_\pm^t a\big)(x)m(\mathrm{d}x),$$

where the scalar-valued $a$ is either in $\mathcal{A}$ with support away from $S$ or an observable in $\mathcal{A}_\mathrm{T} \cap \mathcal{A}_\mathrm{FG}$ with the same support property. The measure $\mathcal{T}_\pm^t(\nu_0^\pm\delta(\tau - \lambda^\pm)\mathrm{d}t)$ satisfies the same transport properties and jump conditions at $I \cap S = \{q = 0\}$ as the measure $\nu_{\mathrm{FG}}^\pm$. Hence

$$\mathcal{T}_\pm^t\big(\nu_0^\pm\delta(\tau-\lambda^\pm)\mathrm{d}t\big) = \nu_{\mathrm{FG}}^\pm \quad \text{on } \mathcal{A}_\mathrm{T} \cap \mathcal{A}_\mathrm{FG}.$$

Since the Hamiltonian flow $\Phi_\pm^t$ conserves energy $\lambda^\pm(q,p) = \frac{1}{2}|p|^2 \pm |q|$, and since $\lambda^+(q,p) = \lambda^-(q,p)$ for $(q,p) \in I \cap S = \{q = 0\}$, we have

$$\mathcal{T}_\pm^t\big(\nu_0^\pm\delta(\tau-\lambda^\pm)\mathrm{d}t\big) = \big(\mathcal{T}_\pm^t\nu_0^\pm\big)\delta(\tau-\lambda^\pm)\mathrm{d}t \quad \text{on } \mathcal{A}_\mathrm{T}.$$

On the other hand, by Lemma 4.12

$$\nu_{\mathrm{FG}}^\pm = \nu_t^\pm\delta(\tau-\lambda^\pm)\mathrm{d}t \quad \text{on } \mathcal{A}_\mathrm{T} \cap \mathcal{A}_\mathrm{FG},$$

and therefore

$$\nu_t^{\pm}\delta(\tau-\lambda^{\pm})\mathrm{d}t = \big(\mathcal{T}_{\pm}^t \nu_0^{\pm}\big)\delta(\tau-\lambda^{\pm})\mathrm{d}t \quad \text{on } \mathcal{A}_{\mathrm{T}} \cap \mathcal{A}_{\mathrm{FG}}.$$

By continuity with respect to time $t$, we then have

$$\nu_t^{\pm} = \mathcal{T}_{\pm}^t \nu_0^{\pm} \quad \text{on } C_{\mathrm{c}}^{\infty}(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2))$$

for all times $t \in [0, T]$, and since $\nu_t^{\pm}$ is a positive distribution, by density the claimed identity on $\mathcal{A}$. □

## 5 Proof of the Main Theorem

With the preparation of Section 4 the proof of Theorem 3.2 is now straightforward.

PROOF OF THEOREM 3.2: We will establish (3.1) by proving separately that uniformly in $t \in [0, T]$

$$\lim_{\varepsilon\to 0}\int_{\mathbb{R}^4} \mathrm{tr}\,(W^{\varepsilon}(\psi^{\varepsilon}(t))(q,p)a(q,p))\,\mathrm{d}q\,\mathrm{d}p = \int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \mathrm{tr}\left(a(q,p)\left(\mathcal{T}^t\rho_0\right)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\right) \tag{5.1}$$

where the key ingredient is Lemma 4.14, and

$$\lim_{\varepsilon\to 0}\int_{\mathbb{R}^4} \mathrm{tr}\left(W^{\varepsilon}_{\mathrm{LZ}}(t)(q,p)a(q,p)\right)\mathrm{d}q\,\mathrm{d}p = \int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \mathrm{tr}\left(a(q,p)\left(\mathcal{T}^t\rho_0\right)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\right) \tag{5.2}$$

uniformly in $t \in [0, T]$, which basically holds by construction of the semigroups.

We write the diagonal observables $a$ under consideration again in the form $a = \mathrm{tr}(a\,\Pi^+)\Pi^+ + \mathrm{tr}(a\Pi^-)\Pi^- =: a^+\Pi^+ + a^-\Pi^-$. Note that such observables can be viewed as $\eta$-independent elements of $\mathcal{A}$. By Proposition 4.7, there exists a subsequence $(\varepsilon_k)_{k\in\mathbb{N}}$ depending on $T > 0$ such that

$$\lim_{k\to\infty}\left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\right\rangle_{\mathcal{A}',\mathcal{A}}$$

exists uniformly in $t \in [0, T]$. In the following, we will show that all such convergent subsequences of

$$\left(\left\langle W_2^{\varepsilon}(\psi^{\varepsilon}(t)), a\right\rangle_{\mathcal{A}',\mathcal{A}}\right)_{\varepsilon>0} \tag{5.3}$$

converge to the same limit point

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \mathrm{tr}\left(a(q,p)\big(\mathcal{T}^t\rho_0\big)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\right)$$

uniformly in $t$, and thus the whole sequence itself has to converge towards this limit point uniformly in $t$. By the definition of the measures $\mu_t^\pm$ and $\nu_t^\pm$, we have uniformly in $t$

$$\lim_{k\to\infty}\int_{\mathbb{R}^4} \operatorname{tr}\left(W^{\varepsilon_k}(\psi^{\varepsilon_k}(t))a(q,p)\right)\mathrm{d}q\,\mathrm{d}p = \lim_{k\to\infty}\left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}(t)), a\right\rangle_{\mathcal{A}',\mathcal{A}} =$$
$$\sum_{j\in\{\pm\}}\Bigg(\int_{\mathbb{R}^4\setminus I} a^j(q,p)\mu_t^j(\mathrm{d}q,\mathrm{d}p) + \int_{I\times\overline{\mathbb{R}}} a^j(q,p)\nu_t^j(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\Bigg).$$

By identity (4.6) following Proposition 4.7,

$$\int_{\mathbb{R}^4\setminus I} a^\pm(q,p)\mu_t^\pm(\mathrm{d}q,\mathrm{d}p) = \int_{\mathbb{R}^4\setminus I} \left(a^\pm\circ\Phi_\pm^{-t}\right)(q,p)\mu_0^\pm(\mathrm{d}q,\mathrm{d}p).$$

