Propagation of Wave Packets, Herman-Kluk Propagators and codimension one crossings

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Introduction

This is a joint work with :
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Schedule of the talk.

1. Propagation of wave packets in semi-classical analysis (Hagedorn 1980 & Combescure-R. 1997).
2. Herman-Kluk propagators. From quantum chemistry (Heller 1981, Herman-Kluk 1984, Kay 1994 & 2006) to mathematics (Rousse-Swart 2009, R. 2010, Lasser-Sattleger 2017).
3. Extension to systems of equations with constant multiplicity eigenvalues.
4. Extension to systems with crossing eigenvalues.

Non-Adiabatic Crossing of Energy Levels.

By Clarence Zener, National Research Fellow of U.S.A.

(Communicated by R. H. Fowler, F.R.S.—Received July 19, 1932.)

1. *Introduction.*

The crossing of energy levels has been a matter of considerable discussion.* The essential features may be illustrated in the crossing of a polar and homopolar state of a molecule.

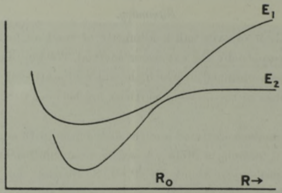

Fig. 1.—Crossing of polar and homopolar states.

Let $\psi_1(x/R)$, $\psi_2(x/R)$ be two electronic eigenfunctions of a molecule with stationary nuclei. Let these eigenfunctions have the property that for $R \gg R_0$, ψ_1 has polar characteristics, ψ_2 homopolar; while at $R \ll R_0$, ψ_2 has polar characteristics, ψ_1 homopolar. In the region $R = R_0$ these two eigenfunctions may be said to exchange their characteristics.

The adiabatic theorem tells us that if the molecule is initially in state ψ_2, and R changes infinitely slowly from $R \gg R_0$ to $R \ll R_0$, then the molecule will remain in state ψ_2. However, if R changes with a finite velocity, the final state $\psi(x/R)$ will be a linear combination

$$\psi(x/R) = A_1(R)\,\psi_1(x/R) + A_2(R)\,\psi_2(x/R). \qquad (1)$$

Neumann and Wigner (*loc. cit.*) have found the conditions for which

$$A_1 \sim 0, \quad |A_2| \sim 1$$

Propagation of wave packets

Let $z=(q,p)\in\mathbb{R}^{2d}$, $a\in\mathcal{S}(\mathbb{R}^d)$, $\|a\|_{L^2(\mathbb{R}^d)}=1$, $\varepsilon\ll 1$.

$$\mathcal{WP}^\varepsilon_z a(x)=\varepsilon^{-\frac{d}{4}}a\left(\frac{x-q}{\sqrt{\varepsilon}}\right)\mathrm{e}^{\frac{i}{\varepsilon}p\cdot(x-q)}$$

Examples :

- Gaussian wave packets: Let Γ a complex symmetric matrix in the positive half Siegel space,

$$a(x)=c_\Gamma\mathrm{e}^{i\Gamma x\cdot x}=:g_0^\Gamma(x).$$

- Hagedorn's wave packets:

$$a(x)=2^{-|\ell|/2}(\ell!)^{-1/2}\pi^{-d/4}[\det A]^{-1/2}\mathcal{H}_\ell(A,|A|^{-1}x)\,\mathrm{e}^{-\frac{1}{2}x\cdot(BA^{-1}x)}$$

for $\ell\in\mathbb{N}^d$ and $\mathcal{H}_\ell$ a Hermite polynomial and A, B adequate matrices.

Notations

- $g_z^{\Gamma,\varepsilon}=\mathcal{WP}^\varepsilon_z g_0^\Gamma$, $g_z^\varepsilon=g_z^{i\mathrm{Id},\varepsilon}$, $g_z^\Gamma=g_z^{\Gamma,1}$, $g_z=g_z^1$.
- $f\in\Sigma^k_\varepsilon$ means $(-\varepsilon^2\triangle_x+|x|^2)^{k/2}\in L^2(\mathbb{R}^d)$.

Wave packets

Let $z=(q,p)\in\mathbb{R}^{2d}$, $a\in\mathcal{S}(\mathbb{R}^d)$, $\|a\|_{L^2(\mathbb{R}^d)}=1$, $\varepsilon\ll 1$.

