Quantum Dynamics with Un-Real Trajectories

Robert E. Wyatt
Department of Chemistry and Biochemistry
The University of Texas at Austin

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Overview

A. Features of complex valued quantum trajectories

B. Background on complex valued trajectories

C. Approximate synthetic quantum trajectories: DPM equations of motion

D. Example: Wave packet barrier scattering on a 2 d.o.f. potential surface

E. Exact wave packet dynamics for barrier scattering in the complex plane

F. Exact (analytic) complex quantum trajectories for barrier scattering

Quantum trajectories in complex space

Some features and advantages…

1. This approach is (in some sense) ‘more fundamental’ (according to C. D. Yang)..
2. Understanding complicated dynamics in the complex space can aid in predicting and understanding dynamics on the real plane.
3. For non-degenerate (and degenerate) stationary states, the trajectories are not fixed points but display non-trivial dynamics. They don’t just sit there.
4. The complex quantum potential in the QHJE is constant if S is quadratic in z, so the quantum force vanishes (e.g., Gaussian wave packet).. But for interesting problems this is temporary!
5. The approximate equations of motion are much easier to derive in complex space.
6. When running approximate QTs (using DPM), the convergence may be faster than for approximate Bohm QTs running in real-space. This method works extremely well for some barrier scattering problems (but not very well for others)

Some disadvantages and challenges…

1. z, p, S, V, etc are all complex valued, as are the equations of motion.
   The is part of the doubling problem: a D-dimensional real space problem becomes a 2D-dim complex space problem
2. Given only point-wise data on the real-axis, it is non-trivial to generate V and it’s derivatives in complex space, especially for multi-dimensional problems
3. The potential off the real-axis may have nasty features, such as multiple poles..
   These mess up trajectory propagation.
4. The complex quantum potential can be just as nasty as the Bohm quantum potential (they are both singular near nodes in the wave function)
5. The isochrone problem: Locating isochrones is difficult, especially for multi-dimensional problems. However, at least for some problems, the isochrone search problem can be circumvented.

Background on complex-valued trajectories

Complex-valued *classical* trajectories have been used for decades in semiclassical approaches to barrier tunneling. For example:

. Stine and Marcus (1972)
. Miller and George (1972-1973)
. Heller, Huber, Littlejohn GGWPD (1987); Grossman, Heller (1995)
. Ikeda (1995-)
. Boiron and Lombardi (1998)
. de Aguiar and co-workers (mid-1990s-)
. Kay (2005-)
. Levkov and Sibiryakov (2005-)

Analytical approach to complex *quantum* trajectories: the wave function is known in advance of the trajectory propagation. These exact quantum trajectories are then generated from this wave function. Why do this? Analysis, insight, interpretation

. Moncy John, 2002: harmonic oscillator, potential step
. C.-D. Yang, 2005-present: H atom eigen-trajectories, harmonic oscillator
rectangular barrier tunneling, double slit diffraction, electron spin, chaos
. Chou and Wyatt, 2006-present: potential steps (soft and hard), barriers
(Eckart and Gaussian), reflection-less potential, and others
. Wyatt and Rowland, 2008: use NAC to study time-dependent barrier scattering
in the complex plane
. Sanz and Miret-Artes, 2008: head on collision of two Gaussian wave packets

Synthetic Approach using Approximate Complex Quantum Trajectories

The Goal: Use QTs to solve the **complex-valued Hamilton-Jacobi equation** for the complex action function, S(z,t).

Develop equations of motion for **approximate individual quantum trajectories** using the **derivative propagation method (DPM)** * The DPM was originally developed for real-space QTs:

C. Trahan, K. Hughes, R. Wyatt, J. Chem. Phys. 118, 9911 (2003).

The idea:

1. Develop an infinite hierarchy of coupled DEs for S and its spatial derivatives along the trajectory.

2. To make progress, truncate the system at some (low) order. Pray for convergence.

The highest spatial derivative retained has order n.

'Regional non-locality' is built in because some of the spatial derivatives of S are retained.