Since $\int_{S_\delta}|W^\varepsilon(\psi_0^\varepsilon)(q,p)|\mathrm{d}q\,\mathrm{d}p\to 0$ as $\varepsilon\to 0$, we also have

$$\int_{\mathbb{R}^4} W^\varepsilon(\psi_0^\varepsilon)(q,p)a(q,p)\mathrm{d}q\,\mathrm{d}p\to 0$$

as $\varepsilon\to 0$ for all $a\in\mathcal{S}(\mathbb{R}^4,\mathcal{L}(\mathbb{C}^2))$ with $\operatorname{supp}(a)\subset S_\delta$. This means $\operatorname{supp}(\mu_0)\cap S_\delta=\varnothing$, which in turn implies $\operatorname{supp}(\rho_0)\cap(S_\delta\times\overline{\mathbb{R}})=\varnothing$. By Lemma 4.14, we then have for the two-scale measures $\nu_t^\pm$

$$\int_{I\times\overline{\mathbb{R}}} a^\pm(q,p)\nu_t^+(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta) = \int_{I\times\overline{\mathbb{R}}} \left(\mathcal{T}_\pm^t a\right)(q,p,\eta)\nu_0^+(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta).$$

Thus, uniformly in $t$

$$\lim_{k\to\infty}\int_{\mathbb{R}^4} \operatorname{tr}\left(W^{\varepsilon_k}(\psi^{\varepsilon_k}(t))a(q,p)\right)\mathrm{d}q\,\mathrm{d}p =$$
$$\sum_{j\in\{\pm\}}\Bigg(\int_{\mathbb{R}^4\setminus I} \left(a^j\circ\Phi_j^{-t}\right)(q,p)\mu_0^j(\mathrm{d}q,\mathrm{d}p) + \int_{I\times\overline{\mathbb{R}}} \left(\mathcal{T}_j^t a\right)(q,p,\eta)\nu_0^j(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\Bigg),$$

and by definition of the measure $\rho_0$ and the semigroup $\mathcal{T}^t$

$$\sum_{j\in\{\pm\}}\int_{I\times\overline{\mathbb{R}}} \left(\mathcal{T}_j^t a\right)(q,p,\eta)\nu_0^j(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta) =$$
$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}((\mathcal{T}^t a)(q,p)\rho_0(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)) - \sum_{j\in\{\pm\}}\int_{\mathbb{R}^4\setminus I} \left(\mathcal{T}_j^t a\right)(q,p,\infty)\mu_0^j(\mathrm{d}q,\mathrm{d}p).$$

Since $T(q,p,\infty)=0$, we have

$$\int_{\mathbb{R}^4\setminus I} \big(\mathcal{T}_\pm^t a\big)(q,p,\infty)\mu_0^\pm(\mathrm{d}q,\mathrm{d}p) = \int_{\mathbb{R}^4\setminus I} \big(a^\pm\circ\Phi_\pm^{-t}\big)(q,p)\mu_0^\pm(\mathrm{d}q,\mathrm{d}p),$$

and therefore, uniformly in $t$,

$$\lim_{k\to\infty}\int_{\mathbb{R}^4} \operatorname{tr}\big(W^{\varepsilon_k}(\psi^{\varepsilon_k}(t))a(q,p)\big)\mathrm{d}q\,\mathrm{d}p = \int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big(a(q,p)\big(\mathcal{T}^t\rho_0\big)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big).$$

The preceding arguments show that all convergent subsequences of the bounded sequence in (5.3) converge to the same limit, and thus the sequence has to converge itself. This proves (5.1).

In order to establish (5.2), i.e., to lift the semigroup acting on the measures to a semigroup acting on functionals, we first have to remove a neighborhood of $S$. Let $\chi\in C^\infty(\mathbb{R},\mathbb{R})$ be a smooth function such that $\chi=0$ on $[-\delta/2,\delta/2]$ and $\chi=1$ on $\mathbb{R}\setminus[-\delta,\delta]$. Since $\operatorname{supp}(\rho_0)\cap(S_\delta\times\overline{\mathbb{R}})=\varnothing$, we have

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big(a(q,p)\big(\mathcal{T}^t\rho_0\big)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big) = \int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big(\chi(q\cdot p)\big(\mathcal{T}^t a\big)(q,p,\eta)\rho_0(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big).$$

Denoting $\widetilde{\chi}(q,p):=\chi(q\cdot p)$, the set $\{\widetilde{\chi}\big(\mathcal{T}^t a\big)\mid t\in[0,T]\}$ is a bounded subset of $\mathcal{A}$. Since weak-star convergence and strong convergence in $\mathcal{A}'$ coincide, we get uniformly in $t$

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big(a(q,p)\big(\mathcal{T}^t\rho_0\big)(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)\big) = \lim_{\varepsilon\to 0}\big\langle W_2^\varepsilon(\psi_0^\varepsilon),\widetilde{\chi}\big(\mathcal{T}^t a\big)\big\rangle_{\mathcal{A}',\mathcal{A}}.$$

Since the initial data have no mass near the jump manifold $S$, we find that

$$\begin{aligned}
&\lim_{\varepsilon\to 0}\big\langle W_2^\varepsilon(\psi_0^\varepsilon),\widetilde{\chi}\big(\mathcal{T}^t a\big)\big\rangle_{\mathcal{A}',\mathcal{A}}\\
&\quad=\lim_{\varepsilon\to 0}\int_{\mathbb{R}^4}\operatorname{tr}\big(W^\varepsilon(\psi_0^\varepsilon)(q,p)\chi(q\cdot p)\big(\mathcal{L}_\varepsilon^t a\big)(q,p)\big)\mathrm{d}q\,\mathrm{d}p\\
&\quad=\lim_{\varepsilon\to 0}\int_{\mathbb{R}^4}\operatorname{tr}\big(W^\varepsilon\big(\psi_0^\varepsilon\big)(q,p)\big(\mathcal{L}_\varepsilon^t a\big)(q,p)\big)\mathrm{d}q\,\mathrm{d}p\\
&\quad=\lim_{\varepsilon\to 0}\int_{\mathbb{R}^4}\operatorname{tr}\big(W_{\mathrm{LZ}}^\varepsilon(t)(q,p)a(q,p)\big)\mathrm{d}q\,\mathrm{d}p,
\end{aligned}$$

uniformly in $t$. This shows (5.2) and the proof is complete. □

# Appendix: Two-Scale Semiclassical Calculus

Here we collect proofs of some of the two-scale results used in Section 4.