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Propagation of wave packets

$$i\varepsilon\partial_t\psi^\varepsilon = \widehat{H}(t)\psi^\varepsilon, \;\; \psi^\varepsilon_{|t=t_0} = \psi^\varepsilon_0 \in L^2(\mathbb{R}^d, \mathbb{C}^N)$$

- H is of subquadratic growth

$$\forall \alpha \in \mathbb{N}^{2d}, \;\; |\alpha| \geq 2, \;\; \exists C_\alpha > 0, \quad \sup_{(t,z)\in\mathbb{R}\times\mathbb{R}^{2d}} \|\partial_z^\alpha H(t,z)\|_{\mathbb{C}^{N,N}} \leq C_\alpha.$$

- $\hat{H}(t)$ is the ε-pseudodifferential operator of symbol $H(t)$: if $f \in \mathcal{S}(\mathbb{R}^d, \mathbb{C}^N)$,

$$\widehat{H}(t)f(x) := (2\pi\varepsilon)^{-d} \iint_{\mathbb{R}^d\times\mathbb{R}^d} \mathrm{e}^{\frac{i}{\varepsilon}\xi\cdot(x-y)} H\left(t, \frac{x+y}{2}, \xi\right) f(y)dy\, d\xi.$$

Examples :

1. *Schrödinger Hamiltonian* (Hagedorn): $H_S(t,x,\xi) = \frac{|\xi|^2}{2}\mathbf{1}_{\mathbb{C}^N} + V(t,x)$
2. *Models arising from solid state physics* (Watson & Weinstein): $H_A(x,\xi) = A(\xi) + V_{ext}(t,x)\mathbf{1}_{\mathbb{C}^2}$

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Scalar equations : $N = 1$, $H = h$ scalar

Theorem (Hagedorn, Combescure & R.)

If $\psi_0^\varepsilon = \mathcal{W}P^\varepsilon_{z_0} a$, *then in* $\Sigma^k_\varepsilon(\mathbb{R}^d)$ *(for any* $k \in \mathbb{N}$*)*

$$\psi^\varepsilon(t) = \mathrm{e}^{\frac{i}{\varepsilon} S(t,t_0,z_0)} \mathcal{W}P^\varepsilon_{z(t)} \left(\sum_{j \geq 0} \varepsilon^{j/2} \varphi_j(t,x) \right) + O(\varepsilon^\infty).$$

- $z(t)$ is the classical trajectory associated with h:

$$z(t) = \Phi_h^{t,t_0}(z_0) = (q(t), p(t)).$$

- $S(t, t_0, z_0)$ is the classical action associated with $z(t)$:

$$\dot{S}(t, t_0, z_0) = p(t) \cdot \dot{q}(t) - h(t, z(t)), \;\; S(t_0, t_0, z_0) = 0.$$

- The profiles $\varphi_j(t)$ express in terms of the propagator $\mathcal{M}[F(t, t_0, z_0)]$ associated with the operator $\mathrm{op}_1^w(\mathrm{Hess}_z h(t, z(t)) z \cdot z)$:

$$\varphi_0(t) = \mathcal{M}[F(t, t_0, z_0)]a, \;\; \varphi_j(t) = \mathcal{M}[F(t, t_0, z_0)]\, b_j(t, t_0, z_0)a.$$

Scalar equations – $N = 1$, $H = h$ scalar

$$F(t, t_0, z_0) := \partial_z \Phi_h^{t,t_0}(z_0) = \begin{pmatrix} A(t, t_0, z_0) & B(t, t_0, z_0) \\ C(t, t_0, z_0) & D(t, t_0, z_0) \end{pmatrix},$$

$b_j(t, t_0, z_0)$ are polynomials in (x, D_x) of degree $\leq 3j$.
Example: If $a = g_0^{\Gamma_0}$ then

$$\varphi(t, x) = g_0^{\Gamma(t,t_0,z_0)}(x)$$

with

$$\Gamma(t, t_0, z_0) = (C(t, t_0, z_0) + D(t, t_0, z_0)\Gamma_0)(A(t, t_0, z_0) + B(t, t_0, z_0)\Gamma_0)^{-1}$$

Herman-Kluk propagators

Scalar equations : Hermann Kluk propagator

The Bargmann formula

$$\psi(x) = (2\pi\varepsilon)^{-d} \int_{z\in\mathbb{R}^{2d}} \langle g_z^\varepsilon, \psi\rangle g_z^\varepsilon(x)dz, \quad \psi \in L^2(\mathbb{R}^d)$$

Theorem (Kay 2006, Rousse & Swart 2009, R.2010)

$$\psi^\varepsilon(t) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} \langle g_z^\varepsilon, \psi_0^\varepsilon\rangle u(t,t_0,z) e^{\frac{i}{\varepsilon}S(t,t_0,z)} g^\varepsilon_{\Phi_h^{t,t_0}(z)} dz + O(\varepsilon),$$

in $L^2(\mathbb{R}^d)$, with the Herman–Kluk prefactor

$$u(t,t_0,z) = 2^{-d/2} \det{}^{1/2}\left(A(t,t_0,z) + D(t,t_0,z) + i(C(t,t_0,z) - B(t,t_0,z))\right).$$

$\Longrightarrow$ Numerical solvers of the Schrödinger equation [Lasser & Sattleger 2017]