* This method has similarities to **CIP** (cubic polynomial pseudo-particle method), used in classical fluid dynamics and transport theory since 1985 (Yabe, Aoki, etc).

The synthetic approach to complex valued quantum trajectories

J. Chem. Phys. **125**, 231103 (2006)

Also see…

J. Chem. Phys. 127, 197101(2007) comment by Sanz and Miret-Artes

J. Chem. Phys. 127, 197102(2007) reply to comment

Bohmian Mechanics with Complex Action: A New Trajectory-Based Formulation of Quantum Mechanics

Yair Goldfarb, Ilan Degani and David J. Tannor

Dept. of Chemical Physics, The Weizmann Institute of Science,Rehovot, 76100 Israel

In recent years there has been a resurgence of interest in Bohmian mechanics as a numerical tool because of its local dynamics, which suggest the possibility of significant computational advantages for the simulation of large quantum systems. However, closer inspection of the Bohmian formulation reveals that the nonlocality of quantum mechanics has not disappeared — it has simply been swept under the rug into the quantum force. In this paper we present a new formulation of Bohmian mechanics in which the quantum action, S, is taken to be complex. This leads to a single equation for complex S, and ultimately complex x and p but there is a reward for this complexification — a significantly higher degree of localization. The quantum force in the new approach vanishes for Gaussian wavepacket dynamics, and its effect on barrier tunneling processes is orders of magnitude lower than that of the classical force. We demonstrate tunneling probabilities that are in virtually perfect agreement with the exact quantum mechanics down to 10^{-7} calculated from strictly localized quantum trajectories that do not communicate with their neighbors. The new formulation may have significant implications for fundamental quantum mechanics, ranging from the interpretation of non-locality to measures of quantum complexity.

More recent work…

Interference effects in reflected wave packet using low-order approx. QTs, 2007

Connection with TD-WKB, 2008

Equations of motion for complex-valued QTs

Ansatz: exponential form for the time-dependent wave function $\psi(x,t) = e^{iS(x,t)/\mathrm{h}}$

S is the complex-valued quantum action function

Substitute into the time-dependent Schrodinger equation to obtain…

$$-\frac{\partial S}{\partial t} = \frac{1}{2m}\left(\frac{\partial S}{\partial x}\right)^2 + V + \frac{\mathrm{h}}{2mi}\frac{\partial^2 S}{\partial x^2}$$

This is the quantum Hamilton-Jacobi equation (QHJE) in the Eulerian frame.

Define

$$Q_C = \frac{\mathrm{h}}{2mi}\frac{\partial^2 S}{\partial x^2}$$

The complex quantum potential.. explicitly brings in quantum effects

Q_C is NOT the same as the Bohm quantum potential Q_B

However, it is just as complicated as Q_B, especially near nodes and quasi-nodes.

See: Rowland and Wyatt, CPL, published on-line.

Origin of complex-valued dynamics

complex action S → complex momentum p → complex coordinate z

$$S(z,t) \qquad p(z,t) = \partial S(z,t)/\partial z \qquad dz/dt = p(z,t)/m$$

How to solve the QHJE using the DPM to generate approximate quantum trajectories

The quantum HJ equation

$$-\frac{\partial S}{\partial t}=\frac{1}{2m}\left(\frac{\partial S}{\partial x}\right)^2+\frac{\mathrm{h}}{2mi}\frac{\partial^2 S}{\partial x^2}+V$$

Notation for derivs.

$$-\frac{\partial S_0}{\partial t}=\frac{1}{2m}S_1^2+\frac{\mathrm{h}}{2mi}S_2+V \qquad S_1=\frac{\partial S}{\partial x},\quad S_2=\frac{\partial^2 S}{\partial x^2},\ldots$$

Now, start the DPM…(1) take the spatial derivatives

Take the x-derivative

$$-\frac{\partial S_1}{\partial t}=\frac{1}{2m}2S_1S_2+\frac{\mathrm{h}}{2mi}S_3+V_1$$

Another x-derivative

$$-\frac{\partial S_2}{\partial t}=\frac{1}{2m}2(S_1S_3+S_2^2)+\frac{\mathrm{h}}{2mi}S_4+V_2$$