## A.1 Moyal Multiplication with Symbols in $S^0_{1/2}$

We start with the proof of Lemma 4.1, which concerns the asymptotic expansion of the Moyal product between the symbol classes $S^0_0(m_1)$ and $S^0_{1/2}(m_2)$.

PROOF OF LEMMA 4.1: By proposition 7.6 in [7], the map

$$\exp\left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right) : \mathcal{S}(\mathbb{R}^{4n}, \mathcal{L}(\mathbb{C}^2)) \to \mathcal{S}(\mathbb{R}^{4n}, \mathcal{L}(\mathbb{C}^2))$$

extends continuously to an operator $S^0_{1/2}(m_1 \otimes m_2) \to S^0_{1/2}(m_1 \otimes m_2)$. Thus, we only have to show the asymptotic expansion. Observing that every differentiation of $b$ produces a factor $\varepsilon^{-1/2}$, it is clear that $c_j \in S^{-j/2}_{1/2}(m_1 m_2)$. Proving (4.1), one defines the smooth mapping

$$E : \mathbb{R} \to \mathcal{L}\big(S^0_{1/2}(m_1 \otimes m_2)\big), \quad t \mapsto E(t) := \exp\left(\frac{\mathrm{i}t}{2}(D_p D_{q'} - D_q D_{p'})\right).$$

Taylor expansion of order $N$ around $t = 0$ gives

$$E(\varepsilon) = \sum_{j=0}^{N} \varepsilon^j \frac{1}{j!}\big(\partial_t^j E\big)(0) + \varepsilon^{N+1}\frac{1}{N!}\int_0^1 (1-t)^N \big(\partial_t^{N+1} E\big)(\varepsilon t)\mathrm{d}t.$$

The first summand is nothing else than

$$\sum_{j=0}^{N} \frac{1}{j!}\left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right)^j,$$

while the remainder term can be rewritten as

$$\frac{1}{N!}\int_0^1 (1-t)^N \left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right)^{N+1} E(\varepsilon t)\mathrm{d}t.$$

Since $E(\varepsilon t)$ preserves the symbol class $S^0_{1/2}(m_1 \otimes m_2)$, and since every differentiation of $b$ produces an extra factor $\varepsilon^{-1/2}$,

$$\text{(A.1)} \quad \int_0^1 (1-t)^N \left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right)^{N+1} E(\varepsilon t) a(q,p) b(q',p')\mathrm{d}t\bigg|_{q'=q,\, p'=p}$$

is a symbol in $S^{-(N+1)/2}_{1/2}(m_1 m_2)$, and we are done. $\square$

For the proof of the Gårding-type inequality we need the following observation on the Moyal product of $\varepsilon$-scaled polynomials.

LEMMA A.1 *Let $a, b \in C^\infty(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2))$ and $g \in C^\infty(\mathbb{R}^4, \mathbb{R})$ be polynomials and $m_1$ and $m_2$ be order functions such that $a_\varepsilon \in S^0_{1/2}(m_1)$ and $b_\varepsilon \in S^0_{1/2}(m_2)$. Then there exists $N \in \mathbb{N}$ depending on the polynomial degrees of $a$, $b$, and $g$ such that*

$$(a_\varepsilon \sharp_\varepsilon b_\varepsilon)(q, p) =$$
$$\sum_{j=0}^{N} \frac{1}{j!}\left(\left(\frac{\mathrm{i}\varepsilon}{2}(D_p D_{q'} - D_q D_{p'})\right)^j a_\varepsilon(q, p) b_\varepsilon(q', p')\right)\bigg|_{q=q', p=p'} =: \sum_{j=0}^{N} c_j(q, p)$$

*with $c_j \in S^{-j/2}_{1/2}(m_1 m_2)$.*

PROOF: The proof of Lemma 4.1 already implies $a_\varepsilon \sharp_\varepsilon b_\varepsilon = \sum_{j=1}^N c_j$. Hence it remains to show that $c_j \in S^{-j/2}_{1/2}(m_1 m_2)$. One computes that

$$\{a_\varepsilon, b_\varepsilon\} = (\partial_p a\, \partial_q b - \partial_q a\, \partial_p b)_\varepsilon$$
$$+ \frac{1}{\sqrt{\varepsilon}}(\partial_p a\, \partial_\eta b\, \partial_q g + \partial_\eta a\, \partial_p g\, \partial_q b - \partial_q a\, \partial_\eta b\, \partial_p g - \partial_\eta a\, \partial_q g\, \partial_p b)_\varepsilon.$$

This identity shows a cancellation of "bad terms" of order $1/\varepsilon$, which multiply $\eta$-derivatives of $a$ with $\eta$-derivatives of $b$. Thus,

$$\varepsilon\{a_\varepsilon, b_\varepsilon\} \in S^{-1/2}_{1/2}(m_1 m_2).$$

The same reasoning yields

$$\varepsilon^j (D_p D_{q'} - D_q D_{p'})^j a_\varepsilon(q, p) b_\varepsilon(q', p')\big|_{q=q', p=p'} \in S^{-j/2}_{1/2}(m_1 m_2)$$

for all $j \in \mathbb{N}$. □

*Remark* A.2. An extension of Lemma A.1 to nonpolynomial symbols would require control on the remainder term (A.1) in the asymptotic expansion, which we have not been able to achieve.

