PROPAGATION THROUGH GENERIC LEVEL CROSSINGS: A SURFACE HOPPING SEMIGROUP

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Abstract. We construct a surface hopping semigroup, which asymptotically describes nuclear propagation through crossings of electron energy levels. The underlying time-dependent Schrödinger equation has a matrix-valued potential, whose eigenvalue surfaces have a generic intersection of codimension two, three or five in Hagedorn's classification. Using microlocal normal forms reminiscent of the Landau-Zener problem, we prove convergence to the true solution with an error of the order $\varepsilon^{1/8}$, where ε is the semi-classical parameter. We present numerical experiments for an algorithmic realization of the semigroup illustrating the convergence of the algorithm.

Key words. Time-dependent Schrödinger system, eigenvalue crossing, microlocal normal form, surface hopping.

1. Introduction. In the framework of time-dependent Born-Oppenheimer approximation, the dynamics of molecules can approximately be reduced to matrix-valued Schrödinger equations on the nucleonic configuration space,

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

see for example [11, 18]. The linear Schrödinger equation (1.1) has a unique global solution $\psi^\varepsilon \in C(\mathbb{R}, L^2(\mathbb{R}^d,\mathbb{C}^N))$ for all square-integrable initial data ψ_0^ε. The parameter $\varepsilon > 0$ is small and causes a highly oscillatory behavior of the solution in space and time. It can be thought of as the square root of the ratio of electronic mass and the average mass of the nuclei. Moreover, the solution itself does not have any direct physical interpretation. It is the position density $|\psi^\varepsilon(q,t)|^2$, which gives the probability of finding the nuclei in the configuration $q\in\mathbb{R}^d$ at time t. We are interested in an asymptotic description for the time evolution of quadratic quantities like the position density with the following properties. First, it shall be effective in the sense, that it unfolds characteristic dynamical properties. Second, it shall be explicit enough, such that it allows an algorithmic realization. Third, the resulting algorithm shall be applicable on high dimensional nucleonic configuration spaces $\mathbb{R}^d$, $d \gg 1$.

G. Hagedorn rigorously derived and classified Schrödinger systems for molecular propagation through electron energy level crossings of minimal multiplicity [12]. He obtained potentials of the form

$$V(q) = v(q)\,\mathrm{Id} + V_\ell\left(\phi(q)\right), \quad \ell\in\{2,3,3',5\},$$

where $v(q)\in\mathcal{C}^\infty(\mathbb{R}^d,\mathbb{R})$ is a smooth real-valued function, Id is the identity matrix in $\mathbb{C}^{2\times2}$ or $\mathbb{C}^{4\times4}$, and $q\mapsto\phi(q)$ is a smooth vector-valued function with $\phi(q)\in\mathbb{R}^2$, $\mathbb{R}^3$ or $\mathbb{R}^5$. The matrices V_ℓ are given by

$$V_2(\phi) = \begin{pmatrix}\phi_1 & \phi_2\\ \phi_2 & -\phi_1\end{pmatrix}, \qquad V_3(\phi) = \begin{pmatrix}\phi_1 & \phi_2+i\phi_3\\ \phi_2-i\phi_3 & -\phi_1\end{pmatrix}, \tag{1.2}$$

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$$(1.3)\qquad V_{3'}(\phi)=\begin{pmatrix}\begin{pmatrix}\phi_1 & \phi_2+i\phi_3\\ \phi_2-i\phi_3 & -\phi_1\end{pmatrix} & \mathbf{0}\\ \mathbf{0} & \begin{pmatrix}\phi_1 & \phi_2-i\phi_3\\ \phi_2+i\phi_3 & -\phi_1\end{pmatrix}\end{pmatrix},$$

$$(1.4)\qquad V_5(\phi)=\begin{pmatrix}\phi_0\ \mathrm{Id} & \begin{pmatrix}\phi_1+i\phi_2 & \phi_3+i\phi_4\\ -\phi_3+i\phi_4 & \phi_1-i\phi_2\end{pmatrix}\\ \begin{pmatrix}\phi_1-i\phi_2 & -\phi_3-i\phi_4\\ \phi_3-i\phi_4 & \phi_1+i\phi_2\end{pmatrix} & -\phi_0\ \mathrm{Id}\end{pmatrix}.$$

For these four matrices, the eigenvalues are $\pm|\phi|$. Therefore, the eigenvalues of $V(q)$ are $v(q)\pm|\phi(q)|$, and $q^*\in\mathbb{R}^d$ is a point of crossing eigenvalues if and only if $\phi(q^*)=0$. We shall say that this crossing is generic, if

$$\mathrm{d}\phi \text{ is of maximal rank on } \{\phi=0\},$$

i. e. of rank 2 for $\ell=2$, of rank 3 for $\ell=3,3'$ and of rank 5 for $\ell=5$. This explains why these crossings are usually referred to as codimension two, three and five crossings, and enlightens the choice of the index ℓ we have made. G. Hagedorn's codimension one crossings are not considered here, since they violate the above rank condition and also show a different dynamical behavior than systems with crossings of higher codimension. We set

$$N(2)=N(3)=2,\quad N(3')=N(5)=4,$$

so that the potential $V(q)$, the wave function $\psi^\varepsilon(q,t)$, and the differential $\mathrm{d}\phi(q)$ belong to $\mathbb{C}^{N(\ell)\times N(\ell)}$, $\mathbb{C}^{N(\ell)}$, and $\mathbb{R}^{\ell\times d}$, respectively. For $\ell=3'$ we set $\mathbb{R}^{3'}=\mathbb{R}^3$. The orthogonal eigenprojectors

$$\Pi^\pm(q)=\tfrac{1}{2}\left(\mathrm{Id}\pm|\phi(q)|^{-1}V_\ell(\phi(q))\right)$$

of the matrix $V(q)$ have a conical singularity at points of crossing eigenvalues q^*, that is $\nabla\Pi^\pm(q)=O(|q-q^*|^{-1})$ as $q\to q^*$. This motivates the notion of conical intersections, by which especially codimension two crossings are frequently referred to.

Eigenvalue crossings are ubiquitous in the quantum mechanical description of polyatomic molecules, that is molecules with more than two nuclei. The collection [4] provides an exposition of this active area of research in theoretical chemistry. As for a prominent example of an ultrafast isomerization on the femtosecond time scale, a codimension two crossing of energy levels explains the effectiveness of the first step of vision, the cis-trans isomerization of retinal in rhodopsin, see also [13] and section 3 below for related numerical experiments.

The analysis of scalar Schrödinger equations teaches that the direct study of the time-evolution of quadratic quantities like the position density $|\psi^\varepsilon(q,t)|^2$ is impossible. The oscillations of $\psi^\varepsilon(q,t)$ have to be taken into account, and one has to work in the space of positions and momenta (q,p), the phase space $\mathbb{R}^d_q\times\mathbb{R}^d_p$. Therefore, one studies the Wigner function of $\psi^\varepsilon(q,t)$ in a suitable ε-dependent scaling, which resolves the highly oscillatory features of the solution,

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

It plays the role of a generalized probability density on phase space. For square-integrable wave functions ψ, the Wigner function $W^\varepsilon(\psi)$ is a square-integrable function on phase space with values in the space of hermitian matrices. One recovers the position density by

$$|\psi(q)|^2 = \mathrm{tr} \int_{\mathbb{R}^d} W^\varepsilon(\psi)(q,p)\,\mathrm{d}p.$$

Besides, the action of the Wigner function against compactly supported smooth test functions $a \in \mathcal{C}_c^\infty(\mathbb{R}^{2d}, \mathbb{C}^{N(\ell)\times N(\ell)})$ is simply expressed in terms of the semi-classical pseudodifferential operator with symbol a, which is defined by

$$\mathrm{op}_\varepsilon(a)\psi(q) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} a\left(\tfrac{1}{2}(q+v), p\right) \mathrm{e}^{\frac{i}{\varepsilon} p\cdot(q-v)} \psi(v)\,\mathrm{d}v\,\mathrm{d}p$$

for $\psi \in L^2(\mathbb{R}^d, \mathbb{C}^{N(\ell)})$. Indeed, we have

$$\mathrm{tr} \int_{\mathbb{R}^{2d}} W^\varepsilon(\psi)(q,p) a(q,p)\,\mathrm{d}q\,\mathrm{d}p = (\mathrm{op}_\varepsilon(a)\psi\,,\,\psi)_{L^2(\mathbb{R}^d,\mathbb{C}^{N(\ell)})}\,.$$

It is our aim to construct an asymptotic semigroup, which approximately propagates the initial data's Wigner function for all generic level crossings of Hagedorn's classification. Our approximation relies on a microlocal normal form for operators with eigenvalue crossings, which has been derived in [1, 2, 6]. Roughly speaking, near a crossing point the Schrödinger operator

$$-i\varepsilon\partial_t - \tfrac{\varepsilon^2}{2}\Delta_q + V(q)$$

is equivalent to the normal form

$$-i\varepsilon\partial_t + V_\ell\Big(t, \mathrm{op}_\varepsilon(|\mathrm{d}\phi(q)p|^{-\frac{1}{2}}\pi_\ell(q,p)\phi(q))\Big)\,,$$

where $\pi_\ell(q,p)$ denotes the orthogonal projection onto the hyperplane normal to the vector $\mathrm{d}\phi(q)p \in \mathbb{R}^\ell$. In the case $\ell = 2$, this resembles the Landau-Zener system

$$i\varepsilon\tfrac{\mathrm{d}}{\mathrm{d}t}\psi(t) = \begin{pmatrix} t & \gamma \\ \gamma & -t \end{pmatrix}\psi(t), \qquad \gamma > 0,$$

for which the probability, that a solution starting at time $t = -\infty$ in the one eigenspace will have passed over to the other eigenspace at time $t = +\infty$, has explicitly been computed by Landau [15] and Zener [20] in the 1930s. This famous Landau-Zener transition rate reads as

$$\exp\Big(-\frac{\pi}{\varepsilon}\gamma^2\Big)\,,$$

and in [8] it is proven, that the rate still gives the correct asymptotics, if γ is replaced by a bounded operator. Our semigroup combines effective transitions between eigenspaces close to points of crossing eigenvalues on the one hand with classical transport in the adiabatic regime on the other hand. More precisely, the V-diagonal components $\Pi^\pm W^\varepsilon(\psi_0^\varepsilon)\Pi^\pm$ of the initial Wigner function are transported along the Hamiltonian curves of the eigenvalues of the Schrödinger operator's symbol

$$\tfrac{1}{2}|p|^2 + v(q) \pm |\phi(q)|.$$

Whenever one of the trajectories $(q^{\pm}(t), p^{\pm}(t))$ attains a local minimal gap between the eigenvalues, there is an effective non-adiabatic transfer of weight according to the ε-dependent transition rate

$$\exp\left(-\frac{\pi}{\varepsilon}\frac{|\pi_\ell(q,p)\phi(q)|^2}{|\mathrm{d}\phi(q)p|}\right).$$

Since the rate is negligibly small, when the eigenvalue gap is larger than $\sqrt{\varepsilon}$, the non-adiabatic transfer of weight is effectively performed at times t^* with

$$t \mapsto |\phi(q^{\pm}(t))| \text{ has a local minimum in } t = t^* \text{ and } |\phi(q^{\pm}(t^*))| \leq R\sqrt{\varepsilon}$$

for some fixed $R > 0$. Our main result is, that this dynamical description yields approximate solutions with an error of order $\varepsilon^{1/8}$ when choosing $R = \varepsilon^{-1/8}$. Moreover, it is explicit enough for an algorithmic realization, whose performance on a model for retinal in rhodopsin is studied here as well. The algorithm is a mathematical counterpart to the popular surface hopping algorithms of chemical physics introduced by Tully and Preston in [19].

Quantum dynamical descriptions in terms of classical transport as described above, that is in the spirit of an Egorov theorem, are well established and have been given for Wigner functions for example in [10]: for Schrödinger systems they hold to leading order in ε, until classical trajectories come close to a point of crossing eigenvalues. Then, as already mentioned, the adiabatic approximation is no longer valid, and there are leading order non-adiabatic transitions between the levels (the energy propagated on one level to the crossing may pass partially or completely on the other level). This phenomenon has been precisely analyzed in the case of Gaussian wave packet propagation by G. Hagedorn [12] for all generic electron level crossings. For initial data, which are less specific than Gaussian wave packets, the evolution of appropriate two-scale Wigner measures has been studied. These measures are weak limits of the Wigner function and incorporate information on concentration effects close to trajectories, which touch points of crossing eigenvalues, with respect to the second scale $\sqrt{\varepsilon}$. These Wigner measures have been analyzed for a linear codimension two crossing in [7], for general two-level systems in [8], and for all of Hagedorn's models in [5]. In [17], the results of [7] have been lifted to a leading order approximation of the Wigner function. Here, we aim at approximating the Wigner function for all generic crossings, while additionally proving a convergence rate.

We will proceed as follows. Section 2 constructs the surface hopping semigroup, states the main result, that is the validity of our approximation with an error of order $\varepsilon^{1/8}$, and discusses the strategy of the proof. In section 3, numerical results are presented for an algorithmic realization of the semigroup applied to a retinal model. In section 4, the proof for propagation away from the crossing is carried out, while the microlocal normal form yields the correct non-adiabatic transition rates, as proven in section 5. In section 6, the main result is extended to observables, which are more pertinent for the crossings with degenerate eigenvalues ($\ell = 3', 5$). Finally, the appendix summarizes basic facts of Weyl calculus.

2. Main Result. Propagation through level crossings can be approximated by a proper combination of classical transport and non-adiabatic transitions. For this, we study the underlying classical flows and combine them with effective non-adiabatic transitions to an asymptotic semigroup.

2.1. Transport and transitions. We consider the classical flows

$$\Phi_\pm^{-t} : \mathbb{R}^{2d} \to \mathbb{R}^{2d}, \quad \Phi_\pm^{-t}(q_0,p_0) = \big(q^\pm(t), p^\pm(t)\big)$$

associated to the Hamiltonian curves of $\frac{1}{2}|p|^2 + v(q) \pm |\phi(q)|$. These curves are solutions to the Hamiltonian systems

$$\begin{cases} \dot q^\pm(t) = p^\pm(t), \;\; \dot p^\pm(t) = -\nabla v\left(q^\pm(t)\right) \mp {}^t\mathrm{d}\phi\left(q^\pm(t)\right) \frac{\phi\left(q^\pm(t)\right)}{|\phi(q^\pm(t))|}, \\ q^\pm(0) = q_0, \;\; p^\pm(0) = p_0. \end{cases}$$

We only consider initial phase space points $(q_0,p_0) \in \mathbb{R}^{2d}$ such that for $t > 0$

$$\phi(q^\pm(t)) = 0 \;\Rightarrow\; \mathrm{d}\phi(q^\pm(t))p^\pm(t) \neq 0. \tag{2.1}$$

This condition guarantees, that classical trajectories arrive transversally at the crossing set and have a unique smooth continuation through this singularity, see Proposition 1 in [5].

For a large class of test functions and under suitable restrictions on the time interval, the classical flows are enough for approximating the dynamics up to an error of order ε. Indeed, one considers observables $a \in \mathcal{C}_c^\infty(\mathbb{R}^{2d}, \mathbb{C}^{N(\ell)\times N(\ell)})$ such that

$$a = a^+\Pi^+ + a^-\Pi^-, \qquad a^\pm \in \mathcal{C}_c^\infty(\mathbb{R}^{2d} \setminus \{\phi = 0\}, \mathbb{C}). \tag{2.2}$$

For $\ell = 2, 3$, the eigenspaces are one-dimensional, and these observables focus on the V-diagonal elements of the Wigner matrix, where V-diagonal means diagonal with respect to the decomposition of $\mathbb{C}^{N(\ell)}$ by the eigenprojectors $\Pi^+(q)$ and $\Pi^-(q)$. For $\ell = 3', 5$, however, the eigenspaces are two-dimensional, and observables of the form (2.2) are not enough to completely resolve all dynamical features within the eigenspaces. We will address this issue in section 6.

For all times $t \in [0,T]$, such that the classical trajectories $\Phi_\pm^t$ arriving on the support of a have not passed the crossing set $\{\phi = 0\}$, the action of the Wigner function on $a = a^\pm\Pi^\pm$ obeys

$$\mathrm{tr}\int_{\mathbb{R}^{2d}} W^\varepsilon(\psi^\varepsilon(t))\,(q,p)\,\Pi^\pm(q) a^\pm(q,p)\,\mathrm{d}q\,\mathrm{d}p \\ - \mathrm{tr}\int_{\mathbb{R}^{2d}} \big(\Pi^\pm W^\varepsilon(\psi_0^\varepsilon)\Pi^\pm \circ \Phi_\pm^{-t}\big)(q,p)\, a^\pm(q,p)\,\mathrm{d}q\,\mathrm{d}p = O(\varepsilon)$$

as $\varepsilon \to 0$. Such Egorov type descriptions hold, until classical trajectories come close to a crossing point $(q,p) \in \{\phi = 0\}$ and leading order non-adiabatic transitions occur. These transitions depend on how the solution $\psi^\varepsilon(t)$ concentrates on the ingoing trajectories with respect to the scale $\sqrt{\varepsilon}$. For the linear codimension two crossing with $\phi(q) = q$, $q \in \mathbb{R}^2$, the two-scale Wigner measure's description of [7] is lifted to an approximation of the Wigner function in [17]. This linear model has specific features, see also [9]. In particular, all classical trajectories which meet the crossing are included in the set $\{q \wedge p = 0\}$, where $q \wedge p := q_2p_1 - q_1p_2$ for $q, p \in \mathbb{R}^2$. The idea of [17] is to propagate the V-diagonal parts of the initial Wigner function along the classical trajectories and to apply the ε-dependent transition coefficient

$$T_{lin}^\varepsilon(q^*,p^*) = \exp\left(-\frac{\pi}{\varepsilon}\frac{|q^*\wedge p^*|^2}{|p^*|^3}\right),$$

as soon as the trajectories reach their minimal distance from the crossing set, which is easy to check since $q \cdot p = 0$ at such a point. Theorem 3.2 in [17] proves, that under suitable conditions on the initial data this ε-dependent propagation is correct in the limit $\varepsilon \to 0$. We construct here an extension, which covers the general situation described above, and give a convergence proof including a convergence rate. Our approach draws from the understanding of the non-adiabatic mechanism as developed in [5].