1. Sampling of the data $\psi_0^\varepsilon(x) \sim (2\pi\varepsilon)^{-d} \sum_{1\leq j\leq N} r_0^\varepsilon(z_j) g^\varepsilon_{z_j}$
2. Approximated formula for $\psi^\varepsilon(t)$ by solving ODEs

$$\psi^\varepsilon(t,x) \sim (2\pi\varepsilon)^{-d} \sum_{1\leq j\leq N} r_0^\varepsilon(z_j) e^{\frac{i}{\varepsilon}S_1(t,t_0,z_j)} u_1(t,t_0,z_j) g^\varepsilon_{\Phi_{z_j}^{t,t_0}}.$$

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Scalar equations : Hermann Kluk propagator

Sketch of Proof

$$\psi_0^\varepsilon(x) = (2\pi\varepsilon)^{-d} \int_{z\in\mathbb{R}^{2d}} \langle g_z^\varepsilon, \psi_0^\varepsilon\rangle g_z^\varepsilon(x) dz$$

$$\Longrightarrow \psi^\varepsilon(t,x) = (2\pi\varepsilon)^{-d} \int_{z\in\mathbb{R}^{2d}} \mathrm{e}^{\frac{i}{\varepsilon}S(t,0,z)} \langle g_z^\varepsilon, \psi\rangle \left(g_{\Phi_h^{t,0}(z)}^{\Gamma(t,0,z),\varepsilon}(x) + \sqrt{\varepsilon}\varphi_1^\varepsilon(t,x,z)\right) dz.$$

We use two lemma

1. Treat the remainder in $O(\sqrt{\varepsilon})$ in order to obtain $O(\varepsilon)$, using the special shape of source term in the equation of $\varphi_1(t)$.
2. Turn the width $\Gamma(t,t_0,z)$ of the Gaussian into the prefactor $u(t,t_0,z)$ by a deformation argument drawing a path form $\Gamma_0 = i\mathrm{Id}$ to $\Gamma_1 = \Gamma(t,0,z)$.

Remark. We could define HK-prefactor connecting $\Gamma_1 = \Gamma(t, t_s 0, z)$ to any Γ_0 instead of $i\mathrm{Id}$.

Scalar equations : Hermann Kluk propagator

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What about systems ?

The constant multiplicity case

Example The Dirac operator

Let $H \in \mathcal{C}^\infty(\mathbb{R} \times \mathbb{R}^{2d}, \mathbb{C}^{N,N})$, subquadratic matrix symbol with two eigenvalues $\lambda_\pm(t, X)$ with multiplicity $m\pm$ $(m_- + m_+ = N)$ s.t $\exists \delta > 0$,

$$|\lambda_+(t, X) - \lambda_-(t, X)| \geq \delta, \ \forall t \in \mathbb{R}, X \in \mathbb{R}^{2d}.$$

The Schrödinger equation

$$i\varepsilon\partial_t\psi^\varepsilon = \widehat{H}(t)\psi^\varepsilon, \ \ \psi^\varepsilon_{|t=t_0} = \psi^\varepsilon_0 \in L^2(\mathbb{R}^d, \mathbb{C}^N)$$

can be decoupled mod $O(\varepsilon^\infty)$ and we are reduced to scalar independent Schrödinger equations (at least for the leading order in ε).
Notations. $\Pi^\pm(t, X)$ the spectral projector for the eigenvalue $\lambda_\pm(t, X)$

Theorem (semi-classical decoupling Emmrich-Weinstein 1996)

There exist unique self-adjoint formal projections $\hat{\Pi}^{\pm,\varepsilon}(t)$, *smooth in* t, *such that* $\hat{\Pi}_0^{\pm}(t,X) = \mathrm{Op}_w^{\varepsilon}(\Pi^{\pm}(t))$ *and*

$$(i\varepsilon\partial_t - H(t))\hat{\Pi}^{\pm,\varepsilon}(t) = \hat{\Pi}^{\pm,\varepsilon}(t)(i\varepsilon\partial_t - H(t)).$$

There exist formal matricial symbols $H^{\pm,\varepsilon}(t)$ *such that* $H_0^{\pm}(t,X) = \lambda^{\pm}(t,X)\mathbf{1}_{m_\pm}$ *and*

$$\Pi^{\pm,\varepsilon}(t)(i\varepsilon\partial_t - \hat{H}(t)) = \hat{\Pi}^{\pm,\varepsilon}(t)(i\varepsilon\partial_t - \hat{H}^{\pm}(t)).$$

Moreover, the subprincipal term $H_1^+(t)$ *of* $H^+(t)$ *is given by the formula*

$$H_1^+(t) = -\frac{1}{2i}(\lambda_+(t) - \lambda_-(t))\{\Pi_+(t), \Pi_+(t)\}$$
$$+i(\partial_t\Pi_+(t) - \{\Pi_+(t), \lambda_+\})(\Pi_+(t) - \Pi_-(t)). \quad (1)$$

Let $\mathcal{U}(t,t_0)$, $\mathcal{U}_\pm(t,t_0)$ the unitary propagators for $\hat{H}(t)$, $\hat{H}^\pm(t)$ (Exist for sub-quadratic Hamiltonians).