## A.2 Gårding Inequalities

The following proof of the Gårding-type inequality, Proposition 4.3, relies on Lemma A.1 and an approximation of the nonnegative symbol $a_\varepsilon$ by the Moyal product $p_\varepsilon \sharp_\varepsilon p_\varepsilon$ of a polynomial $p$.

PROOF OF PROPOSITION 4.3: We proceed by defining a smooth square root, taking a cutoff in the $\eta$-component, approximating polynomially, and finally expanding a Moyal product.

*Step* 1. A SMOOTH SQUARE ROOT. Let $\delta > 0$. Since $a \in \mathcal{A}$ decays superpolynomially in $(q, p)$, one finds a cutoff function $\chi^\delta \in C^\infty_{\mathrm{c}}(\mathbb{R}^4, [0, 1])$ such that $s_5(a - (\chi^\delta)^2 a) < \delta$. One defines

$$b^\delta(q, p, \eta) := \chi^\delta(q, p)\sqrt{a(q, p, \eta) + \delta} \in \mathcal{A},$$

where $\sqrt{\cdot}$ denotes the positive square root of the strictly positive matrix $a(q, p, \eta)+\delta$. By the upper bound (4.4),

$$\begin{aligned} a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q) &= \big(b_\varepsilon^\delta\big)^2(q, -\mathrm{i}\varepsilon\nabla_q) + \big(a_\varepsilon - (\chi^\delta)^2 a_\varepsilon\big)(q, -\mathrm{i}\varepsilon\nabla_q) \\ &\quad - \delta(\chi^\delta)^2(q, -\mathrm{i}\varepsilon\nabla_q) \\ &= \big(b_\varepsilon^\delta\big)^2(q, -\mathrm{i}\varepsilon\nabla_q) + \mathcal{O}(\delta) \end{aligned}$$

uniformly in $\varepsilon > 0$ as $\delta \to 0$.

*Step* 2. A CUTOFF IN $\eta$. Let $R > 0$ and $\chi \in C_{\mathrm{c}}^\infty(\mathbb{R}, [0, 1])$ with $\chi(\eta) = 1$ for $|\eta| < \frac{1}{2}$ and $\chi(\eta) = 0$ for $|\eta| > 1$. We set

$$c^{\delta,R}(q, p, \eta) := \chi\left(\frac{\eta}{R}\right)\big(b^\delta(q, p, \eta) - b_\infty^\delta(q, p)\big) + b_\infty^\delta(q, p) \in \mathcal{A}, \tag{A.2}$$

where the smooth function $b_\infty$ stems from the defining property (P) of the symbol class $\mathcal{A}$. We note that $c^{\delta,R}$ is compactly supported in $(q, p)$ and constant as a function of $\eta$ for $|\eta| > R$. There exists a positive constant $\mathrm{const}_\delta > 0$ such that $\|c_\varepsilon^{\delta,R}(q, -\mathrm{i}\varepsilon\nabla_q)\|_{\mathcal{L}(L^2)} \le \mathrm{const}_\delta$ for all $\varepsilon > 0$ and $R > 0$. (In the following, $\mathrm{const}_*$ will denote any $*$-dependent positive number, which might diverge as $* \to 0$ or $* \to \infty$) We have

$$\begin{aligned} s_5(b^\delta(q, p, \eta) - c^{\delta,R}(q, p, \eta)) &= s_5\left(\left(1 - \chi\left(\frac{\eta}{R}\right)\right)\big(b^\delta(q, p, \eta) - b_\infty^\delta(q, p)\big)\right) \\ &\xrightarrow{R\to\infty} 0 \end{aligned}$$

and thus

$$\big(b_\varepsilon^\delta\big)^2(q, -\mathrm{i}\varepsilon\nabla_q) = \big(c_\varepsilon^{\delta,R}\big)^2(q, -\mathrm{i}\varepsilon\nabla_q) + \mathrm{const}_\delta\, o_R(1)$$

uniformly in $\varepsilon > 0$ as $R \to \infty$.

*Step* 3. POLYNOMIAL APPROXIMATION. According to definition (A.2), we write

$$c^{\delta,R}(q, p, \eta) = c_1^{\delta,R}(q, p, \eta) + c_2^{\delta,R}(q, p),$$

with $c_1^{\delta,R} \in C_{\mathrm{c}}^\infty(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2))$ and $c_2^{\delta,R} \in C_{\mathrm{c}}^\infty(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$. We choose $r = r(\delta, R) > 0$ such that

$$\mathrm{supp}\,\big(c_1^{\delta,R}\big) \subset B_{\mathbb{R}^5}\left(\frac{r}{2}\right), \quad \mathrm{supp}\,\big(c_2^{\delta,R}\big) \subset B_{\mathbb{R}^4}\left(\frac{r}{2}\right),$$

where $B_{\mathbb{R}^d}(\rho)$ denotes the closed ball in $\mathbb{R}^d$ with radius $\rho$. Let $\gamma > 0$. There exist smooth $\mathcal{L}(\mathbb{C}^2)$-valued functions $p_1^{\delta,R,\gamma}$ on $\mathbb{R}^5$ and $p_2^{\delta,R,\gamma}$ on $\mathbb{R}^4$ which are componentwise polynomial such that

$$\sup_{(q,p,\eta)\in B_{\mathbb{R}^5}(r)} \left|\partial^\alpha\big(c_1^{\delta,R}(q, p, \eta) - p_1^{\delta,R,\gamma}(q, p, \eta)\big)\right| \xrightarrow{\gamma\to 0} 0$$

for all $\alpha \in \mathbb{N}_0^5$ with $|\alpha| \leq 5$ and

$$\sup_{(q,p)\in B_{\mathbb{R}^4}(r)} \left|\partial^\beta\left(c_2^{\delta,R}(q,p) - p_2^{\delta,R,\gamma}(q,p)\right)\right| \xrightarrow{\gamma\to 0} 0$$

for all $\beta \in \mathbb{N}_0^4$ with $|\beta| \leq 5$. Moreover, we employ another polynomial function $g^\gamma \in C^\infty(\mathbb{R}^4, \mathbb{R})$ with