2.2. A surface hopping semigroup. Let $R > 0$. In the general case, the crucial points in phase space are those, where the classical trajectories attain a *local minimal gap between the two eigenvalues.* These points fulfill the condition

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\left|\phi\left(q^{\pm}(t)\right)\right|^2\right) = 2\,\mathrm{d}\phi\left(q^{\pm}(t)\right)p^{\pm}(t)\,\cdot\,\phi\left(q^{\pm}(t)\right) = 0,$$

and one performs an effective non-adiabatic transfer of weight, whenever a trajectory passes the set

$$S_{\varepsilon,R} = \left\{(q,p) \in \mathbb{R}^{2d} \mid |\phi(q)| \le R\sqrt{\varepsilon},\ \mathrm{d}\phi(q)p \cdot \phi(q) = 0\right\}.$$

The microlocal normal form, which will be given later in Theorem 5.2, suggests the transition rate

$$T^{\varepsilon}(q^*,p^*) = \exp\left(-\frac{\pi}{\varepsilon}\frac{\left|\pi_\ell(q^*,p^*)\phi(q^*)\right|^2}{\left|\mathrm{d}\phi(q^*)p^*\right|}\right),$$

where $\pi_\ell(q^*,p^*)$ is the orthogonal projection from the Euclidian space $\mathbb{R}^\ell$ into the hyperplane normal to the vector $\mathrm{d}\phi(q^*)p^* \in \mathbb{R}^\ell$. Since $\mathrm{d}\phi(q^{\pm}(t))p^{\pm}(t)$ does not vanish when the considered trajectories arrive at the crossing set $\{\phi = 0\}$, it is also non-zero when arriving at the jump manifold $S_{\varepsilon,R}$, if $R\sqrt{\varepsilon}$ is small enough. Besides, for $\ell = 2$, one has

$$\mid \pi_\ell(q,p)\phi(q) \mid = \mid \phi(q) \wedge \tfrac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|} \mid,$$

and we recover the transition coefficient $T^{\varepsilon}_{lin}(q^*,p^*)$ for $\phi(q) = q$, $q \in \mathbb{R}^2$.

We attach the labels -1 and $+1$ to phase space. For points $(q,p,j) \in \mathbb{R}^{2d}_{\pm} := \mathbb{R}^{2d} \times \{-1,+1\}$, we consider trajectories

$$\mathcal{T}^{(q,p,j)}_{\varepsilon,R} : [0,+\infty) \to \mathbb{R}^{2d}_{\pm},$$

which combine deterministic classical transport and random jumps between the levels at the manifold $S_{\varepsilon,R}$. More precisely, we set $\mathcal{T}^{(q,p,j)}_{\varepsilon,R}(t) = \left(\Phi^t_j(q,p), j\right)$ as long as $\Phi^t_j(q,p) \notin S_{\varepsilon,R}$. Whenever the deterministic flow $\Phi^t_j(q,p)$ hits the manifold $S_{\varepsilon,R}$ at a point (q^*,p^*), a random jump from j to $-j$ occurs with probability $T^{\varepsilon}(q^*,p^*)$. For points (q,p,j) generating classical trajectories, which either violate the non-degeneracy condition (2.1) or do not leave the set $S_{\varepsilon,R}$, there is either no transport or no jump at all. Since the trajectories which touch the crossing set arrive there transversally, each path

$$(q,p,j) \to \mathcal{T}^{(q,p,j)}_{\varepsilon,R}(t)$$

has a finite number of jumps and remains in a bounded region of $\mathbb{R}^{2d}_{\pm}$ within a bounded time-interval $[0,T]$. Away from the jump manifold $S_{\varepsilon,R} \times \{-1,+1\}$ each path is smooth. Hence, the random trajectories define a time-dependent Markov process with state space $\mathbb{R}^{2d}_{\pm}$. The associated transition function $P_{\varepsilon,R}(p,q,j;t,\Gamma)$ describes the probability of being at time t in the measurable set $\Gamma\subset\mathbb{R}^{2d}_{\pm}$ having started in (q,p,j). Its action on bounded measurable scalar functions $f:\mathbb{R}^{2d}_{\pm}\to\mathbb{C}$ defines a semigroup $(\mathcal{L}^t_{\varepsilon,R})_{t\geq 0}$ by

$$(\mathcal{L}^t_{\varepsilon,R}\,f)(q,p,j):=\int_{\mathbb{R}^{2d}\times\{-1,+1\}} f(x,\xi,k)\,P_{\varepsilon,R}(q,p,j;t,\mathrm{d}(x,\xi,k)).$$

For introducing the semigroup's action on Wigner functions, we use the following space of continuous V-diagonal test functions satisfying T^ε-dependent boundary conditions at the jump manifold.

Definition 2.1. *A continuous function $a\in\mathcal{C}_c(\mathbb{R}^{2d}\setminus S_{\varepsilon,R},\mathbb{C}^{N(\ell)\times N(\ell)})$ belongs to the space $\mathcal{C}_{\varepsilon,R}$, if it has the following properties:*

i. $a=a^+\Pi^++a^-\Pi^-$ *with* $a^\pm\in\mathcal{C}_c(\mathbb{R}^{2d}\setminus S_{\varepsilon,R},\mathbb{C})$

ii. The function $f_a:(\mathbb{R}^{2d}\setminus S_{\varepsilon,R})\times\{-1,+1\}\to\mathbb{C}$,

$$f_a(q,p,+)=a^+(q,p),\quad f_a(q,p,-)=a^-(q,p),$$

satisfies for all $(q,p,j)\in S_{\varepsilon,R}\times\{-1,+1\}$

$$\begin{aligned}&\lim_{\delta\to -0} f_a(q+\delta p,p+\delta(-\nabla v(q)-j\ {}^t\mathrm{d}\phi(q)\phi(q)/|\phi(q)|),j)\\ &=\lim_{\delta\to +0}(T^\varepsilon f_a)(q+\delta p,p+\delta(-\nabla v(q)+j\ {}^t\mathrm{d}\phi(q)\phi(q)/|\phi(q)|),-j)\\ &=\lim_{\delta\to +0}((1-T^\varepsilon)f_a)(q+\delta p,p+\delta(-\nabla v(q)-j\ {}^t\mathrm{d}\phi(q)\phi(q)/|\phi(q)|),j).\end{aligned}$$

For test functions $a\in\mathcal{C}_{\varepsilon,R}$, the action of $(\mathcal{L}^t_{\varepsilon,R})_{t\geq 0}$ is naturally given by

$$(\mathcal{L}^t_{\varepsilon,R}\,a)(q,p):=\left(\mathcal{L}^t_{\varepsilon,R}f_a\right)(q,p,+1)\,\Pi^+(q)+\left(\mathcal{L}^t_{\varepsilon,R}f_a\right)(q,p,-1)\,\Pi^-(q).$$

By construction, the semigroup leaves $\mathcal{C}_{\varepsilon,R}$ invariant, and duality allows us to define its action on Wigner functions. More precisely, let $W^\varepsilon(\psi)$ be the Wigner function of some wave function $\psi\in L^2(\mathbb{R}^d,\mathbb{C}^{N(\ell)})$. Then, $\mathcal{L}^t_{\varepsilon,R}W^\varepsilon(\psi)$ acts on $a\in\mathcal{C}_{\varepsilon,R}$ by

$$\left(\mathcal{L}^t_{\varepsilon,R}W^\varepsilon(\psi),a\right)=\mathrm{tr}\int_{\mathbb{R}^{2d}} W^\varepsilon(\psi)(q,p)(\mathcal{L}^t_{\varepsilon,R}\,a)(q,p)\,\mathrm{d}q\,\mathrm{d}p,$$

defining a locally integrable function on phase space. We finally choose an ε-dependent hopping range $R(\varepsilon)=\varepsilon^{-1/8}$ and set

$$(\mathcal{L}^t_\varepsilon)_{t\geq 0}:=(\mathcal{L}^t_{\varepsilon,R(\varepsilon)})_{t\geq 0}.$$

2.3. Assumptions and main result. Let $\psi^\varepsilon(t)$ be the solution of the Schrödinger equation (1.1) with initial datum ψ^ε_0. We now state the precise assumptions, under which the action of the semigroup $(\mathcal{L}^t_\varepsilon)_{t\geq 0}$ on the initial Wigner function $W^\varepsilon(\psi^\varepsilon_0)$ approximates the V-diagonal components of $W^\varepsilon(\psi^\varepsilon(t))$.

(A0) $V\in\mathcal{C}^\infty(\mathbb{R}^d,\mathbb{C}^{N(\ell)\times N(\ell)})$ is of subquadratic growth and of the form

$$V(q)=v(q)\,\mathrm{Id}+V_\ell\left(\phi(q)\right),\quad \ell\in\{2,3,3',5\},$$

where the matrices $V_\ell(\phi(q))$ have been defined in (1.2), (1.3) and (1.4). We assume the eigenvalue crossings to be generic in the sense, that $\mathrm{d}\phi$ is of maximal rank on the crossing set $\{\phi=0\}$.

(A1) $(\psi_0^\varepsilon)_{\varepsilon>0}$ is a bounded family in $L^2\big(\mathbb{R}^d,\mathbb{C}^{N(\ell)}\big)$ associated with $\mathrm{Ran}\Pi^+$,

$$\|\Pi^-\psi_0^\varepsilon\|_{L^2(\mathbb{R}^d,\mathbb{C}^{N(\ell)})} = O(\varepsilon^{\beta_1}),\ \ \beta_1\geq 1/8.$$

We suppose that the initial data are localized away from the crossing $\{\phi=0\}$, i. e. for all $b\in\mathcal{C}_c^\infty(\mathbb{R}^{2d},\mathbb{C}^{N(\ell)\times N(\ell)})$ with $\mathrm{supp}(b)\subset\{|\phi|\leq R\sqrt{\varepsilon}\}$, $R=\varepsilon^{-1/8}$,

$$\int_{\mathbb{R}^{2d}} W^\varepsilon(\psi_0^\varepsilon)(q,p)\, b(q,p)\,\mathrm{d}q\,\mathrm{d}p = O(\varepsilon^{\beta_2}),\ \ \beta_2\geq 1/8.$$

We also assume localization away from the set

$$\left\{(q_0,p_0)\in\mathbb{R}^{2d}\mid \exists t>0:\phi(q^\pm(t))=0,\ \mathrm{d}\phi(q^\pm(t))p^\pm(t)=0\right\},$$

which contains the points issuing classical trajectories, which arrive at the crossing without a unique continuation through it.

(A2) The test function $a\in\mathcal{C}_c^\infty\left(\mathbb{R}^{2d},\mathbb{C}^{N(\ell)\times N(\ell)}\right)$ has its support at a distance larger than $R\sqrt{\varepsilon}$ with $R=\varepsilon^{-1/8}$ from the crossing, that is

$$\mathrm{supp}(a)\cap\left\{(q,p)\in\mathbb{R}^{2d}\mid\ |\phi(q)|\leq R\sqrt{\varepsilon}\right\}=\emptyset,\qquad R=\varepsilon^{-1/8},$$

and

$$a(q,p)=a^+(q,p)\Pi^+(q)+a^-(q,p)\Pi^-(q),\qquad (q,p)\in\mathbb{R}^{2d}$$

with scalar-valued $a^\pm\in\mathcal{C}_c^\infty(\mathbb{R}^{2d},\mathbb{C})$.

(A3) Within the time-interval $[0,T]$, each of the plus-trajectories arriving at the support of a^+ at time T has performed at most one jump generating minus-trajectories arriving at the support of a^-, which have not jumped at all.

Alternatively, assumptions (A1) and (A3) could also require, that the initial data are associated with $\mathrm{Ran}\Pi^-$ and that each of the minus-trajectories arriving at the support of a^- at time T has performed at most one jump generating plus-trajectories arriving at the support of a^+, which have not jumped at all.

Theorem 2.2. *Let the potential V, the initial data $(\psi_0^\varepsilon)_{\varepsilon>0}$, the observable a, and the time-interval $[0,T]$ fulfill the assumptions (A0), (A1), (A2), and (A3). Let $\chi\in\mathcal{C}_c^\infty([0,T],\mathbb{R})$. Then, there exist positive constants $C,\varepsilon_0>0$ such that for all $0<\varepsilon<\varepsilon_0$ the solution $\psi^\varepsilon(t)$ of the Schrödinger equation (1.1) satisfies*

$$\left|\mathrm{tr}\int_{\mathbb{R}^{2d+1}}\chi(t)\left(W^\varepsilon(\psi^\varepsilon(t))-\mathcal{L}_\varepsilon^t W^\varepsilon(\psi_0)\right)(q,p)\,a(q,p)\,\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t\right|\leq C\,\varepsilon^{1/8}. \tag{2.3}$$

Before entering the proof, we add some remarks. First, if one allows initial data in assumption (A1) with $\beta_1,\beta_2>0$, then the result holds with convergence rate $\varepsilon^{\min(\beta_1,\beta_2,1/8)}$. Second, pointwise convergence holds on time intervals without non-adiabatic jumps, that is when the solution has passed by the jump manifold, see also [17]. However, for pointwise convergence only the limit behaviour without convergence rate can be deduced, since the constants C and ε_0 depend on the cut-off function χ and its derivatives in a way, that possible oscillations in time are not controlled. Finally, in section 6 an extension of the approximation for the cases $\ell=3',5$ with degenerate eigenvalues is given. There, assumption (A2) is generalized to observables a which commute with V, that is $a=\Pi^+a\Pi^++\Pi^-a\Pi^-$.

2.4. Strategy of the proof. For notational convenience, we suppose $\beta_1 = \beta_2 = 1/2$. Otherwise, one has to add $O(\varepsilon^{1/8})$ in all the estimates. We work with the semigroup $(\mathcal{L}^t_{\varepsilon,R})_{t\geq 0}$ and prove convergence with an error of order

$$O(1/(R^5\sqrt{\varepsilon})) + O(R^3\sqrt{\varepsilon}) + O(1/R^2) + O(\sqrt{\varepsilon}\,|\ln\varepsilon|)$$

as $\varepsilon \to 0$ and $R \to +\infty$, which gives the claimed rate when choosing $R = \varepsilon^{-1/8}$. We distinguish between regions of large and small eigenvalue gap, that is between sets $\{|\phi| > C\,R\sqrt{\varepsilon}\}$ with $C = \frac{1}{2}, 1$ and $\{|\phi| \leq R\sqrt{\varepsilon}\}$ on the other hand. For large gap we prove classical transport with an error of size $O(1/(R^5\sqrt{\varepsilon})) + O(1/R^2) + O(\sqrt{\varepsilon})$. Close to the crossing set, proving the relevance of non-adiabatic transitions, we use a microlocal normal form, which reduces the Schrödinger equation to a Landau-Zener type problem with explicitly computable transition rates. There, we introduce an error of order $O(R^3\sqrt{\varepsilon}) + O(1/R^2) + O(\sqrt{\varepsilon}|\ln\varepsilon|) + O(1/(R^5\sqrt{\varepsilon}))$. The combination of both errors will then yield the final estimate of Theorem 2.2.