Corollary

$$\mathcal{U}(t,t_0) = \hat{\Pi}^{+,\varepsilon}(t)\mathcal{U}_+(t,t_0)\hat{\Pi}^{+,\varepsilon}(t_0) + \hat{\Pi}^{-,\varepsilon}(t)\mathcal{U}_-(t,t_0)\hat{\Pi}^{-,\varepsilon}(t_0) + O(\varepsilon^\infty)$$

Remark The sub-principal terms $H_1^\pm$ in $H^\pm(t)$ mix the states in the modes $+$ and $-$. This is clear by computing time evolution of coherent states.

Let $\mathcal{R}^\pm(t,t_0)$ the matrix satisfying
$$\partial_t \mathcal{R}^\pm(t,t_0) + iH_1^\pm(t, z^\pm(t)) = 0,\quad \mathcal{R}^\pm(t_0,t_0) = \mathbf{1}_{\mathbb{C}^N}.$$

Corollary (Bily Ph.D thesis 2001)

$a := g$ is a Gaussian.
For every $(z_0, v_0) \in \mathbb{R}^{2d} \times \mathbb{C}^N$, we have $\psi^\varepsilon(t) \approx \psi^{+,\varepsilon}(t) + \psi^{-,\varepsilon}(t)$ where

$$\psi^{\pm,\varepsilon}(t) = \mathrm{e}^{\frac{i}{\varepsilon} S^\pm(t,t_0,z_0)} \mathcal{W}P^\varepsilon_{z^\pm(t)} \left(\sum_{j\geq 0} \varepsilon^{j/2} \varphi_j^\pm(t,x) \right) + O(\varepsilon^\infty).$$

$$\varphi_j^\pm(t) = \mathcal{R}^\pm(t,t_0)\mathcal{M}[F^\pm(t,t_0,z_0)]\, b_j^\pm(t,t_0,z_0,v_0)g.$$

$b_j^\pm(t)$, polynomials of degree $\leq 3j$, with coefficients in $\mathbb{C}^N$.

$$b_0^\pm(t) = \pi_\pm(t,z)\mathcal{R}^\pm(t,t_0)\pi_\pm(t_0,z)v.$$

For $j \geq 2$ polarizations $+$ and $-$ are no more separated in $b_j^\pm(t)$.
Explicite computations are possible for $b_2^\pm(t)$!
To follow the phase of states: adiabatic decoupling for systems with a (small) spectral gap.

adiabatic decoupling

Let $h(t,z)$ be an eigenvalue of $H(t,z)$ and $\Pi(t,z)$ the associated eigenprojector :

$$H(t,z)\Pi(t,z) = \Pi(t,z)H(t,z) = h(t,z)\Pi(t,z)$$

Gap assumption: $\exists \delta > 0,\ \forall (t,z),\ \mathrm{dist}\left(h(t,z), \sigma(H(t,z)) \setminus \{h(t,z)\}\right) > \delta.$

Theorem (Kato revisited)

If $\psi_0^\varepsilon = \widehat{\vec{V}_0} v_0^\varepsilon$ *with* $v_0^\varepsilon \in L^2(\mathbb{R}^d, \mathbb{C})$ *and* $\Pi(t_0,z)\vec{V}_0(z) = \vec{V}_0(z)$, *then in* $L^2(\mathbb{R}^d, \mathbb{C}^N)$

$$\psi^\varepsilon(t) = \widehat{\vec{V}(t)} v^\varepsilon(t) + O(\varepsilon)$$

where $\Pi(t,z)\vec{V}(t,z) = \vec{V}(t,z)$ *and* $v^\varepsilon(t)$ *solves*

$$i\varepsilon\partial_t v^\varepsilon = \widehat{h}(t) v^\varepsilon \quad v^\varepsilon_{|t=t_0} = v_0^\varepsilon.$$

Besides, if $(\psi_0^\varepsilon)_{\varepsilon>0}$ *is unif. bounded in* Σ_ε^k $(k \in \mathbb{N})$, *then convergence in* Σ_ε^k.

$\Longrightarrow$ Who is $\vec{V}(t,z)$?

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$\Longrightarrow$ Who is $\vec{V}(t,z)$?

Gapped systems - parallel transport

We set

$$\Omega(t,z) = \Pi(t,z)\{\Pi, H\}(t,z)\Pi(t,z),$$
$$K(t,z) = (\mathrm{Id}_{\mathbb{C}^N} - \Pi(t,z))\,(\partial_t\Pi(t,z) + \{h,\Pi\}(t,z))\,\Pi(t,z).$$

Then,

- $\Omega(t,z)$ is a skew-symmetric matrix,
- $K(t,z)$ and $\Omega(t,z)$ are smooth,.