$$\sup_{(q,p)\in B_{\mathbb{R}^4}(r)} \left|\partial^\beta\left(g(q,p) - g^\gamma(q,p)\right)\right| \xrightarrow{\gamma\to 0} 0$$

for all $\beta \in \mathbb{N}_0^4$ with $|\beta| \leq 5$. We set

$$p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}(q,p) := p_1^{\delta,R,\gamma}\left(q, p, \frac{g^\gamma(q,p)}{\sqrt{\varepsilon}}\right) + p_2^{\delta,R,\gamma}(q,p)$$

and introduce a cutoff function $\chi^{\delta,R} \in C_c^\infty(\mathbb{R}^4, [0,1])$ such that $\chi^{\delta,R}(q,p) = 1$ for $|(q,p)| \leq r/2$ and $\chi^{\delta,R}(q,p) = 0$ for $|(q,p)| \geq r$. Then,

$$\sup_{(q,p)\in\mathbb{R}^4} \left|\partial^\beta\left(c^{\delta,R}\left(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p, \frac{g(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)}{\sqrt{\varepsilon}}\right) - \left(\chi^{\delta,R} p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)\right)\right| \xrightarrow{\gamma\to 0} 0$$

uniformly in $\varepsilon > 0$ for all $\beta \in \mathbb{N}_0^4$ with $|\beta| \leq 5$. According to Lemma 4.2, this convergence translates to

$$\left(c_\varepsilon^{\delta,R}\right)^2(q, -\mathrm{i}\varepsilon\nabla_q) = \left(\chi^{\delta,R} p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)^2(q, -\mathrm{i}\varepsilon\nabla_q) + \mathrm{const}_{\delta,R}\, o_\gamma(1)$$

uniformly in $\varepsilon > 0$ as $\gamma \to 0$.

*Step* 4. MOYAL PRODUCT. Since $\chi^{\delta,R} \in S_0^0(1)$ is scalar-valued and cuts off the polynomial growth of $p^{\delta,R,\gamma}$, one has by Lemma 4.1 that

$$\left(\chi^{\delta,R} p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)^2 = \chi^{\delta,R} \sharp_\varepsilon \left(p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)^2 \sharp_\varepsilon \chi^{\delta,R} + \sqrt{\varepsilon} r^{\delta,R,\gamma,\varepsilon}$$

with $r^{\delta,R,\gamma,\varepsilon} \in S_{1/2}^0(1)$. By Lemma A.1, one then obtains

$$\left(\chi^{\delta,R} p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)^2 = \chi^{\delta,R} \sharp_\varepsilon p_{\varepsilon,g^\gamma}^{\delta,R,\gamma} \sharp_\varepsilon p_{\varepsilon,g^\gamma}^{\delta,R,\gamma} \sharp_\varepsilon \chi^{\delta,R} + \sqrt{\varepsilon} s^{\delta,R,\gamma,\varepsilon}$$

with $s^{\delta,R,\gamma,\varepsilon} \in S_{1/2}^0(1)$.

*Step* 5. CONCLUSION. Putting all the previous pieces together while using the real-valuedness and symmetry, respectively, of the symbols $\chi^{\delta,R}$ and $p^{\delta,R,\gamma}$, one obtains

$$\begin{aligned}\langle\psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2} &= \left\|\left(\chi^{\delta,R} \sharp_\varepsilon p_{\varepsilon,g^\gamma}^{\delta,R,\gamma}\right)(q, -\mathrm{i}\varepsilon\nabla_q)\psi\right\|_{L^2}^2 \\ &\quad + \left(\mathcal{O}(\delta) + \mathrm{const}_\delta o_R(1) + \mathrm{const}_{\delta,R} o_\gamma(1) + \mathrm{const}_{\delta,R,\gamma}\mathcal{O}(\sqrt{\varepsilon})\right)\|\psi\|_{L^2}^2\end{aligned}$$

as $\delta, \gamma, \varepsilon \to 0$ and $R \to \infty$. Since the first summand on the right-hand side is nonnegative, we have

$$\langle\psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2} \geq$$

$$- |\mathcal{O}(\delta) + \mathrm{const}_\delta o_R(1) + \mathrm{const}_{\delta,R} o_\gamma(1) + \mathrm{const}_{\delta,R,\gamma}\mathcal{O}(\sqrt{\varepsilon})| \|\psi\|^2_{L^2}$$

as $\delta, \gamma, \varepsilon \to 0$ and $R \to \infty$. For $n \in \mathbb{N}$ we choose $\delta_n, R_n, \gamma_n, \varepsilon_n > 0$ (exactly in this order) such that

$$|\mathcal{O}(\delta_n)|, |\mathrm{const}_{\delta_n} o_{R_n}(1)|, |\mathrm{const}_{\delta_n,R_n} o_{\gamma_n}(1)|, |\mathrm{const}_{\delta_n,R_n,\gamma_n}\mathcal{O}(\sqrt{\varepsilon_n})| \le \frac{1}{4n}$$

and define $c(\varepsilon) := \sum_{n=1}^{\infty} 1\!\!1_{[\varepsilon_{n+1},\varepsilon_n[}(\varepsilon)\frac{1}{n}$ with $\varepsilon_1 = 1$. Then,

$$\langle \psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} \ge -c(\varepsilon)\|\psi\|^2_{L^2(\mathbb{R}^2)}.$$

□

By using Fourier integral operators, the sharp, two-scale Gårding inequality of Remark 4.4 can be proven along the following lines, which have been communicated to us by the anonymous referee.

PROOF OF REMARK 4.4: First, one assumes $g(q, p) = p_1$. In this easy case one has

$$a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q) = a_{\varepsilon,3}(q, -\mathrm{i}\sqrt{\varepsilon}\nabla_q)$$

with $a_{\varepsilon,3}(q, p) := a(q, \sqrt{\varepsilon}p, g(q, p)) = a(q, \sqrt{\varepsilon}p, p_1)$. For symbols $a \in \mathcal{A}$ one has $\|\partial^\alpha a_{\varepsilon,3}\|_\infty \le C_\alpha \sqrt{\varepsilon}^\alpha$. Therefore, the claimed sharp Gårding inequality can be proven in $\sqrt{\varepsilon}$-symbolic calculus.