Proposition 2.3. *Let $c \in \mathcal{C}^\infty_c(\mathbb{R}^{2d},\mathbb{C})$, and let $b \in \mathcal{C}^\infty(\mathbb{R}^\ell,\mathbb{C})$ with ∇b compactly supported. If there exist $C > 0$ and $s_0 > 0$ such that*

$$\forall r \in [-s_0, s_0]: \quad \Phi^r_\pm(\mathrm{supp}(c)) \subset \{|\phi| > C\,R\sqrt{\varepsilon}\},$$

then for all $\chi \in \mathcal{C}^\infty_c(\mathbb{R},\mathbb{R})$ and for all $s \in [t - s_0, t + s_0]$

$$\begin{aligned}
&\mathrm{tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, c(q,p)\, b\Big(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\Big)\, \Pi^\pm(q) W^\varepsilon(\psi^\varepsilon(t))(q,p)\,\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t = \\
&\mathrm{tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, c(q,p)\, b\Big(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\Big)\, \big(\Pi^\pm W^\varepsilon(\psi^\varepsilon(s))\,\Pi^\pm \circ \Phi^{-t+s}_\pm\big)(q,p)\,\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t \\
&\qquad\qquad + O(1/(R^5\sqrt{\varepsilon})) + O(1/R^2) + O(\sqrt{\varepsilon}).
\end{aligned}$$

Proposition 2.3 will be proved in section 4. To use it for the main proof, we need to specify which points of $\mathrm{supp}(a^\pm)$ arrive close to the crossing. We denote the sets of trajectories arriving at respectively arising from the crossing set $\{\phi = 0\}$ by

$$\begin{aligned}
M^{\pm,in} &= \left\{\Phi^t_\pm(q,p) \in \mathbb{R}^{2d} \mid \Phi^t_\pm(q,p) \notin \{\phi = 0\}, \exists t_0 < t : \Phi^{t_0}_\pm(q,p) \in \{\phi = 0\}\right\}, \\
M^{\pm,out} &= \left\{\Phi^t_\pm(q,p) \in \mathbb{R}^{2d} \mid \Phi^t_\pm(q,p) \notin \{\phi = 0\}, \exists t_0 > t : \Phi^{t_0}_\pm(q,p) \in \{\phi = 0\}\right\}.
\end{aligned}$$

The sets $M^{\pm,in/out}$ are smooth submanifolds of $\mathbb{R}^{2d}$. By construction of the semigroup, all phase space points generating backward trajectories passing through the zone of small gap $\{|\phi| \leq R\sqrt{\varepsilon}\}$ and performing a jump are contained in a neighborhood $\Omega^\pm$ of the intersection of the support of $a^\pm$ with $M^{\pm,out}$.

Some of the random trajectories reaching $\Omega^\pm$ touch the crossing set. We consider one of them, which arrives at time t_0 at (q_0, p_0) with $\phi(q_0) = 0$, $\mathrm{d}\phi(q_0)p_0 \neq 0$, and choose the associated point (q_0, t_0, p_0, τ_0) in the phase space of space-time, where $\tau_0 = -\frac{1}{2}|p_0|^2 - v(q_0)$ is the energy-coordinate. The normal form theorems [1, 2, 6] give neighborhoods of these points, on which the Schrödinger equation (1.1) microlocally reduces to a Landau-Zener type problem

$$-i\varepsilon\partial_s v^\varepsilon = V_\ell(s, \tilde{z} + \gamma_\varepsilon(z,\zeta))v^\varepsilon + O(\varepsilon^\infty).$$

These model problems have explicitly computable transition rates in the scattering regime, see [7, 8]. The compact subset of $\{|\phi| < R\sqrt{\varepsilon}\}$, which is touched by the backwards trajectories coming from $\Omega^\pm$, can be covered by finitely many of these neighborhoods projected to $\mathbb{R}^{2d}$, and without loss of generality we assume that one of them suffices.

Moreover, for each point in $\Omega^\pm$ being reached by a random trajectory at time t there are positive numbers $0 < t_{\rm f}^\pm < t_{\rm i}^\pm$, such that at time $t - t_{\rm i}^\pm$ and $t - t_{\rm f}^\pm$ the trajectories are contained in an annulus $\{C_1\sqrt{\varepsilon} < |\phi| < C_2\sqrt{\varepsilon}\}$ with $C_1, C_2 > 0$ and have performed their only jump within the interval $]t - t_{\rm i}^\pm, t - t_{\rm f}^\pm[$, whose length is denoted by $\delta_t^\pm = t_{\rm i}^\pm - t_{\rm f}^\pm$. These quantities are well-defined, since the trajectories are transverse to the crossing set. Choosing $C_1 = \frac{R}{2}$ and $C_2 = R$, $\Omega^\pm$ can be covered by finitely many open sets, such that each of these sets can be associated with such points of time $t_{\rm i}^\pm$ and $t_{\rm f}^\pm$. Without loss of generality, we assume that one of them is enough. Then, we have by Proposition 2.3 with $C = \frac{1}{2}$

$$\begin{aligned}
&{\rm tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, a^\pm(q,p)\, \Pi^\pm(q) W^\varepsilon(\psi^\varepsilon(t))(q,p)\, {\rm d}q\, {\rm d}p\, {\rm d}t = \\
&{\rm tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, a^\pm(q,p)\, \left(\Pi^\pm W^\varepsilon\big(\psi^\varepsilon(t - t_{\rm f}^\pm)\big)\, \Pi^\pm \circ \Phi_\pm^{-t_{\rm f}^\pm}\right)(q,p)\, {\rm d}q\, {\rm d}p\, {\rm d}t \\
&\qquad\qquad + O(1/(R^5\sqrt{\varepsilon})) + O(1/R^2) + O(\sqrt{\varepsilon}).
\end{aligned}$$

Then, we perform a cut-off of the symbol $a^\pm \circ \Phi_\pm^{t_{\rm f}^\pm}$. We consider a smooth compactly supported function $\chi_0 \in \mathcal{C}_c^\infty(\mathbb{R}^\ell, \mathbb{R})$ such that $\chi_0(u) = 1$ on $\{|u| < 1\}$ and $\chi_0(u) = 0$ for $\{|u| > 2\}$. We write

$$\begin{aligned}
\left(a^\pm \circ \Phi_\pm^{t_{\rm f}^\pm}\right)(q,p) &= a_{BO}^\pm(q,p) + a_{LZ}^\pm(q,p), \\
a_{BO}^\pm(q,p) &= \left(a^\pm \circ \Phi_\pm^{t_{\rm f}^\pm}\right)(q,p)\, \left(1 - \chi_0\left(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\right)\right), \\
a_{LZ}^\pm(q,p) &= \left(a^\pm \circ \Phi_\pm^{t_{\rm f}^\pm}\right)(q,p)\, \chi_0\left(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\right).
\end{aligned}$$

Since the trajectories, which pass within the time-interval $]t - t_{\rm i}^\pm, t - t_{\rm f}^\pm[$ through the support of $a_{BO}^\pm$, do not jump, Proposition 2.3 with $C = 1$ is enough to deal with the Born-Oppenheimer part. The analysis of the Landau-Zener part, however, involves non-adiabatic transitions. For points $(q,p) \in {\rm supp}(a_{LZ}^\pm)$ we have $|\phi(q)| \le 2R\sqrt{\varepsilon}$. Hence, not all of the trajectories passing through the support of $a_{LZ}^\pm$ jump. Nevertheless, we argue as if all of them did. Indeed, the transition coefficients generated by these added jumps are exponentially small with respect to ε, since they occur for points (q,p) with $|\phi(q)| > R\sqrt{\varepsilon}$.

Proposition 2.4. *Let $0 < t_{\rm f}^\pm < t_{\rm i}^\pm$ be such that at $t - t_{\rm i}^\pm$ and $t - t_{\rm f}^\pm$ all random trajectories arriving at time t in $\Omega^\pm$ are contained in $\left\{\frac{R}{2}\sqrt{\varepsilon} \le |\phi| \le R\sqrt{\varepsilon}\right\}$ and have performed their only jump within the interval $]t - t_{\rm i}^\pm, t - t_{\rm f}^\pm[$ of length $\delta_t^\pm = t_{\rm i}^\pm - t_{\rm f}^\pm$. Then, for all $\chi \in \mathcal{C}_c^\infty([0,T], \mathbb{R})$*

$$\begin{aligned}
&{\rm tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, W^\varepsilon(\psi^\varepsilon(t))(q,p)\, a_{LZ}^\pm(q,p) \Pi^\pm(q)\, {\rm d}q\, {\rm d}p\, {\rm d}t = \\
&{\rm tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\, W^\varepsilon(\psi^\varepsilon(t - \delta_t^\pm))(q,p) (\mathcal{L}_{\varepsilon,R}^{\delta_t^\pm} a_{LZ}^\pm)(q,p)\, \Pi^+(q)\, {\rm d}q\, {\rm d}p\, {\rm d}t \\
&\qquad + O(1/R^2) + O(R^3\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|) + O(1/(R^5\sqrt{\varepsilon})).
\end{aligned} \tag{2.4}$$

One observes that in the right-hand side of (2.4) only the plus-projector Π^+ appears. This comes from the fact that at time $t - t_{\rm i}^{\pm}$ the contribution on the minus mode is of order $O(1/(R^5\sqrt{\varepsilon})) + O(1/R^2) + O(\sqrt{\varepsilon})$, which is due to Proposition 2.3 and the initial data being only associated with $\operatorname{Ran}\Pi^+$. Finally, again the classical transport result of Proposition 2.3 relates the right-hand side of (2.4) with the initial data, and the proof of our main result Theorem 2.2 is complete.

3. Numerical experiments. Before giving the detailed proof, we present numerical experiments illustrating the theoretical convergence result and the effectiveness of the algorithm. We consider a two-level Schrödinger equation with codimension two crossing in two space dimensions, which models the photoisomerization of retinal in rhodopsin. This conformational change is considered as the first step of vision. In [13], computations with the model Hamiltonian

$$-\frac{\omega}{2}\partial_x^2 - \frac{1}{2m}\partial_\varphi^2 + \tfrac{1}{2}\omega x^2 + \begin{pmatrix} \frac{1}{2}W_0(1-\cos\varphi) & \lambda x \\ \lambda x & E_1 - \frac{1}{2}W_1(1-\cos\varphi) + \kappa x \end{pmatrix}$$

have qualitatively reproduced spectroscopic information of the molecule. The two effective coordinates $(\varphi, x) \in]-\frac{\pi}{2}, \frac{3\pi}{2}] \times \mathbb{R}$ consist of the reaction coordinate ϕ and a collective coordinate x. The parameters are $m^{-1} = 4.84 \cdot 10^{-4}$, $E_1 = 2.48$, $W_0 = 3.6$, $W_1 = 1.09$, $\omega = \lambda = 0.19$, and $\kappa = 0.1$ (all in eV, $\hbar = 1$), see note 18 in [13]. Setting

$$\varepsilon = m^{-1/2} = 0.022, \qquad q_1 = \varphi, \qquad q_2 = \frac{\varepsilon}{\sqrt{\omega}}\, x,$$

one obtains a rescaled Hamiltonian $-\frac{\varepsilon^2}{2}\Delta_q + V(q)$ with potential

$$V(q) = \tfrac{1}{2}(\beta q_2)^2 + \begin{pmatrix} \frac{1}{2}W_0(1-\cos q_1) & \alpha_1 q_2 \\ \alpha_1 q_2 & E_1 - \frac{1}{2}W_1(1-\cos q_2) + \alpha_2 q_2 \end{pmatrix},$$

whose parameters $(\alpha_1, \alpha_2) = \frac{\sqrt{\omega}}{\varepsilon}(\kappa, \lambda) \approx (2,\, 3.8)$ and $\beta = \omega/\varepsilon \approx 8.6$ are of order one with respect to ε. Fixing these values of (α_1, α_2) and β, we run a series of experiments for varying values of the semiclassical parameter ε and hopping ranges R,

$$\varepsilon \in \{0.0005, 0.001, 0.005, 0.01, 0.015, 0.02, 0.022, 0.03\}, \qquad R \in \{1, 2, 3\},$$

in the following set-up. We consider normalized Gaussian initial data associated with the plus-level, that is

$$\psi_0^\varepsilon(q) = (\varepsilon\pi)^{-1/2} \exp\!\big(-\tfrac{1}{2\varepsilon}|q - q_0^\varepsilon|^2 + \tfrac{i}{\varepsilon}\, p_0 \cdot (q - q_0^\varepsilon)\big)\, v^+(q),$$

where $v^+(q)$ is a normalized eigenvector of $V(q)$ for the eigenvalue $v(q) + |\phi(q)|$, which depends smoothly on q. The initial center in position space

$$q_0^\varepsilon = \big(1.63 - 4\sqrt{\varepsilon},\, 0.5\sqrt{\varepsilon}\big)$$

is chosen left of the two crossing points $(\gamma_{\rm l}, 0)$, $(\gamma_{\rm r}, 0)$, where $\gamma_{\rm l,r}$ are the two solutions of $\cos\varphi = 1 - 2E_1/(W_0 + W_1)$ for $\varphi \in]-\frac{\pi}{2}, \frac{3\pi}{2}]$, that is $\gamma_{\rm l} \approx 1.63$ and $\gamma_{\rm r} \approx 4.65$. The initial momentum center and the time-interval,

$$p_0 = (1, 0), \qquad [0, T] = [0, 7\sqrt{\varepsilon}],$$

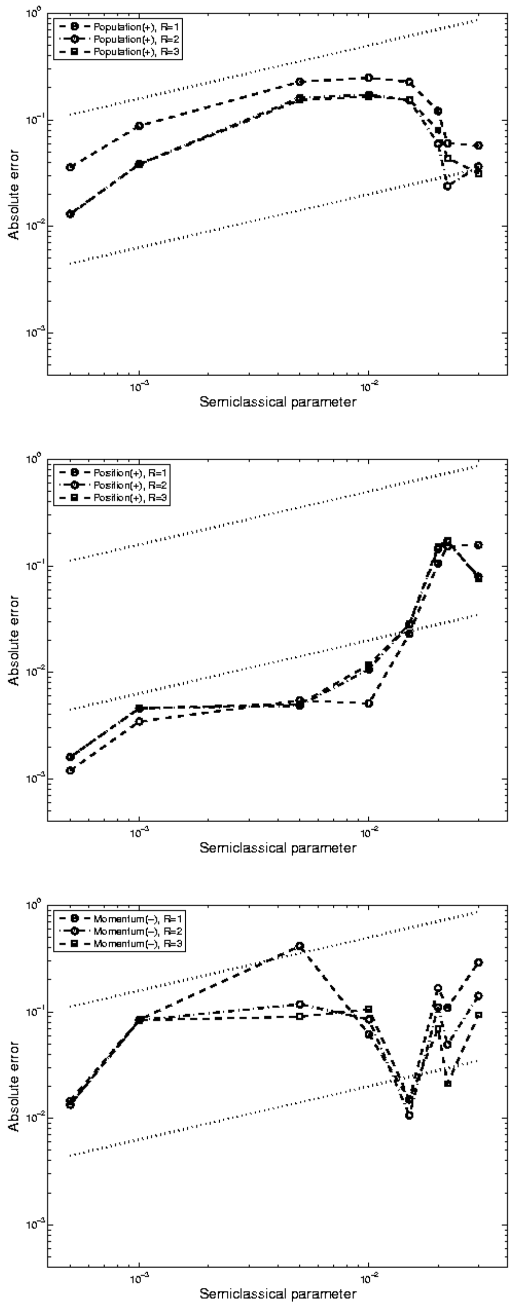

FIG. 3.1. *Double logarithmic plots of the absolute error, when comparing the outcome of the surface hopping algorithm with a numerically converged pseudospectral splitting scheme. The semiclassical parameter ε varies in the set $\{0.0005, 0.001, 0.005, 0.01, 0.015, 0.02, 0.022, 0.03\}$. The dashed, dashed-dotted, and the solid lines refer to a hopping range $R = 1, 2, 3$, respectively. The three plots show the error of the level population $\|\Pi^+(q)\psi^\varepsilon(q,t)\|^2$, of the position expectation value $\langle \Pi^+(q)\psi^\varepsilon(q,t), q\Pi^+(q)\psi^\varepsilon(q,t)\rangle$, and of the momentum expectation $\langle \Pi^-(q)\psi^\varepsilon(q,t), -i\varepsilon\nabla_q\Pi^-(q)\psi^\varepsilon(q,t)\rangle$ at the final time $T = 7\sqrt{\varepsilon}$. All errors lie in and below the corridor defined by the two functions $\varepsilon \mapsto 5\sqrt{\varepsilon}$ and $\varepsilon \mapsto \frac{1}{5}\sqrt{\varepsilon}$, which are represented by dotted lines.*

Table 3.1

The table shows final level populations and particle numbers as well as the accuracy of the reference solver. The population of the upper level $\|\Pi^+\psi^\varepsilon(t)\|^2$ at the final time $T = 7\sqrt{\varepsilon}$ is computed by the reference solver, a pseudo-spectral Strang splitting scheme, and illustrates leading order non-adiabatic transitions for all test cases. Depending on the hopping range R, the final particle numbers of the surface hopping algorithm vary between three and thirty thousand. The reference accuracy is the difference in L^2-norm of the final wave function computed with full and half resolution. For all computations it is less than 10^{-4}.

ε	0.0005	0.001	0.005	0.01	0.015	0.02	0.022	0.03
$\|\Pi^+\psi^\varepsilon(t)\|^2$	0.499	0.576	0.792	0.378	0.263	0.276	0.276	0.239
♯ particles, $R=1$	5396	5229	3730	3548	3316	3274	3482	3292
♯ particles, $R=2$	6650	6353	4732	4766	4801	5569	6106	8281
♯ particles, $R=3$	7392	6881	5809	6155	7119	10 536	12 581	34 485
ref. accuracy $\cdot 10^5$	3.44	2.89	7.57	2.56	2.47	2.45	2.47	2.63

are chosen such that the wave function passes only the left crossing point $(\gamma_1, 0)$ once without returning to it again. The upper level populations $\|\Pi^+\psi^\varepsilon(t)\|^2$ at the final time $T = 7\sqrt{\varepsilon}$, which are given in table 3.1, confirm that the experimental set-up produces leading order non-adiabatic transitions.