Proposition (Hagedorn 1994, FLR 2019)

There exists a smooth vector-valued function $\vec{V}(t,z)$ such that

$$\Pi(t,z)\vec{V}(t,z) = \vec{V}(t,z), \;\; |\vec{V}(t,z)| = 1,$$

$$\partial_t\vec{V}(t,z) + \{h,\vec{V}\}(t,z) = \Omega(t,z)\vec{V}(t,z) + K(t,z)\vec{V}(t,z), \quad \vec{V}(0,z) = \vec{V}_0(z).$$

The result holds independently of the existence of the gap if Π and h are smooth.

Gapped systems - Hermann Kluk propagator

Adiabatic decoupling: If $\psi_0^\varepsilon = \widehat{\vec{V}_0} v_0^\varepsilon + O(\varepsilon)$ with $\Pi(0,z)\vec{V}_0(z) = \vec{V}_0(z)$, then

$$\psi^\varepsilon(t) = \widehat{\vec{V}(t)} v^\varepsilon(t) + O(\varepsilon)$$

where $v^\varepsilon(t)$ solves $i\varepsilon\partial_t v^\varepsilon = \widehat{h}(t) v^\varepsilon \quad v^\varepsilon_{|t=t_0} = v_0^\varepsilon$.

$\Longrightarrow$ Herman-Kluk representation of the propagator of a gapped system

Corollary (FLR 2019)

$$\psi^\varepsilon(t) = (2\pi\varepsilon)^{-d} \int_{\mathbb{R}^{2d}} \langle g_z^\varepsilon, v_0^\varepsilon \rangle \; u(t,t_0,z)\vec{V}(t,\Phi_h^{t,t_0}(z)) \, \mathrm{e}^{\frac{i}{\varepsilon}S(t,t_0,z)} \; g^\varepsilon_{\Phi_h^{t,0}(z)} \, dz + O(\varepsilon).$$

- A vector-valued Herman-Kluk prefactor:

$$\vec{U}(t,t_0,z) = u(t,t_0,z)\vec{V}(t,\Phi_h^{t,t_0}(z)).$$

- Approximation in finite time and in $L^2(\mathbb{R}^d)$.

What about systems with crossings ?

$$H(t,z) = v(t,z)\mathrm{Id} + \begin{pmatrix} p_1(t,z) & p_2(t,z) + ip_3(t,z) \\ p_2(t,z) - ip_3(t,z) & -p_1(t,z) \end{pmatrix}. \quad (2)$$

$v \in \mathbb{R}$ and $p(t,z) = (p_1(t,z), p_2(t,z), p_3(t,z)) \in \mathbb{R}^3$ smooth functions. The eigenvalues of $H(t,z)$, ordered by size, are the real numbers

$$v(t,z) - |p(t,z)| = \lambda_-(t,z) \leq \lambda_+(t,z) = v(t,z) + |p(t,z)|.$$

The functions $(t,z) \mapsto \lambda_\pm(z)$ are smooth in open sets where $p(t,z) \neq 0_{\mathbb{R}^3}$. When $\Upsilon := \{(t,z) \in \mathbb{R} \times \mathbb{R}^{2d}, p(t,z) = 0_{\mathbb{R}^3}\}$ is an hypersurface we have a *codimension 1 crossing* (see Hagedorn's classification).

Assume that there exists a smooth function $(t, z) \mapsto f(t, z)$ such that $df \neq 0$ on Υ. Then

$$p_j(t, z) = f(t, z)u_j(t, z), \;\; j \in \{1, 2, 3\}$$

and we assume further that $u_1(t, z)^2 + u_2(t, z)^2 + u_3(t, z)^2 = 1$ on Υ. Then, the functions

$$h_j(z) = v(z) - (-1)^j f(z), \;\; j \in \{1, 2\}, \;\; v = \frac{1}{2}\mathrm{tr}\, H \tag{3}$$

correspond to a renumbering of the eigenvalues on both sides of the hypersurface Υ. This is a specific feature of codimension 1 crossing: the existence of smooth eigenvalues after renumbering.

Systems with codimension 1 crossings – Notations

- $N = 2$, $H(t,z) = h_1(t,z)\Pi_1(t,z) + h_2(t,z)\Pi_2(t,z)$.
- The functions h_1, h_2, Π_1 and Π_2 are smooth and

$$h_1 = v + f, \quad h_2 = v - f, \quad v = \frac{1}{2}\mathrm{Tr}\, H, \quad f \text{ is the gap.}$$

- Classical quantities associated with h_j $j \in \{1,2\}$

$$\Phi_j^{t,t_0}(z),\; S_j(t,t_0,z),\; F_j(t,t_0,z),\; \Gamma_j(t,t_0,z),\; u_j(t,t_0,z),\; \vec{U}_j(t,t_0,z),\dots$$