For arbitrary functions $g$ one uses a canonical transformation $\kappa$, which maps $\{p_1 = 0\} \cap \Omega$ into $\{g(q, p) = 0\}$ with $\Omega \subset \mathbb{R}^4$. Let $U$ denote the Fourier integral operator associated with $\kappa$ (see, e.g., section 2.2 in [10]). Then one has for all $a \in \mathcal{A}$ with $\mathrm{supp}(a_\varepsilon) \subset \Omega$

$$\langle \psi, a_\varepsilon(q, -\mathrm{i}\varepsilon\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} = \langle \psi, (a \circ \kappa^{-1})_{\varepsilon,3}(q, -\mathrm{i}\sqrt{\varepsilon}\nabla_q)\psi\rangle_{L^2(\mathbb{R}^2)} + \mathcal{O}(\sqrt{\varepsilon})$$

for all $\psi \in L^2(\mathbb{R}^2, \mathbb{C}^2)$. For arbitrary symbols $a \in \mathcal{A}$ one must use a partition of unity on the support of $a$ to prove the sharp Gårding inequality. □

## A.3 Two-Scale Wigner Measures

Next, we present the self-contained construction of two-scale Wigner measures (Proposition 4.5). We proceed analogously to the standard construction of Wigner measures, using the Calderon-Vaillancourt theorem and the Gårding-type inequality in Proposition 4.3.

PROOF OF PROPOSITION 4.5: We proceed via different steps, first showing a uniform bound, second positivity of the limit points, then extending the linear form to continuous functions, and finally proving the claimed relation to the Wigner measure $\mu$.

*Step* 1. A UNIFORM BOUND. The upper bound (4.4) resulting from the Calderon-Vaillancourt theorem gives a positive constant $C > 0$ such that

$$\left|\left\langle W_2^\varepsilon(\psi^\varepsilon), a\right\rangle_{\mathcal{A}',\mathcal{A}}\right| \le C s_5(a)\|\psi^\varepsilon\|^2_{L^2(\mathbb{R}^2)}.$$

Since $\mathcal{A}$ is a separable topological vector space, an application of the Banach-Alaoglu theorem, theorem 3.17 in [27], gives a subsequence $(W_2^{\varepsilon_k}(\psi^{\varepsilon_k}))_{\varepsilon_k>0}$ that converges with respect to the weak-star topology to some $\rho \in \mathcal{A}'$.

*Step* 2. POSITIVITY. By Proposition 4.3, we have for nonnegative $0 \le a \in \mathcal{A}$

$$\begin{aligned}\langle \rho, a\rangle_{\mathcal{A}',\mathcal{A}} = \lim_{k\to\infty} \left\langle W_2^{\varepsilon_k}(\psi^{\varepsilon_k}), a\right\rangle_{\mathcal{A}',\mathcal{A}} &= \lim_{k\to\infty} \left\langle \psi^{\varepsilon_k}, a_\varepsilon(q, -\mathrm{i}\varepsilon_k\nabla_q)\psi^{\varepsilon_k}\right\rangle_{L^2(\mathbb{R}^2)} \\ &\ge -\lim_{k\to\infty} c(\varepsilon_k)\|\psi^{\varepsilon_k}\|^2_{L^2(\mathbb{R}^2)} = 0.\end{aligned}$$

Thus, $\rho$ is a bounded positive linear form on $\mathcal{A}$.

*Step* 3. EXTENSION TO $C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$. The following considerations coincide literally with the standard arguments showing that positive distributions are Radon measures. However, since we have to work with matrix-valued measures on $\mathbb{R}^4 \times \overline{\mathbb{R}}$, we follow up the usual argumentation ensuring that the matrix-valuedness and the set $\{\eta = \infty\}$ do not enforce any alterations. For $a \in \mathcal{A}$ with values in $\mathcal{L}_{sa}(\mathbb{C}^2)$ we have $\|a\|_\infty \pm a \ge 0$, where $\|a\|_\infty = \sup_{(q,p,\eta)\in\mathbb{R}^5} \|a(q, p, \eta)\|_{\mathcal{L}(\mathbb{C}^2)}$. Therefore, $\|a\|_\infty\rho(\mathrm{Id}) \pm \rho(a) \ge 0$, that is,

$$|\rho(a)| \le \rho(\mathrm{Id})\|a\|_\infty.$$

For arbitrary $a \in \mathcal{A}$, we choose $\theta \in \mathbb{R}$ such that $\mathrm{e}^{\mathrm{i}\theta}\rho(a) \in \mathbb{R}$. Since $\rho(a^*) = \overline{\rho(a)}$, we have by the preceding observation

$$|\rho(a)| = \frac{1}{2}|\rho(\mathrm{e}^{\mathrm{i}\theta}a + \mathrm{e}^{-\mathrm{i}\theta}a^*)| \le \rho(\mathrm{Id})\frac{1}{2}\|e^{i\theta}a + e^{-i\theta}a^*\|_\infty \le \rho(\mathrm{Id})\|a\|_\infty. \tag{A.3}$$

Clearly, we can identify $C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$ with the space

$$\begin{aligned}\big\{&a \in C(\mathbb{R}^5, \mathcal{L}(\mathbb{C}^2)) : \mathrm{supp}(a) \subset K \times \mathbb{R} \text{ for some compact set } K \subset \mathbb{R}^4, \\ &\exists a_\infty \in C(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2)) : \lim\nolimits_{|\eta|\to\infty} \|a(\cdot, \eta) - a_\infty\|_\infty = 0\big\},\end{aligned}$$

and thus we can view $\mathcal{A}$ as a subspace of $C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$. For $\delta > 0$ and $\phi_\delta \in \mathcal{A}$ with $\int_{\mathbb{R}^5} \phi_\delta(x)\mathrm{d}x = 1$ and $\mathrm{supp}(\phi_\delta) \subset \{x \in \mathbb{R}^5 : |x| \le \delta\}$, one immediately checks that the convolution $a * \phi_\delta$ is a function in $\mathcal{A}$ and that $\mathcal{A}$ is dense in $C_\mathrm{c}(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$ with respect to the supremum norm.