The numerical realization of the surface hopping semigroup $(\mathcal{L}^t_{\varepsilon,R})_{t\geq 0}$ is the same as for the simulations of models with linear isotropic potential matrix presented in [16], up to adding the $R\sqrt{\varepsilon}$-dependent jump criterion

$$t \mapsto |\phi(q^\pm(t))| \text{ has a local minimum in } t = t^* \text{ and } |\phi(q^\pm(t^*))| \leq R\sqrt{\varepsilon}.$$

The initial sampling is performed on 16×16 grids in position and momentum space, and the classical transport is discretized by the explicit Runge-Kutta method of Dormand and Prince `DOPRI45`. For comparison, we have also solved the Schrödinger equation (1.1) by a numerically converged pseudo-spectral Strang splitting scheme. The solutions obtained with a 1024×512 space grid on the computational domain $[1.63-8\sqrt{\varepsilon}, 1.63+16\sqrt{\varepsilon}]\times[-6\sqrt{\varepsilon}, 6\sqrt{\varepsilon}]$ and with 10^4 time steps are regarded as a reference, since they differ in L^2-norm from the corresponding solution with a fourth of the grid points and half the time steps by less than 10^{-4}, see table 3.1. We have computed the following quadratic quantities of the wave function at final time $T = 7\sqrt{\varepsilon}$: the level populations $\|\Pi^\pm\psi^\varepsilon(t)\|^2$ and the expectation values of position and momentum on each level,

$$\langle\Pi^\pm(q)\psi^\varepsilon(q,t), q\Pi^\pm(q)\psi^\varepsilon(q,t)\rangle, \qquad \langle\Pi^\pm(q)\psi^\varepsilon(q,t), -i\varepsilon\nabla_q\Pi^\pm(q)\psi^\varepsilon(q,t)\rangle.$$

We note, that the reference solver restricts the length of the time-interval to $7\sqrt{\varepsilon}$, since the dynamics can only be resolved, as long as the solution stays well-localized in the computational domain, while for the fixed number of 1024×512 grid points the size of the computational domain affects the numerical accuracy.

Comparing the outcome of the two algorithms, we find all errors within and below the corridor $[\frac{1}{5}\sqrt{\varepsilon}, 5\sqrt{\varepsilon}]$, see figure 3.1, which is better than the proven convergence rate $\varepsilon^{1/8}$. Moreover, the errors increase monotonically when increasing the semi-classical parameter, however, when entering the range $\varepsilon \geq 0.01$ we observe different dependencies. The level populations' error starts decreasing, while the error of the momentum expectation oscillates. We have no mathematical explanation for these observations and can only make an educated guess. The good convergence rate might be caused by the localization properties of the initial Gaussian wave packet combined with the short length of the time-interval. Indeed, the error terms $O(1/R^2)$ and

$O(1/(R^5\sqrt{\varepsilon}))$ in the approximation by classical transport in Proposition 2.3 might be negligible in this case as well as the contribution $O(1/R^2)$ in Proposition 2.4, which is due to localization in energy. The tendencies for large semi-classical parameter, however, are clearly beyond the reach of our asymptotic analysis.

Table 3.1 shows, that an increase of the hopping range R increases the number of final particles, that is the number of jumps within the overall time-interval. We obtain particle numbers around three and thirty thousand for $R = 1$ and $R = 3$, respectively, resulting in half a minute and five minutes computing time for our implementation of the algorithm in *Matlab 7.0* on a 3GHz Pentium 4 computer. However, an enlarged hopping range need not improve the accuracy of the approximation. The plots in figure 3.1 mostly display smaller errors for larger R, but the level populations in the physical relevant range of $\varepsilon = 0.022$ have the most accurate computation for $R = 2$.

Summarizing, the numerical experiments are consistent with the theoretical result, but also present a better convergence rate and tendencies in the range of larger semi-classical parameter, which seem to be unexplainable by our asymptotic analysis. A systematic comparison with the well-established surface hopping algorithms of chemical physics is in progress.

4. Propagation outside the crossing zone. We now begin proving our main result. The first step is to establish the validity of the classical transport approximation in the zone of large eigenvalue gap $\{|\phi| \geq C\, R\sqrt{\varepsilon}\}$.

Proof. [Proposition 2.3] Our aim is to prove

$$\mathrm{tr}\int \left\{\chi(t)c(q,p)b\Big(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\Big)\,,\tau + \tfrac{1}{2}|p|^2 + v(q) \pm |\phi(q)\right\}\Pi^{\pm}(q)W^{\varepsilon}(\psi^{\varepsilon}(t))(q,p)\,\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t$$
$$= O(1/R^2) + O(1/(R^5\sqrt{\varepsilon})) + O(\sqrt{\varepsilon}),$$

since then classical transport follows immediately. The key argument is the estimation of the action of the commutator

$$K = \tfrac{1}{\varepsilon}\left[\chi(t)\mathrm{op}_{\varepsilon}\Big(c(q,p)b\Big(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\Big)\,\Pi^{\pm}(q)\Big)\ ,\ -i\varepsilon\partial_t - \tfrac{\varepsilon^2}{2}\Delta_q + V(q)\right]$$

on the solution of the Schrödinger equation (1.1). Indeed, observing that

$$(K\psi^{\varepsilon},\psi^{\varepsilon})_{L^2(\mathbb{R}^{d+1})} = 0,$$

we are going to prove

$$(K\psi^{\varepsilon},\psi^{\varepsilon})_{L^2(\mathbb{R}^{d+1})} = O(1/R^2) + O(1/(R^5\sqrt{\varepsilon})) + O(\sqrt{\varepsilon}) +$$
$$\Big(\mathrm{op}_{\varepsilon}\Big(\Big\{\chi(t)c(q,p)b\Big(\tfrac{\phi(q)}{R\sqrt{\varepsilon}}\Big)\,,\tau + \tfrac{1}{2}|p|^2 + v(q) \pm |\phi(q)\Big\}\,\Pi^{\pm}(q)\Big)\,\psi^{\varepsilon},\psi^{\varepsilon}\Big)_{L^2(\mathbb{R}^{d+1})},$$

where $\mathrm{op}_{\varepsilon}$ also denotes Weyl quantized operators acting on space-time variables. We use the scaling operator

$$T : L^2_{\mathrm{loc}}(\mathbb{R}^{d+1}) \to L^2_{\mathrm{loc}}(\mathbb{R}^{d+1}), \quad (T\psi)(t,q) = \varepsilon^{d/4}\psi(t,\sqrt{\varepsilon}q) \tag{4.1}$$

and write

$$T^*KT = \tfrac{1}{\varepsilon}\left[\mathrm{op}_1\Big(\chi(t)c(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)b\Big(\tfrac{\phi(\sqrt{\varepsilon}q)}{R\sqrt{\varepsilon}}\Big)\,\Pi^{\pm}(\sqrt{\varepsilon}q)\Big)\ ,\ -i\varepsilon\partial_t - \tfrac{\varepsilon}{2}\Delta_q + V(\sqrt{\varepsilon}q)\right].$$

We have to deal carefully with the ε, R dependence of the symbol in the left-hand side of the commutator. For all multi-indeces $\alpha \in \mathbb{N}^d$,

$$D^{\alpha}\big(\Pi^{\pm}(q)\big) = O(|\phi(q)|^{-|\alpha|}).$$

Since $|\phi(q)| > C\, R\sqrt{\varepsilon}$ on the support of $b_{\varepsilon,R}(q,p) := c(q,p)b\Big(\frac{\phi(q)}{R\sqrt{\varepsilon}}\Big)$, we have

$$D_q^\alpha\big((b_{\varepsilon,R}\Pi^\pm)(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(1), \quad D_p^\alpha\big((b_{\varepsilon,R}\Pi^\pm)(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(\varepsilon^{|\alpha|/2}) \tag{4.2}$$

for $\alpha \in \mathbb{N}^d$. By the symbolic calculus of Lemma A.2,

$$\begin{aligned}\tfrac{1}{\varepsilon}\left[\mathrm{op}_1\big((b_{\varepsilon,R}\Pi^\pm)\left(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p\right)\big), \mathrm{op}_1\big(\tfrac{\varepsilon}{2}|p|^2\big)\right] &= \tfrac{1}{i}\,\mathrm{op}_1(\{(b_{\varepsilon,R}\Pi^\pm)\left(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p\right), \tfrac12|p|^2\})\\ &= \tfrac{1}{i}\,\mathrm{op}_1\big(\{b_{\varepsilon,R}(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p),\tfrac12|p|^2\}\Pi^\pm(\sqrt{\varepsilon}q)\big) - \tfrac{1}{i}\,\mathrm{op}_1\big(r_0(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big)\end{aligned}$$

where

$$r_0(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) = b_{\varepsilon,R}(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\sum_{j=1}^d \sqrt{\varepsilon}p_j(\partial_{q_j}\Pi^\pm)(\sqrt{\varepsilon}q). \tag{4.3}$$

Moreover, in view of $[\Pi^\pm, V] = 0$ and (4.2),

$$\begin{aligned}\tfrac{1}{\varepsilon}\left[\mathrm{op}_1\big((b_{\varepsilon,R}\Pi^\pm)\left(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p\right)\big)\ ,\ V(\sqrt{\varepsilon}q)\right] &= \tfrac{1}{2i\varepsilon}\,\mathrm{op}_1(\{(b_{\varepsilon,R}\Pi^\pm)(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p), V(\sqrt{\varepsilon}q)\})\\ &\quad -\tfrac{1}{2i\varepsilon}\,\mathrm{op}_1(\{V(\sqrt{\varepsilon}q),(b_{\varepsilon,R}\Pi^\pm)(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\}) + O(\varepsilon).\end{aligned}$$

Working on the Poisson brackets involving $V = v + V_\ell(\phi)$, we first observe

$$\big\{b_{\varepsilon,R}\Pi^\pm, V_\ell(\phi)\big\} - \big\{V_\ell(\phi), b_{\varepsilon,R}\Pi^\pm\big\} = \Pi^\pm\{b_{\varepsilon,R}, V_\ell(\phi)\} - \{V_\ell(\phi), b_{\varepsilon,R}\}\,\Pi^\pm.$$

Using that $V_\ell(\phi) = |\phi|(\Pi^+ - \Pi^-)$ and $\partial_{q_j}\Pi^\pm = \Pi^\pm(\partial_{q_j}\Pi^\pm) + (\partial_{q_j}\Pi^\pm)\Pi^\pm$, we get

$$\big\{b_{\varepsilon,R}\Pi^\pm, V_\ell(\phi)\big\} - \big\{V_\ell(\phi), b_{\varepsilon,R}\Pi^\pm\big\} = \pm 2\{b_{\varepsilon,R},|\phi|\}\Pi^\pm \pm 2\,|\phi|\sum_{j=1}^d\big(\partial_{p_j}b_{\varepsilon,R}\big)\,\partial_{q_j}\Pi^\pm$$

and set

$$\begin{aligned} r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) &= \tfrac{1}{\varepsilon}|\phi(\sqrt{\varepsilon}q)|\sum_{j=1}^d \partial_{p_j}\big(b_{\varepsilon,R}(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big)\,\partial_{q_j}\big(\Pi^\pm(\sqrt{\varepsilon}q)\big)\\ &= |\phi(\sqrt{\varepsilon}q)|\sum_{j=1}^d(\partial_{p_j}c)(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,b\Big(\tfrac{\phi(\sqrt{\varepsilon}q)}{R\sqrt{\varepsilon}}\Big)\,(\partial_{q_j}\Pi^\pm)(\sqrt{\varepsilon}q).\end{aligned} \tag{4.4}$$

Now, collecting all the different pieces, we have

$$\begin{aligned}K = {}&\mathrm{op}_\varepsilon\big(\big\{\chi(t)b_{\varepsilon,R}(q,p)\,,\,\tau + \tfrac12|p|^2 + v(q) \pm |\phi(q)|\big\}\,\Pi^\pm(q)\big)\\ &+\mathrm{op}_\varepsilon(\chi(t)r_0(q,p)) + \mathrm{op}_\varepsilon(\chi(t)r_1(q,p)) + O(\varepsilon).\end{aligned}$$

Hence, our claim follows from the analysis of r_0 and r_1, which is carried out in the following Lemma 4.1. ◻

Lemma 4.1. *Let ψ^ε solve the Schrödinger equation (1.1). For the matrix-valued functions r_0 and r_1 defined in (4.3) and (4.4), respectively, one has*

$$\begin{aligned}&\left(\mathrm{op}_\varepsilon\left(\chi(t)r_0(q,p)\right)\psi^\varepsilon,\psi^\varepsilon\right)_{L^2(\mathbb{R}^{d+1})} = O(\sqrt{\varepsilon}) + O(1/R^2) + O(1/(R^5\sqrt{\varepsilon})),\\ &\left(\mathrm{op}_\varepsilon\left(\chi(t)r_1(q,p)\right)\psi^\varepsilon,\psi^\varepsilon\right)_{L^2(\mathbb{R}^{d+1})} = O(\sqrt{\varepsilon}) + O(1/R^2).\end{aligned}$$

Proof. Both functions have off-diagonal matrix structure, that is $r_0(q,p)$ and $r_1(q,p)$ do not commute with $V(q)$. However, since r_1 contains an additional factor $|\phi|$, it is less singular than r_0, in the sense that for $\alpha \in \mathbb{N}^d$ and (q,p) with $|\phi(\sqrt{\varepsilon}q)| > CR\sqrt{\varepsilon}$

$$D_q^\alpha\big(r_0(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(R^{-|\alpha|-1}\varepsilon^{-1/2}), \qquad D_q^\alpha\big(r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(R^{-|\alpha|}).$$

We begin with r_1. We write $r_1 = \Pi^- r_1 \Pi^+ + \Pi^+ r_1 \Pi^-$ and work successively with each part. Thus, without loss of generality, we suppose that $\Pi^- r_1 \Pi^+ = r_1$. The strategy is to reuse the Schrödinger equation, since

$$r_1 = \Pi^- r_1 \Pi^+ = \tfrac{1}{2|\phi|}[r_1, V_\ell(\phi)] = \tfrac{1}{2|\phi|}[r_1, \tau + \tfrac12|p|^2 + V].$$

With the scaling operator T defined in (4.1), we obtain

$$\mathrm{op}_\varepsilon(\chi(t)r_1(q,p)) = T^*\mathrm{op}_1\Big(\Big[\tfrac{\chi(t)}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p), \varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big]\Big)\, T.$$

Then, by the symbolic calculus of Lemma A.2,

$$\begin{aligned}&\mathrm{op}_1\Big(\Big[\tfrac{\chi(t)}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p), \varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big]\Big)\\ &= \Big[\mathrm{op}_1\Big(\tfrac{\chi(t)}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\Big), \mathrm{op}_1\big(\varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\big)\Big] + \mathrm{op}_1(r_2(t,\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)),\end{aligned}$$

where

$$r_2(t,\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) = \chi(t)\tilde r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) + \varepsilon\tfrac{\chi'(t)}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) + \varepsilon\chi(t)\tilde r_2(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$$

with

$$\begin{aligned}\tilde r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) = \tfrac{1}{2i}&\Big\{\tfrac{1}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,,\ \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big\}\\ &-\tfrac{1}{2i}\Big\{\tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\,,\ \tfrac{1}{2|\phi(\sqrt{\varepsilon}q)|}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\Big\}.\end{aligned}$$

The term $\tilde r_2(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$ contains the commutator of second derivatives in p of the symbol $|\phi(\sqrt{\varepsilon}q)|^{-1}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$ with second derivatives of $V(\sqrt{\varepsilon}q)$ and a linear combination of derivatives in (q,p) of order greater than or equal to three. Hence, by Lemma A.2

$$\mathrm{op}_\varepsilon\left(\chi(t)\tilde r_2(q,p)\right) = O(\varepsilon\sqrt{\varepsilon}) + O(\varepsilon/R^4) \ \text{ in } \ \mathcal{L}(L^2(\mathbb{R}^{d+1})).$$

It remains to study $\tilde r_1$. Since derivatives in p of $r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$ generate powers of $\sqrt{\varepsilon}$, the bracket with $V(\sqrt{\varepsilon}q)$ gives

$$\mathrm{op}_1\Big(\chi(t)\Big\{\tfrac{1}{2|\phi(q\sqrt{\varepsilon})|} r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p), V(\sqrt{\varepsilon}q)\Big\}\Big) = O(\sqrt{\varepsilon}/R) \ \text{ in } \ \mathcal{L}(L^2(\mathbb{R}^{d+1})).$$

For the bracket with $\frac{\varepsilon}{2}|p|^2$ one has

$$\begin{aligned}&\Big\{\tfrac{1}{2|\phi(\sqrt{\varepsilon}q)|} r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,,\ \tfrac{\varepsilon}{2}|p|^2\Big\} =\\ &\qquad \frac{\sqrt{\varepsilon}}{2|\phi(\sqrt{\varepsilon}q)|}\,\sqrt{\varepsilon}p\cdot\nabla_q\left(r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\right) + \frac{\varepsilon}{2}\frac{\mathrm{d}\phi(\sqrt{\varepsilon}q)\sqrt{\varepsilon}p\cdot\phi(\sqrt{\varepsilon}q)}{|\phi(\sqrt{\varepsilon}q)|^3}\, r_1(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p).\end{aligned}$$

Both terms give a contribution of order $O(1/R^2)$, but since they are not purely off-diagonal, the preceding commutator argument cannot be reiterated. Hence,

$$\mathrm{op}_\varepsilon(\chi(t)\tilde r_1(q,p)) = O(1/R^2) + O(\sqrt{\varepsilon}/R), \qquad \mathrm{op}_\varepsilon\Big(\varepsilon\,\tfrac{\chi'(t)}{|\phi(q)|}\,r_1(q,p)\Big) = O(\sqrt{\varepsilon}/R)$$

in $\mathcal{L}(L^2(\mathbb{R}^{d+1}))$, and we have proven one part of the Lemma.