- Two families of eigenvectors obtained by parallel transport:
  with $\vec{V}_j(t_0,z)$ such that $\Pi_j(t_0,z)\vec{V}_j(z) = \vec{V}_j(z)$, $j \in \{1,2\}$ one associates

$$(t,z) \mapsto \vec{V}_j(t,t_0,z), \quad \vec{V}_j(t_0,t_0,z) = \vec{V}_j(z)$$

Systems with codimension 1 crossings – Geometry

- Generic codimension 1 crossing:

$$\partial_t f + \{v, f\} \neq 0.$$

$\Longrightarrow$ The trajectories are transverse to the crossing hypersurface

$$\Upsilon = \{f(t,z) = 0\}.$$

- If z is such that $\Phi_1^{t,t_0}(z)$ crosses Υ we denote by $t^\natural(z)$ the crossing time

$$\Phi_1^{t^\natural(z),t_0}(z) = z^\natural(z) \in \Upsilon.$$

- The map $z \mapsto t^\natural(z) \in \mathbb{R}$ is well-defined and smooth.
- We associate with $z \in \mathbb{R}^{2d}$

$$S^\natural(z) = S_1(t^\natural, t_0, z), \;\; \alpha^\natural(z) = \left(\left(\{v, \Pi_2\} + \partial_t \Pi_2 \right) \vec{V}_1 \cdot \vec{V}_1^\perp \right) (t^\natural, z^\natural).$$

- With $\vec{V}_1(z) = \Pi_1(t_0, z)\vec{V}_1(z))$, we associate $\vec{V}_1(t, t_0, z)$ and $\vec{V}_2(t, t^\natural, z)$ with

$$\vec{V}_2(t^\natural, t^\natural, z) = \vec{V}_1(t^\natural, t_0, z)^\perp.$$

Codimension 1 crossings – Propagation of wave packets

We assume $\psi_0^\varepsilon = \widehat{\vec{V}_1} v_0^\varepsilon, \;\; v_0^\varepsilon = \mathcal{W}P^\varepsilon_{z_0}(a)$

Theorem (Hagedorn 94, Watson & Weinstein 18, FLR 19-20?)

$$\psi^\varepsilon(t) = \widehat{\vec{V}}_1(t) v_1^\varepsilon(t) + \sqrt{\varepsilon}\, \mathbf{1}_{t>t^\natural}\, \widehat{\vec{V}}_2(t) v_2^\varepsilon(t) + O(\varepsilon^{5/8})$$

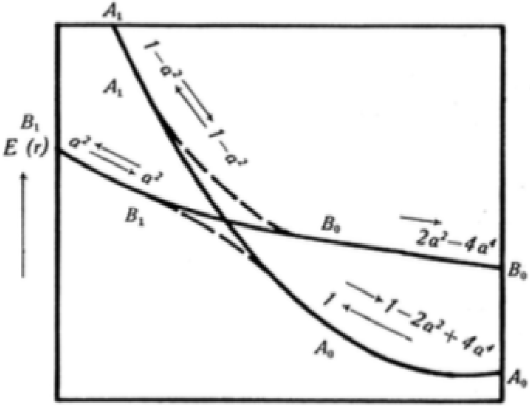
in Σ^k_ε (for any $k \in \mathbb{N}$) where

- $v_1^\varepsilon(t)$ *solves* $i\varepsilon\partial_t v_1^\varepsilon = \widehat{h}_1 v_1^\varepsilon, \;\; v_1^\varepsilon(0) = \mathcal{W}P^\varepsilon_{z_0}(a)$.
- $v_2^\varepsilon(t)$ *solves* $i\varepsilon\partial_t v_2^\varepsilon = \widehat{h}_2 v_2^\varepsilon, \;\; v_2^\varepsilon(t^\natural) = \alpha^\natural\, \mathrm{e}^{iS^\natural/\varepsilon} \mathcal{W}P^\varepsilon_{z^\natural}(\mathcal{T}^\natural \varphi_1(t^\natural))$,

where $\alpha^\natural$, $S^\natural$ are classical quantities associated with the crossing point $z^\sharp$,
$\varphi_1(t^\natural)$ is the profile of $v_1^\varepsilon(t^\natural)$,
and $\mathcal{T}^\sharp$ is a transfer operator mapping $\mathcal{S}(\mathbb{R}^d)$ into itself and preserving Gaussians.