By the bound obtained in (A.3), $\rho$ extends uniquely to a bounded positive linear form on $C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$. By the Riesz representation theorem, $\rho$ is a bounded positive Radon measure on $\mathbb{R}^4 \times \overline{\mathbb{R}}$.

*Step* 4. RELATION TO THE WIGNER MEASURE. Let the sequence

$$\big(W_2^\varepsilon(\psi^\varepsilon)\big)_{\varepsilon>0}$$

converge to $\rho \in \mathcal{A}'$. Since any test function $a \in \mathcal{S}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$ can be viewed as an $\eta$-independent observable in $\mathcal{A}$, we have for such functions $a$

$$\lim_{\varepsilon\to 0} \left\langle W^\varepsilon(\psi^\varepsilon), a\right\rangle_{\mathcal{S}',\mathcal{S}} = \lim_{\varepsilon\to 0} \left\langle W_2^\varepsilon(\psi^\varepsilon), a\right\rangle_{\mathcal{A}',\mathcal{A}}.$$

Thus, $(\langle W^\varepsilon(\psi^\varepsilon), a\rangle_{\mathcal{S}',\mathcal{S}})_{\varepsilon>0}$ converges for all $a \in \mathcal{S}(\mathbb{R}^4, \mathcal{L}(\mathbb{C}^2))$. For $a \in \mathcal{A}_{I_g}$ with

$$\mathcal{A}_{I_g} := \big\{a \in \mathcal{A} \mid \operatorname{supp}(a) \cap (I_g \times \mathbb{R}) = \varnothing, \lim_{|\eta|\to\infty} c_4(a(\cdot,\eta) - a_\infty) = 0\big\},$$

there exists $c = c(a) > 0$ such that $|g(q,p)| \geq c$ for all $(q,p)$ in the support of $a$, and hence $|g(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)/\sqrt{\varepsilon}| \geq c/\sqrt{\varepsilon}$ for all $(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)$ in the support of $a$. We obtain for all $\alpha \in \mathbb{N}_0^4$ with $|\alpha| \leq 5$

$$\lim_{\varepsilon\to 0} \sup_{(q,p)\in\mathbb{R}^4} \left| \partial^\alpha a\left(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p, \frac{g(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)}{\sqrt{\varepsilon}}\right) - \partial^\alpha a_\infty(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p) \right| \leq \lim_{|\eta|\to\infty} c_4(a(\cdot,\eta) - a_\infty) = 0.$$

Denoting $(q,p) \mapsto a_{\infty,\varepsilon}(q,p) := a_\infty(\sqrt{\varepsilon}q, \sqrt{\varepsilon}p)$, one has $\lim_{\varepsilon\to 0} c_4(a_{\varepsilon,2} - a_{\infty,\varepsilon}) = 0$ and therefore by the Calderon-Vaillancourt theorem

$$\begin{aligned}
\langle \rho, a\rangle_{\mathcal{A}',\mathcal{A}} &= \lim_{\varepsilon\to 0} \big\langle S^\varepsilon\psi^\varepsilon, a_{\varepsilon,2}(q, -\mathrm{i}\nabla_q) S^\varepsilon\psi^\varepsilon\big\rangle_{L^2} \\
&= \lim_{\varepsilon\to 0} \big\langle S^\varepsilon\psi^\varepsilon, a_{\infty,\varepsilon}(q, -\mathrm{i}\nabla_q) S^\varepsilon\psi^\varepsilon\big\rangle_{L^2} \\
&= \lim_{\varepsilon\to 0} \big\langle \psi^\varepsilon, a_\infty(q, -\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon\big\rangle_{L^2} = \int_{\mathbb{R}^4} \operatorname{tr}\big(a_\infty(q,p)\mu(\mathrm{d}q, \mathrm{d}p)\big).
\end{aligned}$$

By the same arguments as employed before, we can approximate functions $a \in C_c(\mathbb{R}^4 \times \overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$ with support away from $I_g$ by observables in $(a * \phi_\delta)_{\delta>0}$ in $\mathcal{A}_{I_g}$ with support away from $I_g$, since $|g(q,p)| \geq c$ for $(q,p)$ in the support of $a$ implies $|g(q',p')| \geq c'$ for some $c' = c'(\delta) > 0$ for all $(q',p')$ in the support of $a * \phi_\delta$, and since for all $\alpha \in \mathbb{N}_0^4$ with $|\alpha| \leq 5$

$$\lim_{|\eta|\to\infty} \big\|\partial^\alpha((a*\phi_\delta)(\cdot,\eta) - a_\infty * \phi_{\delta,\infty})\big\|_\infty \leq \lim_{|\eta|\to\infty} \big\|\partial^\alpha(a(\cdot,\eta) - a_\infty)\big\|_\infty \|\phi_\delta\|_{L^1(\mathbb{R}^5)} = 0.$$

Thus,

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}\big(a(q,p,\eta)\rho(\mathrm{d}q, \mathrm{d}p, \mathrm{d}\eta)\big) = \int_{\mathbb{R}^4} \operatorname{tr}\big(a(q,p,\infty)\mu(\mathrm{d}q,\mathrm{d}p)\big)$$

and

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} a(q,p,\eta)\rho(\mathrm{d}q, \mathrm{d}p, \mathrm{d}\eta) = \int_{\mathbb{R}^4} a(q,p,\infty)\mu(\mathrm{d}q,\mathrm{d}p),$$

which means

$$\rho\big|_{(\mathbb{R}^4\setminus I_g)\times\overline{\mathbb{R}}}(q,p,\eta) = \mu\big|_{\mathbb{R}^4\setminus I_g}(q,p) \otimes \delta(\eta - \infty), \quad (q,p,\eta) \in \mathbb{R}^4 \times \overline{\mathbb{R}}.$$