Keep going, do it n times

$$\frac{\partial S_n}{\partial t}=F(S_1,\ldots S_n,S_{n+1},S_{n+2})$$

n nonlinear 1-st order DEs for derivatives

Down-coupling to lower terms

Up coupling to next two ‘higher terms’

Next, convert to the moving frame

(2) Convert equations of motion to the moving frame

$$\frac{df}{dt} = \frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x}, \quad v = \frac{dx}{dt}$$ The 'observer' moves at 'arbitrary' velocity v

Special case: Lagrangian dynamics $v_L = \frac{1}{m} S_1$

This gives $$\frac{dS_n}{dt} = F(S_1, \ldots S_n, S_{n+1}, S_{n+2}) + vS_{n+1}$$ n=0,1,2,…

These are the equations of motion in the ALE frame (Los Alamos!)

We still have a hierarchy of equations. What to do?

(3) Truncation of the hierarchy

Hard truncation: Set the 'next two' higher spatial derivs. to zero $S_{n+1} = 0, \quad S_{n+2} = 0$

This leads to the 'usual' DPM (derivative propagation method)

Soft truncation: Put in approximations for $S_{n+1}, \quad S_{n+2}$

For example, run a small cluster and compute $S_{n+1}, \quad S_{n+2}$ from S_n

This leads to CDPM (cluster derivative propagation method), David and Wyatt

In either case, we now have a closed system of equations:
(n+1) coupled DEs for the functions $S_0, S_1, \ldots S_n$

DPM(2) … truncate the system at n = 2

$$-\frac{\partial S_0}{\partial t} = \frac{1}{2m} S_1^2 + \frac{\mathrm{h}}{2mi} S_2 + V \qquad -\frac{\partial S_1}{\partial t} = \frac{1}{2m} 2 S_1 S_2 + V_1 \qquad -\frac{\partial S_2}{\partial t} = \frac{1}{2m} 2 S_2^2 + V_2$$

Equations of motion in the Eulerian frame

Now have 4 equations in the ALE frame

Q_C is included

$$\frac{dS_0}{dt} = -\frac{1}{2m} S_1^2 - \left(\frac{\mathrm{h}}{2mi} S_2 + V\right) + v S_1$$

$$\frac{dS_1}{dt} = -\frac{1}{m} S_1 S_2 - V_1 + v S_2 \qquad \frac{dz}{dt} = v$$

$$\frac{dS_2}{dt} = -\frac{1}{m} S_2^2 - V_2$$

In the Lagrangian frame

$$\frac{dS_0}{dt} = -\frac{1}{2m} S_1^2 - \left(\frac{\mathrm{h}}{2mi} S_2 + V\right) + v S_1$$

$$\frac{dS_1}{dt} = -V_1 \qquad \frac{dp}{dt} = -V_1 = F_{classical} \qquad \frac{dz}{dt} = p/m$$

$$\frac{dS_2}{dt} = -\frac{1}{m} S_2^2 - V_2$$

The result: complex classical trajectories launched with quantum initial conditions and carrying approximate quantum phase

Application to wave packet barrier scattering on a 2 degree of freedom potential surface

R. E. Wyatt and B. A. Rowland, J. Chem. Phys. **127**, 044103 (2007).

barrier potential + harmonic vibrational potential
2 real coordinates: x_1 translation, x_2 vibration

$$V(x_1, x_2) = V_{trans}(x_1) + \frac{1}{2} k(x_1) x_2^2 .$$

trans. harmonic vib.