Gaussian Wave packets: If $a = g_0^{\Gamma_0}$ is Gaussian then $\varphi_1(t^\natural) = g_0^{\Gamma_1(t^\natural, t, z_0)}$,

$$\mathcal{T}^\natural g_0^{\Gamma_1(t^\natural, t, z_0)} = g_0^{\Gamma^\natural}.$$



A_1
$1-a^2$
A_1
$1-a^2$
B_1
$E\ (r)$
a^2
a^2
B_1
B_0
$2a^2-4a^4$
B_0
1
$1-2a^2+4a^4$
A_0
A_0

Systems with codimension 1 crossings – Gaussian data

Sketch of proof. In the case $H = H(z)$ (no time dependence)

$$w_1^\varepsilon(t) = \widehat{\Pi}_1\psi^\varepsilon(t) - \widehat{\vec{V}_1(t)}v_1^\varepsilon(t), \quad w_2^\varepsilon(t) = \widehat{\Pi}_2\psi^\varepsilon(t), \quad w^\varepsilon = (w_1^\varepsilon, w_2^\varepsilon).$$

Then $w_1^\varepsilon(0) = w_2^\varepsilon(0) = 0$ and for $s, t \in \mathbb{R}$, in $L^2(\mathbb{R}^d)$,

$$w_1^\varepsilon(t) = w_1^\varepsilon(s) + \int_s^t Q_1(\sigma)w^\varepsilon(\sigma)d\sigma + O(\varepsilon),$$

$$w_2^\varepsilon(t) = w_2^\varepsilon(s) + \int_s^t Q_2(\sigma)w^\varepsilon(\sigma)d\sigma + \widetilde{\mathcal{T}}^\varepsilon(t,s)v_0^\varepsilon + O(\varepsilon)$$

where

$$\widetilde{\mathcal{T}}^\varepsilon(t,s) = \int_s^t \mathrm{e}^{-\frac{i}{\varepsilon}(t-\sigma)\widehat{h}_2}\widehat{\alpha\vec{V}_2}(\sigma)\mathrm{e}^{-\frac{i}{\varepsilon}\sigma\widehat{h}_1}d\sigma$$

the operators Q_1 and Q_2 are bounded.

1. From 0 to $t^\natural - \delta$, one can use adiabaticity and get $\mathcal{T}^\varepsilon(t, t_0) = O(\varepsilon\delta^{-1})$.
2. From $t^\natural - \delta$ to $t^\natural + \delta$, analysis of the operator $\mathcal{T}^\varepsilon(t^\natural + \delta, t^\natural - \delta)$ for small δ.

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$$w_2^\varepsilon(t) = w_2^\varepsilon(s) + \int_s^t Q_2(\sigma) w^\varepsilon(\sigma) d\sigma + \widetilde{\mathcal{T}}^\varepsilon(t,s) v_0^\varepsilon + O(\varepsilon)$$

where

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Codimension 1 crossings – The transfer operator

- Writing $\widetilde{\mathcal{T}}^{\varepsilon}(t^{\natural}+\delta, t^{\natural}-\delta)v_0^{\varepsilon}$ as an operator on $v_1^{\varepsilon}(t^{\natural})$.

$$\widetilde{\mathcal{T}}^{\varepsilon}(t^{\natural}+\delta, t^{\natural}-\delta)v_0^{\varepsilon} = \mathrm{e}^{-\frac{i}{\varepsilon}\delta\widehat{h_2}}\left(\int_{-\delta}^{\delta}\mathrm{e}^{\frac{i}{\varepsilon}\sigma\widehat{h_2}}\widehat{\alpha\vec{V}_2}(t^{\natural}+\sigma)\mathrm{e}^{-\frac{i}{\varepsilon}\sigma\widehat{h_1}}d\sigma\right)v_1^{\varepsilon}(t^{\natural})$$

- Egorov theorem and approximation of the trajectories

$$\widetilde{\mathcal{T}}^{\varepsilon}(t^{\natural}+\delta, t^{\natural}-\delta)v_0^{\varepsilon} = \mathrm{e}^{-\frac{i}{\varepsilon}\delta\widehat{h_2}}\widehat{\alpha\vec{V}_2}(t^{\natural})\left(\int_{-\delta}^{\delta}\mathrm{e}^{\frac{i}{\varepsilon}\sigma\widehat{h_2}}\mathrm{e}^{-\frac{i}{\varepsilon}\sigma\widehat{h_1}}d\sigma\right)v_1^{\varepsilon}(t^{\natural})+O(\delta^2)$$

- Wave packet approximation

$$\widetilde{\mathcal{T}}^{\varepsilon}(t^{\natural}+\delta, t^{\natural}-\delta)v_0^{\varepsilon} = \mathrm{e}^{-\frac{i}{\varepsilon}\delta\widehat{h_2}}\,\mathrm{e}^{iS^{\natural}/\varepsilon}\widehat{\alpha\vec{V}_2}(t^{\natural})\mathcal{T}_{\delta}^{\varepsilon}\varphi_1(t^{\natural})+O(\varepsilon)+O(\delta^2).$$

Codimension 1 crossings – The transfer operator

$$\mathcal{T}_\delta^\varepsilon = \int_{-\delta}^{+\delta} \mathrm{e}^{\frac{i}{\varepsilon}\left(S_1(t^\natural+\sigma,t^\natural,z^\natural)+S_2(t^\natural,t^\natural+\sigma,\Phi_1^{t^\natural+\sigma,t^\natural}(z^\natural))\right)} \mathcal{W}P^\varepsilon_{\zeta(\sigma)} d\sigma.$$

$$\zeta(\sigma) = \Phi_2^{t^\natural,t^\natural+\sigma} \circ \Phi_1^{t^\natural+\sigma,t^\natural}(z^\natural).$$