In the case of the more singular symbol r_0, the previous strategy results in an error of size

$$\tfrac{1}{R\sqrt{\varepsilon}}\left(O(\varepsilon\sqrt{\varepsilon}) + O(\varepsilon/R^4) + O(\sqrt{\varepsilon}/R) + O(1/R^2)\right)$$
$$= O(\sqrt{\varepsilon}) + O(1/R^2) + O(1/(R^3\sqrt{\varepsilon})).$$

However, the special form of r_0 allows to ameliorate the term of order $O(1/(R^3\sqrt{\varepsilon}))$, which stems from the Poisson bracket with $\frac{\varepsilon}{2}|p|^2$. Indeed, we observe that

$$p\cdot\nabla\Pi^+(q) = \tfrac{1}{4|\phi(q)|^3}\,[V_\ell(\phi(q))\,,\,[V_\ell(\phi(q)),V_\ell(\mathrm{d}\phi(q)p)]]\,.$$

Therefore, the bracket with $\frac{\varepsilon}{2}|p|^2$ in the $\tilde r_1$ term writes as

$$\Big\{\tfrac{1}{4|\phi(\sqrt{\varepsilon}q)|^3}\,b_{\varepsilon,R}(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,\big[V_\ell(\phi(\sqrt{\varepsilon}q)),V_\ell(\mathrm{d}\phi(\sqrt{\varepsilon}q)\sqrt{\varepsilon}p)\big]\,,\,\tfrac{\varepsilon}{2}|p|^2\Big\}$$
$$= \tfrac{\varepsilon}{|\phi(\sqrt{\varepsilon}q)|^4}\,\big[V_\ell(\phi(\sqrt{\varepsilon}q)),G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big]$$

for some matrix-valued function G_ε with

$$D_q^\alpha\big(G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(1), \qquad D_p^\alpha\big(G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big) = O(\varepsilon^{|\alpha|/2})$$

for all $\alpha\in\mathbb{N}^d$. We then set

$$\tilde r_3(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p) := \tfrac{\varepsilon}{|\phi(\sqrt{\varepsilon}q)|^4}\,\big[V_\ell(\phi(\sqrt{\varepsilon}q)),G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\big]$$
$$= -\Big[\tfrac{\varepsilon}{|\phi(\sqrt{\varepsilon}q)|^4}\,G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,,\,\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big]\,,$$

and obtain

$$\begin{aligned}
&(\mathrm{op}_\varepsilon(\chi(t)\tilde r_3(q,p))\,\psi^\varepsilon\,,\,\psi^\varepsilon)_{L^2(\mathbb{R}^{d+1})}\\
&= \Big(T^*\mathrm{op}_1\Big(\Big[\tfrac{\varepsilon\chi(t)}{|\phi(\sqrt{\varepsilon}q)|^4}\,G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,,\,\varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big]\Big)\,T\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}\\
&= \tfrac{1}{2i}\Big(T^*\mathrm{op}_1\Big(\Big\{\tfrac{\varepsilon\chi(t)}{|\phi(\sqrt{\varepsilon}q)|^4}\,G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\,,\,\varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\Big\}\Big)\,T\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}\\
&- \tfrac{1}{2i}\Big(T^*\mathrm{op}_1\Big(\Big\{\varepsilon\tau + \tfrac{\varepsilon}{2}|p|^2 + V(\sqrt{\varepsilon}q)\,,\,\tfrac{\varepsilon\chi(t)}{|\phi(\sqrt{\varepsilon}q)|^4}\,G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)\Big\}\Big)\,T\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}\\
&+ \big(T^*\mathrm{op}_1(\tilde r_4(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p))T\psi^\varepsilon\,,\,\psi^\varepsilon\big)_{L^2(\mathbb{R}^{d+1})}\,,
\end{aligned}$$

where $\tilde r_4(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$ contains the commutator of second order derivatives in p of $\varepsilon\,\chi(t)|\phi(\sqrt{\varepsilon}q)|^{-4}\,G_\varepsilon(\sqrt{\varepsilon}q,\sqrt{\varepsilon}p)$ with second derivatives of $V(\sqrt{\varepsilon}q)$ and a linear combination of higher order derivatives in (q,p). Hence, $\mathrm{op}_\varepsilon(\tilde r_4(q\sqrt{\varepsilon},p\sqrt{\varepsilon})) = O(\varepsilon/R^4)$ in $\mathcal{L}(L^2(\mathbb{R}^{d+1}))$. Since the Poisson brackets give a contribution of order $O(1/(R^5\sqrt{\varepsilon})) + O(1/R^4)$, the other part of the Lemma is proven, too. □

5. Transitions near the crossing. The microlocal normal form used for proving Proposition 2.4 holds locally near some point $(q_0, t_0, p_0, \tau_0) \in \mathbb{R}^{2d+2}$ of the phase space of space-time, which is a crossing point in the sense that $\phi(q_0) = 0$ and $\tau_0 + v(q_0) + \frac{1}{2}|p_0|^2 = 0$.

5.1. Localization in energy. For localization in energy, we consider a cut-off function $\theta \in \mathcal{C}_c^\infty(\mathbb{R})$, $0 \le \theta \le 1$, with $\theta(x) = 1$ for $|x| \le \frac{1}{2}$ and $\theta(x) = 0$ for $|x| > 1$. We set

$$\lambda^\pm(q,p,\tau) = \tau + v(q) + \tfrac{1}{2}|p|^2 \pm |\phi(q)|,$$

and crucially use the following lemma for suitably reformulating Proposition 2.4.

LEMMA 5.1. *Let* $c \in \mathcal{C}_c^\infty(\mathbb{R}^{2d+1+\ell}, \mathbb{C})$. *If* $c_{\varepsilon,R}(t,q,p) = c(t,q,p,\phi(q)/(R\sqrt{\varepsilon}))$ *is supported in* $\{|\phi| \ge \frac{R}{2}\sqrt{\varepsilon}\}$*, then*

$$\operatorname{tr}\int_{\mathbb{R}^{2d+1}} W^\varepsilon(\psi^\varepsilon(t))(q,p) c_{\varepsilon,R}(t,q,p)\Pi^\pm(q)\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t = O(1/R^2) + O(\sqrt{\varepsilon}) + \left(\mathrm{op}_\varepsilon\left(c_{\varepsilon,R}(t,q,p)\theta\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right)\Pi^\pm(q)\right)\psi^\varepsilon, \psi^\varepsilon\right)_{L^2(\mathbb{R}^{d+1})}.$$

Proof. Writing $1-\theta(u) = u\,G(u)$ with $G \in \mathcal{C}^\infty(\mathbb{R})$, we have

$$1-\theta\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right) = \tfrac{1}{R\sqrt{\varepsilon}}\lambda^\pm(q,p,\tau)G\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right).$$

We now argue as in section 4, using the estimates on $\Pi^\pm$ and $\lambda^\pm(q,p,\tau)\Pi^\pm(q) = \Pi^\pm(q)\left(\tau + \frac{1}{2}|p|^2 + V(q)\right)$. The symbolic calculus of Lemma A.2 yields in $\mathcal{L}(L^2(\mathbb{R}^{d+1}))$

$$\mathrm{op}_\varepsilon\left(c_{\varepsilon,R}(t,q,p)\left(1-\theta\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right)\right)\Pi^\pm(q)\right) = O(\sqrt{\varepsilon}) + O(1/R^2) + \tfrac{1}{R\sqrt{\varepsilon}}\,\mathrm{op}_\varepsilon\left(c_{\varepsilon,R}(t,q,p)G\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right)\Pi^\pm(q)\right)\mathrm{op}_\varepsilon\left(\tau + \tfrac{1}{2}|p|^2 + V(q)\right)$$

Indeed, the derivatives of the projectors are less harmful than in section 4, since they are only divided by $\sqrt{\varepsilon}$ and not by ε. Since ψ^ε solves the equation, we obtain

$$\left(\mathrm{op}_\varepsilon\left(c_{\varepsilon,R}(t,q,p)\left(1-\theta\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right)\right)\Pi^\pm(q)\right)\psi^\varepsilon, \psi^\varepsilon\right)_{L^2(\mathbb{R}^{d+1})} = O(1/R^2) + O(\sqrt{\varepsilon}).$$

□

By Lemma 5.1, we introduce the energy cut-off on both sides of equality (2.4) adding an error of order $O(1/R^2) + O(\sqrt{\varepsilon})$. Then, the left hand side reads

$$\operatorname{tr}\int_{\mathbb{R}^{2d+1}} \chi(t)\,W^\varepsilon(\psi^\varepsilon(t))(q,p)a^\pm_{LZ}(q,p)\,\Pi^\pm(q)\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t = O(1/R^2) + O(\sqrt{\varepsilon}) + \left(\mathrm{op}_\varepsilon\left(a^\pm_{LZ}(q,p)\chi(t)\,\theta\left(\tfrac{\lambda^\pm(q,p,\tau)}{R\sqrt{\varepsilon}}\right)\Pi^\pm(q)\right)\psi^\varepsilon\,,\,\psi^\varepsilon\right)_{L^2(\mathbb{R}^{d+1})}.$$

Using the notation

$$f_a^\pm(q,p,j) = \mathbf{1}_{\{j=\pm1\}}(j)\,a^j(q,p), \qquad (q,p,j) \in \mathbb{R}^{2d}_\pm$$

for a V-diagonal matrix-valued symbol a, the action of the semigroup on $a^\pm_{LZ}$ can be written as

$$(\mathcal{L}^{\delta_t^\pm}_{\varepsilon,R}a^\pm_{LZ})(q,p)\,\Pi^\pm(q) = (\mathcal{L}^{\delta_t^\pm}_{\varepsilon,R}f^\pm_{a_{LZ}})(q,p,+)\,\Pi^+(q).$$

Then, the right hand side of (2.4) is

$$\operatorname{tr}\int_{\mathbb{R}^{2d+1}}\chi(t+\delta_t^{\pm})\,W^\varepsilon(\psi^\varepsilon(t))(q,p)\,(\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}a_{LZ}^{\pm})(q,p)\,\Pi^+(q)\,\mathrm{d}q\,\mathrm{d}p\,\mathrm{d}t$$
$$=\Big(\operatorname{op}_\varepsilon\Big(\chi(t+\delta_t)\,\theta\Big(\tfrac{\lambda^+(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,(\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}f_{a_{LZ}}^{\pm})(q,p,+)\,\Pi^+(q)\Big)\,\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}$$
$$+O\left(1/R^2\right)+O(\sqrt{\varepsilon}),$$

and the proof of Proposition 2.4 reduces to showing that

$$\Big(\operatorname{op}_\varepsilon\Big(a_{LZ}^{\pm}(q,p)\chi(t)\theta\Big(\tfrac{\lambda^{\pm}(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,\Pi^{\pm}(q)\Big)\,\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}=$$
$$\Big(\operatorname{op}_\varepsilon\Big(\chi(t+\delta_t^{\pm})\theta\Big(\tfrac{\lambda^+(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,(\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}f_{a_{LZ}}^{\pm})(q,p,+)\,\Pi^+(q)\Big)\,\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}$$
$$+O(1/R^2)+O(R^3\sqrt{\varepsilon})+O(\sqrt{\varepsilon}|\ln\varepsilon|)+O(1/(R^5\sqrt{\varepsilon})). \tag{5.1}$$

For points $(q,t,p,\tau,j)\in\mathbb{R}^{2d+2}\times\{\pm1\}$, we consider random trajectories

$$\mathcal{T}_{\varepsilon,R}^{(q,t,p,\tau,j)}:[0,+\infty)\to\left(\mathbb{R}^{2d+2}\times\{\pm1\}\right)$$

with $\mathcal{T}_{\varepsilon,R}^{(q,t,p,\tau,j)}(r)=\left(q^j(r),r+t,p^j(r),\tau,j\right)$ as long as $(q^j(r),p^j(r))\notin S_{\varepsilon,R}$ and a jump from j to $-j$ with probability $T^\varepsilon(q^*,p^*)$, whenever $(q^j(r),p^j(r))$ hits $S_{\varepsilon,R}$ at a point (q^*,p^*). We keep the notation $(\mathcal{L}_{\varepsilon,R}^r)_{r\geq0}$ for the associated semigroup and set

$$c_{\varepsilon,R}^{\pm}(q,t,p,\tau)=a_{LZ}^{\pm}(q,p)\,\chi(t)\,\theta\Big(\tfrac{\lambda^{\pm}(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,, \tag{5.2}$$

Since $r\mapsto\lambda^{\pm}(q^{\pm}(r),p^{\pm}(r),\tau)$ is a constant function, and since within $[t-t_{\mathrm{i}},t-t_{\mathrm{f}}]$ all involved random trajectories perform a jump, we have

$$(\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}f_{c_{\varepsilon,R}}^{+})(q,t,p,\tau,+)=\chi(t+\delta_t)\,\theta\Big(\tfrac{\lambda^+(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,(\mathcal{L}_{\varepsilon,R}^{\delta_t}f_{a_{LZ}}^{+})(q,p,+),$$
$$(\mathcal{L}_{\varepsilon,R}^{\delta_t}f_{c_{\varepsilon,R}}^{-})(q,t,p,\tau,+)=\chi(t+\delta_t)\,\theta\Big(\tfrac{\lambda^+(q,p,\tau)}{R\sqrt{\varepsilon}}\Big)\,(\mathcal{L}_{\varepsilon,R}^{\delta_t}f_{a_{LZ}}^{-})(q,p,+).$$

With this notation, equation (5.1) and consequently Proposition 2.4 are equivalent to

$$\Big(\operatorname{op}_\varepsilon\Big(c_{\varepsilon,R}^{\pm}(q,t,p,\tau)\Pi^{\pm}(q)\Big)\,\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}=$$
$$\Big(\operatorname{op}_\varepsilon\Big((\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}f_{c_{\varepsilon,R}}^{\pm})(q,t,p,\tau,+)\,\Pi^+(q)\Big)\,\psi^\varepsilon\,,\,\psi^\varepsilon\Big)_{L^2(\mathbb{R}^{d+1})}$$
$$+O(1/R^2)+O(R^3\sqrt{\varepsilon})+O(\sqrt{\varepsilon}|\ln\varepsilon|)+O(1/(R^5\sqrt{\varepsilon})). \tag{5.3}$$

We emphasize, that the symbols $c_{\varepsilon,R}^{\pm}$ and $\mathcal{L}_{\varepsilon,R}^{\delta_t^{\pm}}f_{c_{\varepsilon,R}}^{\pm}$ are compactly supported inside the annulus $\{\frac{R}{2}\sqrt{\varepsilon}<|\phi|<R\sqrt{\varepsilon}\}$ at a distance of order $R\sqrt{\varepsilon}$ of $J^{\pm,out}$ where

$$J^{\pm,in/out}=\Big\{(q,t,p,\tau)\in\mathbb{R}^{2d+2}\mid(q,p)\in M^{\pm,in/out},\,\tau+v(q)+\tfrac12|p|^2\pm|\phi(q)|=0\Big\}$$

denote the submanifolds, which consist of all Hamiltonian trajectories entering (respectively leaving) the crossing set.

5.2. The normal form. Let us first recall some basic facts about canonical transforms and Fourier integral operators. The phase space $T^*\mathbb{R}^{d+1} = \mathbb{R}^{d+1} \times \mathbb{R}^{d+1}$ is a symplectic space, once endowed with the symplectic form $\omega = \mathrm{d}\tau \wedge \mathrm{d}t + \mathrm{d}p \wedge \mathrm{d}q$. A canonical transform $\kappa : T^*\mathbb{R}^{d+1} \to T^*\mathbb{R}^{d+1}$ is a change of coordinates, which preserves the symplectic form. To a canonical transform κ, one associates a unitary operator $U : L^2(\mathbb{R}^{d+1}) \to L^2(\mathbb{R}^{d+1})$ such that for all $a \in \mathcal{C}_c^\infty(\mathbb{R}^{2d+2}, \mathbb{C}^{N(\ell)\times N(\ell)})$

$$U^* \mathrm{op}_\varepsilon\,(a)\, U = \mathrm{op}_\varepsilon(a \circ \kappa) + O(\varepsilon^2)$$

as bounded operators on $L^2(\mathbb{R}^{d+1})$, see for example section 2.2 in [7]. The operator U is a Fourier integral operator. The last equality extends to symbols of the form

$$b_{\varepsilon,R}(q,t,p,\tau) = b\Big(q,t,p,\tau, \tfrac{f(q,p)}{R\sqrt{\varepsilon}}\Big)$$

with $b \in \mathcal{C}_c^\infty(\mathbb{R}^{2d+3}, \mathbb{C}^{N(\ell)\times N(\ell)})$ and $f \in \mathcal{C}^\infty(\mathbb{R}^{2d}, \mathbb{R})$ according to

$$U^* \mathrm{op}_\varepsilon\,(b_{\varepsilon,R})\, U = \mathrm{op}_\varepsilon(b_{\varepsilon,R} \circ \kappa) + O(\sqrt{\varepsilon}). \tag{5.4}$$

The proof of this statement follows the proof of Lemma 2 in [7]: one uses symbolic calculus for the commutator of a usual semi-classical pseudodifferential operator and a two-scale one of the form $\mathrm{op}_\varepsilon(b_{\varepsilon,R})$, hence the gain of $\sqrt{\varepsilon}$.