The vibrational force constant is lower in the barrier region

Translational potential: Eckart or Gaussian barrier centered at $x_1 = 0$

V_0 sech^2 (αx_1) $\qquad$ $V_0 \exp(-\beta x_1^2)$

Veiled Complexity: These potentials look very similar along the real axis but differ greatly in the complex plane

Analytic continuation of the potential surfaces

$$x_1 \rightarrow z_1 = x_1 + i\, y_1, \quad x_2 \rightarrow z_2 = x_2 + i\, y_2 .$$

$$V(x_1, x_2) \rightarrow V(z_1, z_2)$$

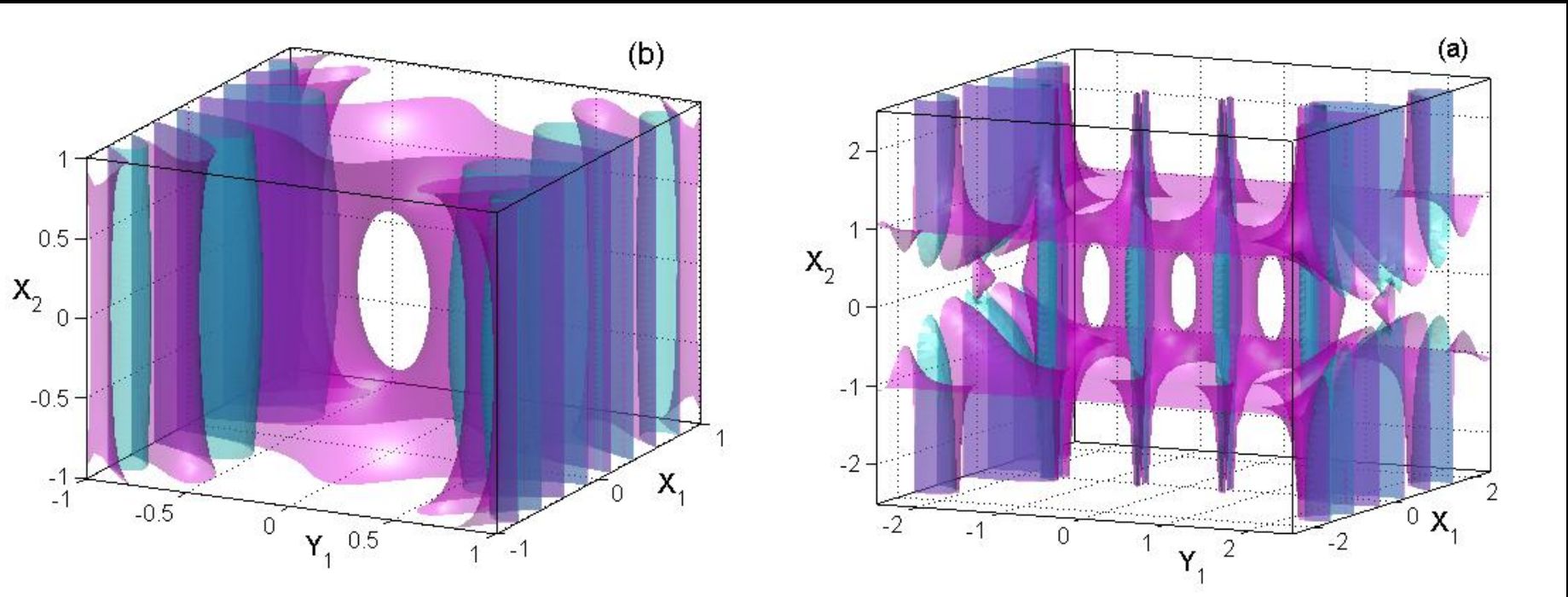
Now have 4 coordinates

The complex-valued potential energy surfaces

The real parts of these complex functions are shown in x_1, y_1, x_2 coordinates

Gaussian

Eckart



No poles in finite complex plane

Periodic string of poles along the imaginary axis
These poles move closer to the real axis as the barrier becomes thinner.

Transmitted trajectories need to make it through ‘holes’ in the potential

Results for DPM(2) trajectories

These are ‘classical trajectories’ propagating in complex phase space.

These trajectories are launched with quantum initial conditions
(for S and its derivatives) and carry quantum phase information,
but follow ‘classical paths’.

A ‘thin’ Eckart barrier and a Gaussian barrier were used.