After Taylor expansions, one can find $\lambda^\natural$, $\beta^\natural = (\beta_q^\natural, \beta_p^\natural)$ such that

$$\mathcal{T}_\delta^\varepsilon = \int_{-\infty}^{+\infty} \mathrm{e}^{i\lambda^\natural s^2} \mathrm{e}^{is\beta_p^\natural\cdot y} \varphi(y - s\beta_q^\natural) ds + O(\varepsilon) + O(\delta^2) + O(\varepsilon^{-1/2}\delta^3).$$

In particular, if $\mu^\natural := \lambda^\natural + \frac{\beta_p^\natural\cdot\beta_q^\natural}{2} \neq 0$, then $\mathcal{T}^\natural = \sqrt{2\pi}\,(\sqrt{\mu^\natural})^{-1} \mathrm{e}^{\frac{i}{4\mu^\natural}(\beta_p^\natural\cdot y - \beta_q^\natural\cdot D_y)^2}$.

- If $H = H_S$, as in [Hagedorn 1994], $\beta_q = 0$.
- If $H = H_A$ as in [Watson-Weinstein 2019], $\beta_p = 0$.

Codimension 1 crossings – Herman-Kluk propagator

Theorem

Assume $\psi_0^\varepsilon = \widehat{\vec{V}_1 v_0^\varepsilon}$. *then*

$$\psi^\varepsilon(t,x) = (2\pi\varepsilon)^{-d}\int_{\mathbb{R}^{2d}} \mathrm{e}^{\frac{i}{\varepsilon}S_1(t,0,z)}\vec{U}_1^\varepsilon(t,z)\langle g_z^\varepsilon, v_0^\varepsilon\rangle g^\varepsilon_{\Phi_1^{t,0}(z)}(x)dz$$

$$+\sqrt{\varepsilon}\,(2\pi\varepsilon)^{-d}\int_{\mathbb{R}^{2d}} \mathbf{1}_{[t^\natural(z)<t]}\alpha^\natural(z)\mathrm{e}^{\frac{i}{\varepsilon}S^\natural(z)}\mathrm{e}^{\frac{i}{\varepsilon}S_2(t,t^\natural,z^\natural)}\vec{U}_2^\varepsilon(t,z)$$

$$\times\langle g_z^\varepsilon, v_0^\varepsilon\rangle g^\varepsilon_{\Phi_2^{t,t^\natural}(z^\natural)}(x)dz + O(\varepsilon^{5/8})$$

with vector-valued Herman-Kluk prefactors

$$\vec{U}_1^\varepsilon(t,z) = \vec{V}_1\left(t,t_0,\Phi_1^{t,t_0}(z)\right)\,u_1(t,t_0,z),$$

$$\vec{U}_2^\varepsilon(t,z) = \vec{V}_2\left(t,t^\natural(z),\Phi_2^{t,t^\natural(z)}(z^\natural(z))\right)\,u_2(t,t^\natural(z),z).$$

Codimension 1 crossings – Proofs

Assume $\partial_t f + \{v, f\} > 0$

- Smoothen the cut off

$$\mathbf{1}_{[t^\natural(z)<t]} = \mathbf{1}_{\Omega(t)}, \;\; \Omega(t) = \{f(z) < 0, \; f(\Phi_1^{t,0}(z)) > 0\}.$$

$\Longrightarrow$ A new parameter $\delta > 0$ and a prefactor of the form $\chi(\delta^{-1}(t - t^\natural(z))$.

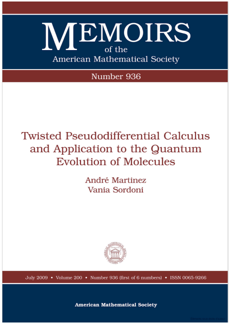
- Identify a canonical transform in the map (broken flow)

$$z \mapsto \Phi_2^{t,t^\natural(z)} \circ \Phi_1^{t^\natural(z),t_0}(z).$$

- Revisit the two lemmas of the scalar proof for this new canonical transform and compute the rest in terms of the derivatives of the prefactor in order to control the lost in δ.

Perspectives

- The Herman-Kluk approximation fits to a numerical realization $\Longrightarrow$ after a phasis of implementation of the initial data, one is reduced to propagate classical quantities along the trajectories.
- The Herman-Kluk approximation of the propagator that we propose for codimension 1 crossing contains the generation of new trajectories when one hits the crossing hypersurface, which is reminiscent from surface hoppings algorithms of quantum chemistry.
- The next step would be to extend this approach to codimension 2 crossings with conical intersections.



Happy birthday André !!!