We shall crucially use the following microlocal normal form result, which for codimension two and three crossings is proven in [1, 2], however, without the explicit equations (5.6)–(5.9). These equations including the normal form for codimension five crossings are provided in Theorem 1 and Proposition 4 of [6].

Theorem 5.2 ([6]). *We consider $\rho_0 = (q_0, t_0, p_0, \tau_0 = -v(q_0) - \frac{1}{2}|p_0|^2)$ such that $\phi(q_0) = 0$, $\mathrm{d}\phi(q_0)p_0 \neq 0$, and $\mathrm{d}\phi$ is of maximal rank near q_0. Then, there exists a local canonical transform κ from a neighborhood of ρ_0 into some neighborhood Ω of 0,*

$$\kappa : (q,t,p,\tau) \mapsto (z,s,\zeta,\sigma), \quad \kappa(\rho_0) = 0.$$

There exist a Fourier integral operator U associated with κ^{-1} and an invertible matrix-valued symbol $A_\varepsilon = A_0 + \varepsilon A_1 + \varepsilon^2 A_2 + \dots$ such that $v^\varepsilon = U^* \mathrm{op}_\varepsilon(A_\varepsilon)^{-1}\psi^\varepsilon$ satisfies for all $\varphi \in \mathcal{C}_c^\infty(\Omega, \mathbb{R})$

$$\mathrm{op}_\varepsilon(\varphi)\, \mathrm{op}_\varepsilon\left(-\sigma + V_\ell\left(s, \tilde{z} + \gamma_\varepsilon(z,\zeta)\right)\right) v^\varepsilon = O(\varepsilon^\infty) \tag{5.5}$$

in $L^2(\mathbb{R}^{d+1})$, where $z = (\tilde{z}, z') \in \mathbb{R}^d$ with $\tilde{z} \in \mathbb{R}^{\ell-1}$ and $\gamma_\varepsilon = \gamma_\varepsilon(z,\zeta)$ is a vector-valued symbol $\gamma_\varepsilon = \gamma_0 + \varepsilon\gamma_1 + \varepsilon^2\gamma_2 + \ldots \in \mathbb{R}^{\ell-1}$ with

$$\gamma_\varepsilon = 0 \;\text{ for }\; \ell = 2, \qquad \gamma_\varepsilon = O(|\tilde{z}|^2) \;\text{ for }\; \ell > 2.$$

$\tilde{z} \in \mathbb{R}^{\ell-1}$ contains the coordinates of the vector $|\mathrm{d}\phi(q)p|^{-1/2}\pi_\ell(q,p)\phi(q)$ in an orthonormal basis of the hyperplane normal to $\mathrm{d}\phi(q)p$ up to $O(|\phi(q)|^2)$, while

$$\begin{aligned} s &= -|\mathrm{d}\phi(q)p|^{-1/2}\, \tfrac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|} \cdot \phi(q) + O(s^2 + \sigma^2 + |\tilde{z}|^2), \\ \sigma &= |\mathrm{d}\phi(q)p|^{-1/2} \left(\tau + \tfrac{1}{2}|p|^2 + v(q)\right) + O(s^2 + \sigma^2 + |\tilde{z}|^2). \end{aligned} \tag{5.6}$$

Moreover,

$$J^{\pm,in} = \{\sigma \mp s = 0,\ \tilde{z} = 0,\ s \leq 0\}, \quad J^{\pm,out} = \{\sigma \pm s = 0,\ \tilde{z} = 0,\ s \geq 0\}, \tag{5.7}$$

and there exists $\gamma \in \{-1,+1\}$ *such that for all* $\rho = (q,t,p,\tau)$ *with* $\kappa(\rho) = (z,s,\zeta,\sigma)$

$$A_0^*(\rho)\left(\tau + \tfrac{1}{2}|p|^2 + V(q)\right)A_0(\rho) = \gamma\left(-\sigma + V_\ell(s, \tilde{z} + \gamma_0(z,\zeta))\right). \tag{5.8}$$

$$A_0^*(\rho)V_\ell\left(\tfrac{\pi_\ell(q,p)\phi(q)}{|\pi_\ell(q,p)\phi(q)|}\right)A_0(\rho) = \gamma\, V_\ell\left(0, \tfrac{\tilde{z}}{|\tilde{z}|}\right) + O(\sqrt{\sigma^2 + s^2 + |\tilde{z}|^2}). \tag{5.9}$$

We denote by

$$\widetilde{\Pi}^{\pm}(z,s,\zeta) = \tfrac{1}{2}\left(\mathrm{Id} \mp \tfrac{1}{\sqrt{s^2 + |\tilde{z}+\gamma_0(z,\zeta)|^2}} V_\ell(s, \tilde{z} + \gamma_0(z,\zeta))\right),$$
$$\tilde{\lambda}^{\pm}(z,s,\zeta,\sigma) = -\sigma \mp \sqrt{s^2 + |\tilde{z} + \gamma_0(z,\zeta)|^2},$$

the spectral projectors and the eigenvalues of $-\sigma + V_\ell(s, \tilde{z} + \gamma_0(z,\zeta))$. Due to the relation $J^{\pm,in/out} \subseteq \{-\sigma \mp \sqrt{s^2 + |\tilde{z}|^2} = 0, \tilde{z} = 0\}$, the labelling of $\widetilde{\Pi}^{\pm}$ coincides on $J^{\pm,in/out}$ with the one for $\Pi^{\pm}$.

Proposition 5.3. *There exists functions* $k^{\pm}$ *such that if* $\kappa(q,t,p,\tau) = (z,s,\zeta,\sigma)$, *the projectors* $\Pi^{\pm}$ *and* $\widetilde{\Pi}^{\pm}$ *are related by*

$$\widetilde{\Pi}^{\pm}(z,s,\zeta,\sigma) = \left(k^{\pm}A_0^*\Pi^{\pm}A_0\right)(q,t,p,\tau) \quad on \quad \Sigma^{\mp} = \{\lambda^{\mp} = 0\}. \tag{5.10}$$

If $S = \{\phi(q) = 0,\ \tau + \frac{1}{2}|p|^2 + v(q) = 0\}$, *then* $k^+_{|S} = k^-_{|S} = e \neq 0$ *and* $(e\,A_0^*A_0)_{|S} = \mathrm{Id}_{|S}$. *Moreover, on* $\Sigma^{\pm} \cap \{0 < |\phi| \leq R\sqrt{\varepsilon}\}$,

$$\widetilde{\Pi}^{\pm}(z,s,\zeta,\sigma) = e(A_0^*\Pi^{\pm}A_0)(q,t,p,\tau) + O(R\sqrt{\varepsilon}). \tag{5.11}$$

Proof. For convenience, we set

$$P = \tau + \tfrac{1}{2}|p|^2 + V(q), \quad \widetilde{P} = -\sigma + V_\ell(s, \tilde{z} + \gamma_0(z,\zeta)).$$

By (5.8), the use of determinants gives $\Sigma^+ \cup \Sigma^- = \kappa^{-1}(\{\tilde{\lambda}^+ = 0\} \cup \{\tilde{\lambda}^- = 0\})$. Considering the equations of $J^{\pm,in/out}$, the only possibility is

$$\Sigma^{\pm} = \kappa^{-1}\left(\{\tilde{\lambda}^{\pm} = 0\}\right).$$

Therefore, for $\rho \in \Sigma^+$ we have

$$\gamma\widetilde{P}(\kappa(\rho)) = \gamma(\tilde{\lambda}^-\widetilde{\Pi}^-)(\kappa(\rho)) = (A_0^*PA_0)(\rho) = (\lambda^-A_0^*\Pi^-A_0)(\rho).$$

The same argument for Σ^- gives (5.10).

The fact $k^+_{|S} = k^-_{|S}$ comes from the precise analysis of the Hamiltonian vector fields associated with the eigenvalues $\lambda^{\pm}$. Let $\rho \in S$. We find in [6], section 5, that if

$$H(\rho) = \lim_{\alpha \to 0^{\mp}} H_{\lambda^{\pm}}(\Phi^{\alpha}_{\pm}(q,p)), \quad H'(\rho) = \lim_{\alpha \to 0^{\pm}} H_{\lambda^{\pm}}(\Phi^{\alpha}_{\pm}(q,p)),$$

then there exists a non zero function e such that

$$H = e(\partial_s + \partial_\sigma), \quad H' = e(\partial_s - \partial_\sigma) \;\text{ on }\; S.$$

Since $\lambda^\pm = 0$ and $\tilde\lambda^\pm = 0$ on S, we have $k^+_{|S} = k^-_{|S} = e$. Next, we consider the limit of the projectors $\Pi^\pm$ along outgoing trajectories,

$$\Pi^\mp_S(\rho) = \lim_{\alpha\to 0^-} \Pi^\mp(\Phi^\alpha_\pm(q,p)) = \tfrac{1}{2}\left(\mathrm{Id} \mp V_\ell\left(\tfrac{\mathrm{d}\phi(q)p}{|\mathrm{d}\phi(q)p|}\right)\right).$$

Then, equation (5.10) gives on S

$$eA_0^* A_0 = eA_0^*\left(\Pi^+_S + \Pi^-_S\right)A_0 = \left(\widetilde\Pi^+ + \widetilde\Pi^-\right)\circ\kappa = \mathrm{Id}.$$

Finally, let $\rho \in \Sigma^+$. By relation (5.8), we have for any vector $w \in \mathbb{C}^{N(\ell)}$ that $w \in \mathrm{Ker}\,\widetilde P(\kappa(\rho))$ if and only if $A_0(\rho)w \in \mathrm{Ker}\,P(\rho)$. Moreover, $\mathrm{Ker}\,P(\rho) = \mathrm{Ran}\,\Pi^+(\rho)$ and $\mathrm{Ker}\,\widetilde P(\kappa(\rho)) = \mathrm{Ran}\,\widetilde\Pi^+(\kappa(\rho))$. We therefore obtain

$$\mathrm{Ran}\,\widetilde\Pi^+(\kappa(\rho)) = \mathrm{Ran}\,(A_0^{-1}\Pi^+A_0)(\rho).$$

Since $\sqrt{e}A_0$ is unitary on S, we have

$$(A_0^{-1}\Pi^+A_0)^*(\rho) = (A_0^{-1}\Pi^+A_0)(\rho) + O(R\sqrt\varepsilon)$$

for $\rho \in \Sigma^+ \cap \{|\phi| \le R\sqrt\varepsilon\}$. Since the two projectors $\widetilde\Pi^+(\kappa(\rho))$ and $(A_0^{-1}\Pi^+A_0)(\rho)$ have the same range, while one of them is orthogonal and the other orthogonal up to $O(R\sqrt\varepsilon)$, they coincide up to $O(R\sqrt\varepsilon)$. The same argument holds for $\rho\in\Sigma^-$, and we have proven relation (5.11). □

As the next step towards proving the claimed identity (5.3), we perform the canonical change of coordinates for arriving at the microlocal normal form.

Proposition 5.4. *Let $c^\pm_{\varepsilon,R} \in \mathcal{C}^\infty_c(\mathbb{R}^{2d+2},\mathbb{C})$ be the functions defined in (5.2) and $v^\varepsilon = U^*\mathrm{op}_\varepsilon(A_\varepsilon)^{-1}\psi^\varepsilon$. Denote*

$$\widetilde T^\varepsilon(\tilde z) = \exp(-\tfrac{\pi}{\varepsilon}|\tilde z|^2). \tag{5.12}$$

Then, there exist functions $b^\pm \in \mathcal{C}^\infty_c(\mathbb{R}^{2d+2},\mathbb{C})$ and $s_1^\pm \in \mathbb{R}$, such that $b^\pm(z,s,\zeta,\eta)$ and $b^\pm(z,s_1^\pm+s,\zeta,\eta)$ are compactly supported in $\{s>0\}$ and $\{s<0\}$, respectively, and satisfy

$$\left(\mathrm{op}_\varepsilon\left(c^+_{\varepsilon,R}(q,t,p,\tau)\Pi^+(q)\right)\psi^\varepsilon(q,t),\psi^\varepsilon(q,t)\right)_{L^2} = \left(\mathrm{op}_\varepsilon\left(b^+\left(z,s,\zeta,\tfrac{\tilde z}{R\sqrt\varepsilon}\right)\right)v_2^\varepsilon(z,s),v_2^\varepsilon(z,s)\right)_{L^2} + O(R\sqrt\varepsilon).$$

$$\left(\mathrm{op}_\varepsilon\left((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})(q,t,p,\tau,+)\Pi^+(q)\right)\psi^\varepsilon(q,t),\psi^\varepsilon(q,t)\right)_{L^2} = \left(\mathrm{op}_\varepsilon\left((1-\widetilde T^\varepsilon(\tilde z))b^+\left(z,s+s_1^+,\zeta,\tfrac{\tilde z}{R\sqrt\varepsilon}\right)\right)v_1^\varepsilon(z,s),v_1^\varepsilon(z,s)\right)_{L^2} + O(R^3\sqrt\varepsilon),$$

$$\left(\mathrm{op}_\varepsilon\left(c^-_{\varepsilon,R}(q,t,p,\tau)\Pi^-(q)\right)\psi^\varepsilon(q,t),\psi^\varepsilon(q,t)\right)_{L^2} = \left(\mathrm{op}_\varepsilon\left(b^-\left(z,s,\zeta,\tfrac{\tilde z}{R\sqrt\varepsilon}\right)\right)v_1^\varepsilon(z,s),v_1^\varepsilon(z,s)\right)_{L^2} + O(R\sqrt\varepsilon),$$

$$\left(\mathrm{op}_\varepsilon\left((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^-_{c_{\varepsilon,R}})(q,t,p,\tau,+)\Pi^+(q)\right)\psi^\varepsilon(q,t),\psi^\varepsilon(q,t)\right)_{L^2} = \left(\mathrm{op}_\varepsilon\left(\widetilde T^\varepsilon(\tilde z)\,b^-\left(z,s+s_1^-,\zeta,\tfrac{\tilde z}{R\sqrt\varepsilon}\right)\right)v_1^\varepsilon(z,s),v_1^\varepsilon(z,s)\right)_{L^2} + O(R^3\sqrt\varepsilon).$$

Proof. We only prove the first two equalities, since one deals similarly with the other two. By symbolic calculus and the transformation property (5.4), the canonical transform κ^{-1} of Theorem 5.2 acts as

$$\left(\mathrm{op}_\varepsilon\left(c^+_{\varepsilon,R}(q,t,p,\tau)\Pi^+(q)\right)\psi^\varepsilon(q,t),\psi^\varepsilon(q,t)\right)_{L^2} = \left(\mathrm{op}_\varepsilon\left(\left((c^+_{\varepsilon,R}A_0^*\Pi^+A_0)\circ\kappa^{-1}\right)(z,s,\zeta,\sigma)\right)v^\varepsilon(z,s)\ ,\ v^\varepsilon(z,s)\right)_{L^2} + O(\sqrt{\varepsilon}).$$

The compactly supported function $c^+_{\varepsilon,R}\circ\kappa^{-1}$ is localized near $J^{+,out}$, that is near $\{\sigma+s=0,\ \tilde z=0,\ s>0\}$. The relation (5.11) between the projectors gives a function $b\in\mathcal{C}_c^\infty(\mathbb{R}^{2d+2+\ell},\mathbb{C})$ compactly supported in $\{s>0\}$ such that

$$\begin{aligned}\left((c^+_{\varepsilon,R}A_0^*\Pi^+A_0)\circ\kappa^{-1}\right)&(z,s,\zeta,\sigma)\\ &= b\left(z,s,\zeta,\sigma,\tfrac{\tilde z}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z,s,\zeta,\sigma)}{R\sqrt{\varepsilon}}\right)\widetilde\Pi^+(z,s,\zeta)+O(R\sqrt{\varepsilon})\\ &=: b_{\varepsilon,R}(z,s,\zeta,\sigma)\widetilde\Pi^+(z,s,\zeta)+O(R\sqrt{\varepsilon})\end{aligned}$$

as functions in $\mathcal{C}_c^\infty(\mathbb{R}^{2d+2},\mathbb{C})$. Hence, we obtain

$$\begin{aligned}&\left(\mathrm{op}_\varepsilon\left(c^+_{\varepsilon,R}(q,t,p,\tau)\Pi^+(q)\right)\psi^\varepsilon(q,t)\ ,\ \psi^\varepsilon(q,t)\right)_{L^2}\\ &\qquad= \left(\mathrm{op}_\varepsilon\left(b_{\varepsilon,R}(z,s,\zeta,\sigma)\widetilde\Pi^+(z,s,\zeta)\right)v^\varepsilon(z,s)\ ,\ v^\varepsilon(z,s)\right)_{L^2}+O(R\sqrt{\varepsilon}).\end{aligned}$$