Defining $\nu := \rho|_{I_g\times\overline{\mathbb{R}}}$ as the restriction of the measure $\rho$ to $I_g \times \overline{\mathbb{R}}$, we obtain

$$\rho(q,p,\eta) = \mu\big|_{\mathbb{R}^4\setminus I_g}(q,p) \otimes \delta(\eta - \infty) + \nu(q,p,\eta).$$

For $a(q,p) = a \in \mathcal{A}$ just depending on $(q,p)$ we have

$$\int_{\mathbb{R}^4\times\overline{\mathbb{R}}} \operatorname{tr}(a(q,p)\rho(\mathrm{d}q,\mathrm{d}p,\mathrm{d}\eta)) = \lim_{\varepsilon\to 0}\left\langle \psi^\varepsilon, a(q,-\mathrm{i}\varepsilon\nabla_q)\psi^\varepsilon\right\rangle_{L^2}$$

$$= \int_{\mathbb{R}^4} \operatorname{tr}\big(a(q,p)\mu(\mathrm{d}q,\mathrm{d}p)\big),$$

and thus $\int_{\overline{\mathbb{R}}} \nu(\,\cdot\,,\mathrm{d}\eta) = \mu\big|_{I_g}$.

□

## A.4 Localization of Two-Scale Wigner Measures

Finally, we provide the proof of the localization property of two-scale Wigner measures in the cotangent space of space-time, Lemma 4.11.

PROOF OF LEMMA 4.11: We define a linear operator

$$\widetilde{H}^\varepsilon := -\mathrm{i}\varepsilon\partial_t - H^\varepsilon = (\tau + h)((t,q), -\mathrm{i}\varepsilon\nabla_{t,q})$$

with domain

$$D(\widetilde{H}^\varepsilon) := \{\psi \in L^2(\mathbb{R}^3,\mathbb{C}^2) \mid \psi(\,\cdot\,,q) \in C^1(\mathbb{R},\mathbb{C}^2), q \in \mathbb{R}^2;$$
$$\psi(t,\,\cdot\,) \in D(H^\varepsilon), t \in \mathbb{R}\}.$$

For initial data $\psi_0^\varepsilon \in D(H^\varepsilon)$ the solution $\psi^\varepsilon$ is in $C^1(\mathbb{R}, D(H^\varepsilon))$. Thus, $\chi_\mathrm{T}\psi^\varepsilon \in D(\widetilde{H}^\varepsilon)$ and

$$\left\|\widetilde{H}^\varepsilon\big(\chi_\mathrm{T}\psi^\varepsilon\big)\right\|_{L^2(\mathbb{R}^3)} = \left\|(-\mathrm{i}\varepsilon\partial_t\chi_\mathrm{T})\psi^\varepsilon\right\|_{L^2(\mathbb{R}^3)} \overset{\varepsilon\to 0}{\longrightarrow} 0.$$

The symbol $a_\varepsilon$ need not have any decay properties for large $\tau$. However, since $\tau + h$ is linear in $\tau$, the reasoning of Lemma 4.1's proof gives for $a \in \mathcal{A}_\mathrm{T}$

$$a_\varepsilon \,\sharp_\varepsilon\, (\tau + h) - a_\varepsilon(\tau + h) \in S^{-1/2}_{1/2}(1).$$

For a well-defined pairing with $\rho_\mathrm{T}$, we restrict ourselves to symbols $a \in \mathcal{A}_\mathrm{T}$ with support $\operatorname{supp}(a) \subset [0,T] \times \mathbb{R}^2_q \times [\tau_1,\tau_2] \times \mathbb{R}^3_{p,\eta}$ for some $\tau_1, \tau_2 \in \mathbb{R}$ and have

$$\langle \rho_\mathrm{T}, a(\tau + h)\rangle_{\mathcal{A}'_\mathrm{T},\mathcal{A}_\mathrm{T}} = \lim_{k\to\infty}\left\langle \chi_\mathrm{T}\psi^{\varepsilon_k}, (a_\varepsilon(\tau + h))((t,q), -\mathrm{i}\varepsilon_k\nabla_{t,q})\big(\chi_\mathrm{T}\psi^{\varepsilon_k}\big)\right\rangle_{L^2(\mathbb{R}^3)}$$
$$= \lim_{k\to\infty}\left\langle \chi_\mathrm{T}\psi^{\varepsilon_k}, a_\varepsilon((t,q), -\mathrm{i}\varepsilon_k\nabla_{t,q})\widetilde{H}^{\varepsilon_k}\big(\chi_\mathrm{T}\psi^{\varepsilon_k}\big)\right\rangle_{L^2(\mathbb{R}^3)} = 0.$$

Since $\rho_\mathrm{T}$ is a distribution of order 0, and since the set of symbols used in the preceding lines is dense in $C_c(\mathbb{R}^6\times\overline{\mathbb{R}}, \mathcal{L}(\mathbb{C}^2))$, we have $\rho_\mathrm{T}(\tau + h) = 0$ as measures, provided initial data $\psi_0^\varepsilon \in D(H^\varepsilon)$. A $\|\cdot\|_{L^2(\mathbb{R}^2)}$ density argument proves

$$\langle \rho_\mathrm{T}, a(\tau + h)\rangle_{\mathcal{A}'_\mathrm{T},\mathcal{A}_\mathrm{T}} = 0$$

for general initial data $\psi_0^\varepsilon \in L^2(\mathbb{R}^2,\mathbb{C}^2)$ and observables $a \in \mathcal{A}_\mathrm{T}$ with compact $\tau$ support, while another $\|\cdot\|_\infty$ density argument gives $\rho_\mathrm{T}(\tau + h) = 0$ in the sense of measures. Observing that $V(q)^2 = |q|^2\,\mathrm{Id}$, we finally obtain the claimed assertion on the support of $\rho_{\mathrm{T}}$. $\square$

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