For $|\tilde z| = O(R\sqrt{\varepsilon})$ we have

$$\widetilde\Pi^+(z,s,\zeta) = \begin{pmatrix}0 & 0\\ 0 & \mathrm{Id}\end{pmatrix} + O(R\sqrt{\varepsilon})\ \text{ in }\ \{s>0\},$$

$$\widetilde\Pi^+(z,s,\zeta) = \begin{pmatrix}\mathrm{Id} & 0\\ 0 & 0\end{pmatrix} + O(R\sqrt{\varepsilon})\ \text{ in }\ \{s<0\}, \tag{5.13}$$

and therefore

$$\begin{aligned}&\left(\mathrm{op}_\varepsilon\left(b_{\varepsilon,R}(z,s,\zeta,\sigma)\widetilde\Pi^+(z,s,\zeta)\right)v^\varepsilon(z,s)\,,\,v^\varepsilon(z,s)\right)_{L^2}\\ &\qquad= \left(\mathrm{op}_\varepsilon\left(b_{\varepsilon,R}(z,s,\zeta,\sigma)\right)v_2^\varepsilon(z,s)\,,\,v_2^\varepsilon(z,s)\right)_{L^2}+O(R\sqrt{\varepsilon}).\end{aligned}$$

We now remove the σ-dependence of the symbol. Taylor expanding around $\sigma = -\sqrt{s^2+|\tilde z+\gamma_0(z,\zeta)|^2}$, we write

$$\begin{aligned}b_{\varepsilon,R}(z,s,\zeta,\sigma) &= b\left(z,s,\zeta,-\sqrt{s^2+|\tilde z+\gamma_0(z,\zeta)|^2},\tfrac{\tilde z}{R\sqrt{\varepsilon}},0\right)\\ &\quad+\tfrac{1}{R\sqrt{\varepsilon}}\tilde\lambda^+(s,z,\sigma,\zeta)\,G\left(z,s,\zeta,\sigma,\tfrac{\tilde z}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z,s,\sigma,\zeta)}{R\sqrt{\varepsilon}}\right)\end{aligned}$$

with $G\in\mathcal{C}_c^\infty(\mathbb{R}^{2d+2+\ell},\mathbb{C})$. Since

$$\tilde\lambda^+(z,s,\zeta,\sigma)\tilde\Pi^+(z,s,\zeta) = \widetilde\Pi^+(z,s,\zeta)\left(\sigma - V_\ell(s,\tilde z+\gamma_0(z,\zeta))\right),$$

and since v^ε solves the Landau-Zener type problem (5.5), an argument analogous to the proof of Lemma 5.1 yields

$$\begin{aligned}&\left(\mathrm{op}_\varepsilon\left(b_{\varepsilon,R}(z,s,\zeta,\sigma)\right)v_2^\varepsilon(z,s)\,,\,v_2^\varepsilon(z,s)\right)_{L^2} = O(1/R^2)+O(\sqrt{\varepsilon})\\ &\quad+\left(\mathrm{op}_\varepsilon\left(b\left(z,s,\zeta,-\sqrt{s^2+|\tilde z+\gamma_0(z,\zeta)|^2},\tfrac{\tilde z}{R\sqrt{\varepsilon}},0\right)\right)v_2^\varepsilon(z,s)\,,\,v_2^\varepsilon(z,s)\right)_{L^2}.\end{aligned}$$

Setting $b^+(z,s,\zeta,\eta) = b(z,s,\zeta,-s,\eta,0)$, we obtain

$$\left(\mathrm{op}_\varepsilon\left(c^+_{\varepsilon,R}(q,t,p,\tau)\Pi^+(q)\right)\psi^\varepsilon(q,t)\ ,\ \psi^\varepsilon(q,t)\right)_{L^2}$$
$$= \left(\mathrm{op}_\varepsilon\left(b^+\left(z,s,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\right)\right)v_2^\varepsilon(z,s)\ ,\ v_2^\varepsilon(z,s)\right)_{L^2} + O(R\sqrt{\varepsilon}) + O(1/R^2).$$

Next, we focus on the second claimed identity, which contains non-adiabatic transitions. We have

$$\left(\mathrm{op}_\varepsilon\left((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})(q,t,p,\tau,+)\Pi^+(q)\right)\psi^\varepsilon(q,t)\ ,\ \psi^\varepsilon(q,t)\right)_{L^2} =$$
$$\left(\mathrm{op}_\varepsilon\left(((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})A_0^*\Pi^+A_0)(\kappa^{-1}(z,s,\zeta,\sigma),+)\right)v^\varepsilon(z,s)\ ,\ v^\varepsilon(z,s)\right)_{L^2} + O(\sqrt{\varepsilon}).$$

Let $r\mapsto(z^+(r),s^+(r),\zeta^+(r),\sigma^+(r))$ be the Hamiltonian trajectory of

$$\begin{aligned}
&\dot z = \partial_\zeta\tilde\lambda^+ = {}^t\mathrm{d}_\zeta\gamma_0(z,\zeta)(\tilde z+\gamma_0(z,\zeta))\,(s^2+|\tilde z+\gamma_0(z,\zeta)|^2)^{-1/2},\\
&\dot s = \partial_\sigma\tilde\lambda^+ = 1,\\
&\dot\zeta = -\partial_z\tilde\lambda^+ = -\,{}^t\mathrm{d}_z(\tilde z+\gamma_0(z,\zeta))(\tilde z+\gamma_0(z,\zeta))(s^2+|\tilde z+\gamma_0(z,\zeta)|^2)^{-1/2},\\
&\dot\sigma = -\partial_s\tilde\lambda^+ = -s\,(s^2+|\tilde z+\gamma_0(z,\zeta)|^2)^{-1/2}.
\end{aligned}$$

with $(z(0),s(0),\zeta(0),\sigma(0)) = (z,s,\zeta,\sigma) = \kappa(q,t,p,\tau)$. A trajectory jumps for $r=r_*$, if $s^+(r_*) = O(|\phi(q)|^2) = O(R^2\varepsilon)$. Then, $\sigma^+(r_*) = O(R^2\varepsilon)$ as well. Since

$$\tfrac{\mathrm{d}}{\mathrm{d}r}\left(s^+(r)+\sigma^+(r)\right) = O(|\tilde z|^2)\quad\text{on}\quad J^{+,out} = \{\sigma+s=0,\ \tilde z=0,\ s>0\},$$

and since $|\tilde z| = O(R\sqrt{\varepsilon})$ on the support of our symbol, we have

$$\begin{aligned}
&z^+(r) = z + O(R^3\varepsilon^{3/2}),\quad s^+(r) = s+r,\\
&\zeta^+(r) = \zeta + O(R\sqrt{\varepsilon}),\quad \sigma^+(r) = -s^+(r) + O(R^2\varepsilon).
\end{aligned}$$

These asymptotics together with conservation of energy along Hamiltonian flows yield

$$b\Big(z^+(r),s^+(r),\zeta^+(r),\sigma^+(r),\tfrac{\tilde z^+(r)}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z^+(r),s^+(r),\zeta^+(r),\sigma^+(r))}{R\sqrt{\varepsilon}}\Big) =$$
$$b\Big(z,s+r,\zeta,-(s+r),\tfrac{\tilde z}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z,s,\zeta,\sigma)}{R\sqrt{\varepsilon}}\Big) + O(R\sqrt{\varepsilon}).$$

Jumps occur for $|\phi(q)|\le R\sqrt{\varepsilon}$, that is for

$$\begin{aligned}
|\tilde z|^2 &= |\mathrm{d}\phi(q)p|^{-1}|\pi_\ell(q,p)\phi(q)|^2 + O(|\phi(q)|^3)\\
&= |\mathrm{d}\phi(q)p|^{-1}|\pi_\ell(q,p)\phi(q)|^2 + O(R^3\varepsilon^{3/2}).
\end{aligned}$$

Therefore, the transition rate $T^\varepsilon(q,p)$ reads in the new coordinates as

$$\begin{aligned}
T^\varepsilon(q,p) &= \exp\big(-\tfrac{\pi}{\varepsilon}|\mathrm{d}\phi(q)p|^{-1}|\pi_\ell(q,p)\phi(q)|^2\big)\\
&= \exp\big(-\tfrac{\pi}{\varepsilon}|\tilde z|^2\big) + O(R^3\sqrt{\varepsilon}) = \widetilde T^\varepsilon(\tilde z) + O(R^3\sqrt{\varepsilon}),
\end{aligned}$$

and there exists $s_1^+\in\mathbb{R}$ such that

$$\begin{aligned}
&\big((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})A_0^*\Pi^+A_0\big)(\kappa^{-1}(z,s,\zeta,\sigma),+)\\
&= \Big(1-\widetilde T^\varepsilon(\tilde z)\Big)\,b\Big(z,s+s_1^+,\zeta,-(s+s_1^\pm),\tfrac{\tilde z}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z,s,\zeta,\sigma)}{R\sqrt{\varepsilon}}\Big)\,\widetilde\Pi^+(z,s,\zeta) + O(R^3\sqrt{\varepsilon}).
\end{aligned}$$

By the asymptotics (5.13) of $\widetilde{\Pi}^+$ above $\{s<0\}$, we then get

$$\Big(\mathrm{op}_\varepsilon\Big(((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})A_0^*\Pi^+A_0)(\kappa^{-1}(z,s,\zeta,\sigma),+)\Big)\,v^\varepsilon(z,s)\;,\;v^\varepsilon(z,s)\Big)_{L^2}=O(R^3\sqrt{\varepsilon})$$
$$+\Big(\mathrm{op}_\varepsilon\Big((1-\widetilde{T}^\varepsilon(\tilde z))b\Big(z,s+s_1^+,\zeta,-(s+s_1^\pm),\tfrac{\tilde z}{R\sqrt{\varepsilon}},\tfrac{\tilde\lambda^+(z,s,\zeta,\sigma)}{R\sqrt{\varepsilon}}\Big)\Big)\,v_1^\varepsilon(z,s),v_1^\varepsilon(z,s)\Big)_{L^2}.$$

As before, we remove the σ-dependence of the symbol and obtain

$$\Big(\mathrm{op}_\varepsilon\Big((\mathcal{L}^{\delta_t}_{\varepsilon,R}f^+_{c_{\varepsilon,R}})(q,t,p,\tau,+)\Pi^+(q)\Big)\,\psi^\varepsilon(q,t)\;,\;\psi^\varepsilon(q,t)\Big)_{L^2}$$
$$=\Big(\mathrm{op}_\varepsilon\Big((1-\widetilde{T}^\varepsilon(\tilde z))b^+\Big(z,s+s_1^+,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,v_1^\varepsilon(z,s)\;,\;v_1^\varepsilon(z,s)\Big)_{L^2}+O(R^3\sqrt{\varepsilon}).$$

□

5.3. The transitions. It remains to analyze $v^\varepsilon=U^*\mathrm{op}_\varepsilon(A_\varepsilon^{-1})\psi^\varepsilon$ for proving the equality of each pair in Proposition 5.4 up to an error of $O(1/R^2)+O(R^3\sqrt{\varepsilon})+O(\sqrt{\varepsilon}|\ln\varepsilon|)+O(1/(R^5\sqrt{\varepsilon}))$, concluding the proof of our main result. We consider the solution u^ε of

$$-i\varepsilon\partial_s u^\varepsilon=\begin{pmatrix} s\,\mathrm{Id} & \sqrt{\varepsilon}\,G\\ \sqrt{\varepsilon}G^* & -s\,\mathrm{Id}\end{pmatrix}u^\varepsilon,\qquad u^\varepsilon_{|s=0}=v^\varepsilon_{|s=0},\tag{5.14}$$

where G is one of the operators

$$G_2=\tfrac{1}{\sqrt{\varepsilon}}\varphi(\tfrac{\tilde z}{R\sqrt{\varepsilon}})\tilde z,\qquad G_3=\tfrac{1}{\sqrt{\varepsilon}}\varphi(\tfrac{\tilde z}{R\sqrt{\varepsilon}})(Z_1+iZ_2),$$
$$G_5=\tfrac{1}{\sqrt{\varepsilon}}\varphi(\tfrac{\tilde z}{R\sqrt{\varepsilon}})\begin{pmatrix} Z_1+iZ_2 & Z_3+iZ_4\\ -Z_3+iZ_4 & Z_1-iZ_2\end{pmatrix}$$

with $Z=\mathrm{op}_\varepsilon(\tilde z+\gamma_\varepsilon(z,\zeta))$ and $\varphi\in\mathcal{C}_c^\infty(\mathbb{R}^{\ell-1},\mathbb{R})$. In all three cases, G is a bounded operator on $L^2(\mathbb{R}^d)$ with $\|G\|=O(R)$, and we have

$$\|u^\varepsilon-v^\varepsilon\|_{L^2_{loc}(\mathbb{R}^{d+1})}=O(\varepsilon^\infty).$$

The following Landau-Zener type formula is given in Proposition 7 of [8], up to the explicit error terms.

Proposition 5.5. *Let u^ε be the solution of equation (5.14). There exist vector-valued functions $\alpha^\varepsilon=(\alpha_1^\varepsilon,\alpha_2^\varepsilon)$, $\omega^\varepsilon=(\omega_1^\varepsilon,\omega_2^\varepsilon)\in L^2(\mathbb{R}^d,\mathbb{C}^{N(\ell)})$ such that for any function $\chi\in\mathcal{C}_c^\infty(\{x\in\mathbb{R}\mid |x|\le R^2\},\mathbb{R})$ the families $(\chi(GG^*)\alpha_1^\varepsilon)_{\varepsilon>0}$, $(\chi(G^*G)\alpha_2^\varepsilon)_{\varepsilon>0}$, $(\chi(GG^*)\omega_1^\varepsilon)_{\varepsilon>0}$, $(\chi(G^*G)\omega_2^\varepsilon)_{\varepsilon>0}$ are bounded in $L^2(\mathbb{R}^d,\mathbb{C})$ and satisfy*

$$\chi(GG^*)u_1^\varepsilon(z,s)=\chi(GG^*)\mathrm{e}^{is^2/(2\varepsilon)}\left|\tfrac{s}{\sqrt{\varepsilon}}\right|^{i\frac{GG^*}{2}}k_1^\varepsilon(z)+O(R^2\sqrt{\varepsilon}),$$
$$\chi(G^*G)u_2^\varepsilon(z,s)=\chi(G^*G)\mathrm{e}^{-is^2/(2\varepsilon)}\left|\tfrac{s}{\sqrt{\varepsilon}}\right|^{-i\frac{G^*G}{2}}k_2^\varepsilon(z)+O(R^2\sqrt{\varepsilon})$$

in $L^2(\mathbb{R}^d,\mathbb{C})$, where $k_j^\varepsilon=\alpha_j^\varepsilon$ and $k_j^\varepsilon=\omega_j^\varepsilon$ for $s<0$ and $s>0$, respectively, $j\in\{1,2\}$. Moreover,

$$\begin{pmatrix}\omega_1^\varepsilon\\ \omega_2^\varepsilon\end{pmatrix}=\begin{pmatrix} a(GG^*) & -\bar b(GG^*)G\\ b(G^*G)G^* & a(G^*G)\end{pmatrix}\begin{pmatrix}\alpha_1^\varepsilon\\ \alpha_2^\varepsilon\end{pmatrix}\tag{5.15}$$

with

$$a(\lambda)=\mathrm{e}^{-\pi\lambda/2},\qquad b(\lambda)=\frac{2i\mathrm{e}^{i\pi/4}}{\lambda\sqrt{\pi}}2^{-i\lambda/2}\mathrm{e}^{-\pi\lambda/4}\Gamma(1+i\tfrac{\lambda}{2})\sinh(\tfrac{\pi\lambda}{2}).$$

Proof. Lemma 7 in [8] is the crucial step in the proof of Proposition 7 for which we have to check that the leading order error estimate is indeed $O(R^2\sqrt{\varepsilon})$. For this, we turn to the explicit calculations in the proof of Lemma 11 in [7] and study the two integrals

$$A_0 = s^{-1+i\eta^2/2}\mathrm{e}^{-is^2/2}\int_{\mathbb{R}}\tilde\chi\Big(\sqrt{2}-\sqrt{2-\tfrac{2z}{s^2}}\Big)\,\Big|\sqrt{2}-\sqrt{2-\tfrac{2z}{s^2}}\Big|^{i\eta^2/2}\,\frac{\mathrm{e}^{iz}}{\sqrt{2-\frac{2z}{s^2}}}\,\mathrm{d}z,$$

$$B_0 = s^{1+i\eta^2/2}\int_{\mathbb{R}}(1-\tilde\chi(y))\,\mathrm{e}^{-\frac{i}{2}s^2(1+y^2-2\sqrt{2}y)}|y|^{i\eta^2/2}\mathrm{d}y,$$

where $\tilde\chi\in\mathcal{C}_c^\infty(\mathbb{R},\mathbb{R})$ is a function with $0\le\tilde\chi\le 1$, $\tilde\chi(y)=0$ for $|y|\ge\sqrt{2}/2$ and $\tilde\chi(y)=1$ for $|y|\le\sqrt{2}/4$. The phase function $y\mapsto -\frac12(1+y^2-2\sqrt{2}y)$ of B_0 has the stationary point $y=\sqrt{2}$, and Taylor expansion of $y\mapsto\ln|y|$ around $y=\sqrt{2}$ yields

$$B_0=\sqrt{2\pi}\mathrm{e}^{-i\pi/4}2^{i\eta^2/4}s^{i\eta^2/2}\mathrm{e}^{is^2/2}+O(\eta^2 s^{-2}),$$

while integration by parts gives

$$A_0 = O(\eta^2 s^{-1})$$

as $\eta, s\to\infty$. Since the asymptotics of the other relevant integrals can be obtained analogously, the claimed error estimates follow by setting $s=O(\varepsilon^{-1/2})$ and $\eta=O(R)$. □

For implementing these Landau-Zener asymptotics, we need the following additional relations, which are literally contained in the proofs of Lemma 8 and Lemma 9 in [8].

Lemma 5.6. *For any $\chi\in\mathcal{C}_c^\infty(\mathbb{R},\mathbb{R})$ and $b\in\mathcal{C}_c^\infty(\mathbb{R}^{2d+N(\ell)-1},\mathbb{C})$, we have*

$$\chi\Big(\tfrac{|\tilde z|^2}{\varepsilon}\Big)=\chi(GG^*)+O(\sqrt{\varepsilon})=\chi(G^*G)+O(\sqrt{\varepsilon}),$$

$$\Big|\tfrac{s}{\sqrt{\varepsilon}}\Big|^{\pm i\frac{G^*G}{2}}\,\mathrm{op}_\varepsilon\Big(b\Big(z,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,\Big|\tfrac{s}{\sqrt{\varepsilon}}\Big|^{\mp i\frac{G^*G}{2}}=\mathrm{op}_\varepsilon\Big(b\Big(z,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)+O(\sqrt{\varepsilon}|\ln\varepsilon|),$$

Now, we are ready to conclude the proof of Theorem 2.2. We only discuss the first pair of terms in Proposition 5.4, since the other pair can be dealt with analogously. We set

$$I^1_{\varepsilon,R}=\Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z,s,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,u_2^\varepsilon(z,s),u_2^\varepsilon(z,s)\Big)_{L^2},$$
$$I^2_{\varepsilon,R}=\Big(\mathrm{op}_\varepsilon\Big((1-\widetilde{T}^\varepsilon(\tilde z))b^+\Big(z,s+s_1^+,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,u_1^\varepsilon(z,s),u_1^\varepsilon(z,s)\Big)_{L^2}$$

By Lemma 5.6 and Proposition 5.5, we have

$$\begin{aligned}
I^1_{\varepsilon,R}&=\Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z,s,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,\chi(G^*G)u_2^\varepsilon(z,s),\chi(G^*G)u_2^\varepsilon(z,s)\Big)_{L^2}+O(\sqrt{\varepsilon}),\\
&=\Bigg(\mathrm{op}_\varepsilon\Big(b^+\Big(z,s,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,\mathrm{e}^{-i\frac{s^2}{2\varepsilon}}\,\Big|\tfrac{s}{\sqrt{\varepsilon}}\Big|^{-i\frac{G^*G}{2}}\,\omega_2^\varepsilon(z),\mathrm{e}^{-i\frac{s^2}{2\varepsilon}}\,\Big|\tfrac{s}{\sqrt{\varepsilon}}\Big|^{-i\frac{G^*G}{2}}\,\omega_2^\varepsilon(z)\Bigg)_{L^2}\\
&\qquad+O(R^2\sqrt{\varepsilon})\\
&=\Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z,s,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,\omega_2^\varepsilon(z),\omega_2^\varepsilon(z)\Big)_{L^2}+O(R^2\sqrt{\varepsilon})+O(\sqrt{\varepsilon}|\ln\varepsilon|)\\
&=\Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z,s+s_1^+,\zeta,\tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,\omega_2^\varepsilon(z),\omega_2^\varepsilon(z)\Big)_{L^2}+O(R^2\sqrt{\varepsilon})+O(\sqrt{\varepsilon}|\ln\varepsilon|)
\end{aligned}$$

because $s_1^\pm = O(R\sqrt{\varepsilon})$ and analogously

$$I^2_{\varepsilon,R} = \Big(\mathrm{op}_\varepsilon\Big((1-\widetilde{T}^\varepsilon(\tilde z))\, b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, \alpha_1^\varepsilon(z), \alpha_1^\varepsilon(z)\Big)_{L^2} + O(R^2\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|).$$

By the scattering identity (5.15), we have $\omega_2^\varepsilon = b(G^*G)G^*\alpha_1^\varepsilon + a(G^*G)\alpha_2^\varepsilon$ and

$$I^1_{\varepsilon,R} = \Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, (b(G^*G)G^*\alpha_1^\varepsilon(z) + a(G^*G)\alpha_2^\varepsilon(z))\,, \; b(G^*G)G^*\alpha_1^\varepsilon(z) + a(G^*G)\alpha_2^\varepsilon(z)\Big)_{L^2} + O(R^2\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|).$$

Since the wave function $\psi^\varepsilon(q,t)$ is of order $O(1/R^2)+O(\sqrt{\varepsilon})+O(1/(R^5\sqrt{\varepsilon}))$ near the set $J^{-,in} = \{\sigma+s=0,\ \tilde z = 0,\ s<0\}$, we have

$$\begin{pmatrix} 0 \\ v_2^\varepsilon(z,s)\end{pmatrix} = \mathrm{op}_\varepsilon\Big(\widetilde{\Pi}^-(z,s,\zeta)\Big)\, v^\varepsilon(z,s) + O(R\sqrt{\varepsilon}) = O(1/R^2) + O(R\sqrt{\varepsilon}) + O(1/(R^5\sqrt{\varepsilon}))$$

as functions in $L^2_{loc}(\mathbb{R}^{d+1})$ localized near $J^{-,in}$. The preceding arguments expressing $I^1_{\varepsilon,R}$ and $I^2_{\varepsilon,R}$ in terms of α^ε and ω^ε then yield

$$a(G^*G)\alpha_2^\varepsilon(z) = O(1/R^2) + O(R^2\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|) + O(1/(R^5\sqrt{\varepsilon}))$$

near $J^{-,in}$, and hence

$$I^1_{\varepsilon,R} = \Big(\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, b(G^*G)G^*\alpha_1^\varepsilon(z), b(G^*G)G^*\alpha_1^\varepsilon(z)\Big)_{L^2} + O(1/R^2) + O(R^2\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|) + O(1/(R^5\sqrt{\varepsilon})).$$

Lemma 5.6 together with the relations $G^*b(GG^*) = b(G^*G)G^*$ and

$$\lambda|b(\lambda)|^2 = 1 - \mathrm{e}^{-\pi\lambda}$$

implies

$$\begin{aligned}
&G\,\bar b(G^*G)\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, b(G^*G)G^*\alpha_1^\varepsilon(z)\\
&= G\,\bar b(G^*G) b(G^*G)G^*\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, \alpha_1^\varepsilon(z) + O(\sqrt{\varepsilon})\\
&= GG^*\bar b(GG^*) b(GG^*)\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, \alpha_1^\varepsilon(z) + O(\sqrt{\varepsilon})\\
&= (1-\widetilde{T}^\varepsilon(\tilde z))\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, \alpha_1^\varepsilon(z) + O(\sqrt{\varepsilon}) \qquad (5.16)
\end{aligned}$$

and finally

$$I^1_{\varepsilon,R} = I^2_{\varepsilon,R} + O(1/R^2) + O(R^2\sqrt{\varepsilon}) + O(\sqrt{\varepsilon}|\ln\varepsilon|) + O(1/(R^5\sqrt{\varepsilon})).$$

6. Eigenvalues of multiplicity two. We have assumed that the matrix-valued observable a is V-diagonal in the sense that $a = a^+\Pi^+ + a^-\Pi^-$ with scalar-valued functions $a^\pm$. The more natural assumption, that a commutes with V,

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

does not change the situation in the case $\ell = 2, 3$, but enlarges the class of observables for $\ell = 3', 5$. For observables of this form, one has to modify the Markov process to account for a polarization effect. The state space requires an additional component $w \in \mathbb{C}^4$, and when the deterministic flow $\Phi_j^t(q,p)$ hits the jump manifold $S_{\varepsilon,R}$ in a point (q^*, p^*) a more general branching occurs. The state (q^*, p^*, j, w) changes with probability $T^\varepsilon(q^*, p^*)$ to $(q^*, p^*, -j, w)$ and with probability $1 - T^\varepsilon(q^*, p^*)$ to $(q^*, p^*, j, \mathcal{R}(q^*, p^*)w)$, where

$$\mathcal{R}(q,p) = V_\ell\Big(\tfrac{\pi_\ell(q,p)\phi(q)}{|\pi_\ell(q,p)\phi(q)|}\Big).$$

This phenomenon is also described in Theorem 1 of [5] for two-scale Wigner measures. Our main result Theorem 2.2 for the propagation of Wigner functions still applies for the semigroup, which encorporates polarization.

Proof. [Theorem 2.2 for $a = \Pi^+ a\Pi^+ + \Pi^- a\Pi^-$ if $\ell = 3', 5$] Let us first prove classical transport. We set $A^+ = \Pi^+ a\Pi^+$ and we focus on the $+$ mode. We extensively use $\Pi^+ A^+ = A^+\Pi^+ = A^+$. The strategy is similar to the one of section 4, and we have to focus on the Poisson brackets

$$\tfrac12\{A^+(q,p), \tau + \tfrac12|p|^2 + v(q) + V_\ell(\phi(q))\} - \tfrac12\{\tau + \tfrac12|p|^2 + v(q) + V_\ell(\phi(q)), A^+(q,p)\}.$$

We set $\mu(q,p,\tau) = \tau + \frac12|p|^2 + v(q)$ and write

$$\{A^+, \mu\} = \Pi^+\{A^+, \mu\}\Pi^+ + A^+\{\Pi^+, \mu\} + \{\Pi^+, \mu\}A^+.$$

We observe that

$$r_0 = A^+\{\Pi^+, \mu\} + \{\Pi^+, \mu\}A^+ = -A^+(\nabla_q\Pi^+\cdot p) - (\nabla_q\Pi^+\cdot p)A^+$$

can be treated as in section 4. Indeed, since A^+ commutes with $V_\ell(\phi)$, one has for any matrix G that $A^+[V_\ell(\phi), G] = [V_\ell(\phi), A^+G]$, $[V_\ell(\phi), G]A^+ = [V_\ell(\phi), GA^+]$, and consequently

$$\begin{aligned} r_0 &= -\tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), \left[V_\ell(\phi), A^+V_\ell(\mathrm{d}\phi p)\right]\right] - \tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), \left[V_\ell(\phi), V_\ell(\mathrm{d}\phi p)A^+\right]\right] \\ &= -\Big[\mu + V_\ell(\phi), \tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), A^+V_\ell(\mathrm{d}\phi p)\right]\Big] - \Big[\mu + V_\ell(\phi), \tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), V_\ell(\mathrm{d}\phi p)A^+\right]\Big]. \end{aligned}$$

The most harmful of the arising terms contains the brackets with $\frac12|p|^2$, that is

$$\tilde r_0 = -\Big\{\tfrac12|p|^2, \tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), A^+V_\ell(\mathrm{d}\phi p)\right]\Big\} - \Big\{\tfrac12|p|^2, \tfrac{1}{4|\phi|^3}\left[V_\ell(\phi), V_\ell(\mathrm{d}\phi p)A^+\right]\Big\}.$$

Since the term containing the derivatives of $V_\ell(\phi)$ vanishes,

$$-\tfrac{1}{4|\phi|^3}\left[V_\ell(\mathrm{d}\phi p), A^+V_\ell(\mathrm{d}\phi p)\right] - \tfrac{1}{4|\phi|^3}\left[V_\ell(\mathrm{d}\phi p), V_\ell(\mathrm{d}\phi p)A^+\right] = 0,$$

there is a matrix-valued function G_ε with suitable bounds on its derivatives, such that $\tilde r_0 = |\phi|^{-4}\left[V_\ell(\phi), G_\varepsilon\right]$, and hence the other arguments of Lemma 4.1 apply for the analysis of r_0.

For the brackets with the matrix part, we write

$$\tfrac12\{A^+, V_\ell(\phi)\} - \tfrac12\{V_\ell(\phi), A^+\} = \Pi^+\{A^+, |\phi|\}\Pi^+ + |\phi|\left(\{A^+, \Pi^+\} - \{\Pi^+, A^+\}\right).$$

The second part

$$r_1 = |\phi|\left(\{A^+, \Pi^+\} - \{\Pi^+, A^+\}\right) = |\phi|\left(\nabla_p A^+\cdot\nabla_q\Pi^+ + \nabla_q\Pi^+\nabla_p A^+\right)$$

is off-diagonal with respect to V, since $\nabla_p A^+ = \Pi^+\nabla_p A^+ = \nabla_p A^+\Pi^+$, $\Pi^\pm\Pi^\mp = 0$, and $\Pi^\pm\nabla_q\Pi^+\Pi^\pm = 0$ imply

$$\Pi^\pm\left(\nabla_p A^+\cdot\nabla_q\Pi^+ + \nabla_q\Pi^+\nabla_p A^+\right)\Pi^\pm = 0.$$

Hence, Lemma 4.1 applies.

The importance of $\mathcal{R}(q,p)$ for the non-adiabatic transitions becomes clear, when recasting equation (5.16) in the previous section as

$$\begin{aligned}
&\begin{pmatrix} 0 & G^* \\ G & 0\end{pmatrix}\bar{b}(G^*G)\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\, b(G^*G)\begin{pmatrix} 0 & G \\ G^* & 0\end{pmatrix}\begin{pmatrix}\alpha_1^\varepsilon(z)\\ 0\end{pmatrix}\\
&= V_\ell^*(0,\tfrac{\tilde z}{|\tilde z|})(G^*G)^{\frac12}\,\bar{b}(G^*G)\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\\
&\qquad\qquad b(G^*G)(G^*G)^{\frac12}\,V_\ell^*(0,\tfrac{\tilde z}{|\tilde z|})\begin{pmatrix}\alpha_1^\varepsilon(z)\\ 0\end{pmatrix} + O(\sqrt{\varepsilon})\\
&= (1-\widetilde{T}^\varepsilon(\tilde z))\,V_\ell^*(0,\tfrac{\tilde z}{|\tilde z|})\,\mathrm{op}_\varepsilon\Big(b^+\Big(z, s+s_1^+, \zeta, \tfrac{\tilde z}{R\sqrt{\varepsilon}}\Big)\Big)\,V_\ell(0,\tfrac{\tilde z}{|\tilde z|})\begin{pmatrix}\alpha_1^\varepsilon(z)\\ 0\end{pmatrix} + O(\sqrt{\varepsilon})
\end{aligned}$$

and observing that the normal form transformation relates $V_\ell(0,\tilde z/|\tilde z|)$ and $\mathcal{R}(q,p)$ by identity (5.9) of Theorem 5.2. □

Appendix A. Weyl calculus. For the convenience of the reader, we formulate the key technical lemma of the calculus of Weyl quantized pseudodifferential operators.

Definition A.1. *A smooth matrix-valued function $a\in\mathcal{C}^\infty(\mathbb{R}^{2d},\mathbb{C}^{N\times N})$ is of subquadratic growth, if for all $|\alpha|+|\beta|\ge 2$ there exists $C_{\alpha,\beta}>0$ such that*

$$\|\partial_q^\alpha\partial_p^\beta a\|_\infty \le C_\alpha.$$

Lemma A.2. *Let $a\in\mathcal{C}_c^\infty(\mathbb{R}^{2d},\mathbb{C}^{N\times N})$ and let $b\in\mathcal{C}^\infty(\mathbb{R}^{2d},\mathbb{C}^{N\times N})$ be of subquadratic growth. Then, for all $\psi\in\mathcal{C}_c^\infty(\mathbb{R}^d,\mathbb{C}^N)$*

$$\mathrm{op}_1(a)\mathrm{op}_1(b)\psi = \left(\mathrm{op}_1(ab) + \tfrac{1}{2i}\mathrm{op}_1\left(\{a,b\}\right) + \mathrm{op}_1(c) + \mathrm{op}_1(r)\right)\psi,$$

with $\{a,b\} = \partial_p a\partial_q b - \partial_q a\partial_p b$ the Poisson bracket, c a linear combination of

$$\partial_{q_j,p_j}a\,\partial_{q_j,p_j}b,\ \partial_{q_j}^2 a\,\partial_{p_j}^2 b,\ \partial_{p_j}^2 a\,\partial_{q_j}^2 b,$$

and $r\in\mathcal{C}^\infty(\mathbb{R}^{2d},\mathbb{C}^{N\times N})$ such that

$$N_k(r) := \operatorname*{Sup}_{|\alpha|+|\beta|\le k}\|\partial_q^\alpha\partial_p^\beta r\|_\infty \le C_k \sum_{m+m'=k}\left(N_m(D^3a)\,N_{m'}(D^3b)\right)$$

for all $k\in\mathbb{N}$.

For a proof of this classical lemma, the reader can refer to [14] or to [3]. The Theorem of Calderon-Vaillancourt implies, that $\mathrm{op}_1(r)$ is a bounded operator on $L^2(\mathbb{R}^d,\mathbb{C}^N)$.

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