Launch Points: Isochrones for barrier scattering

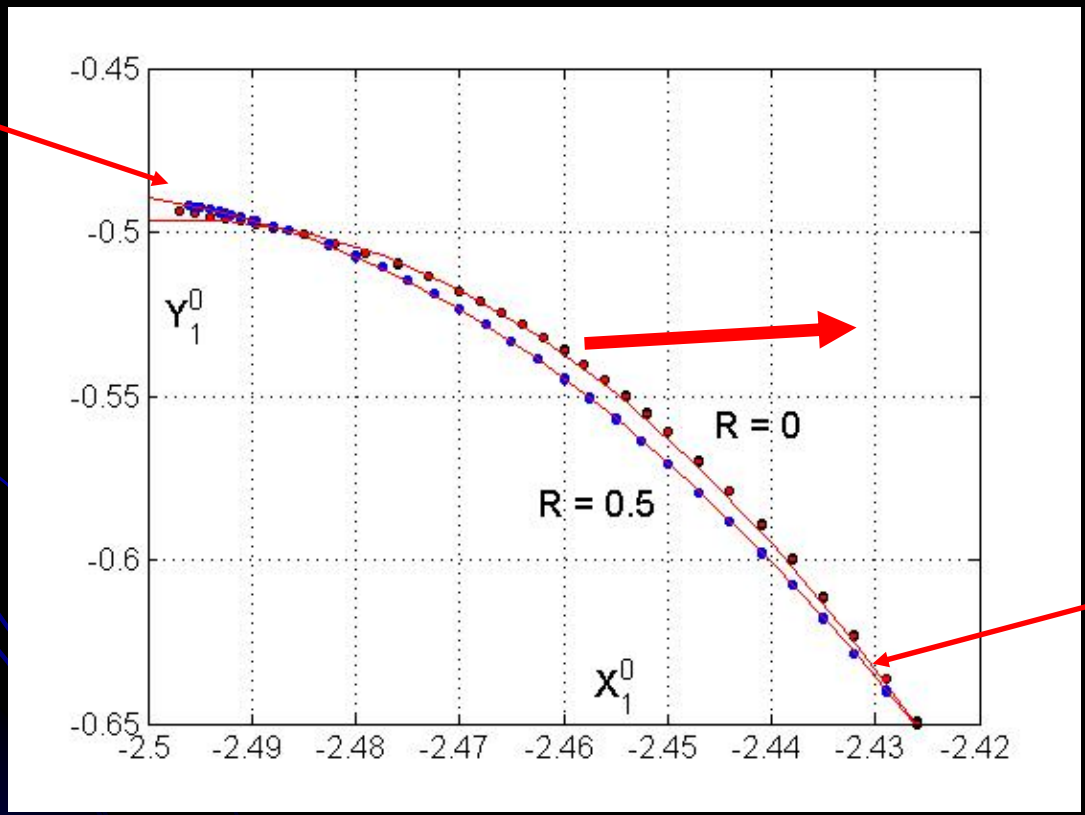
Isochrone: the set of launch points such that trajectories will be 'detected' on the real plane in the transmitted region at the same time. These points fall on a surface in 'initial coordinate space'.

Center of initial wave packet

Arrival time 1200 a.u.

R denotes the initial vibrational displacement

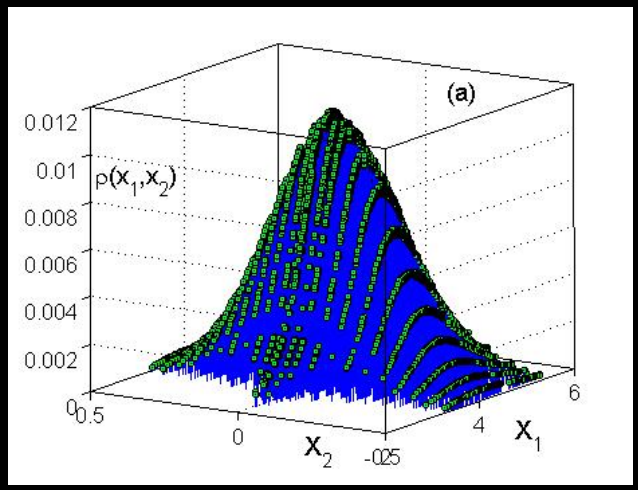
Slow moving



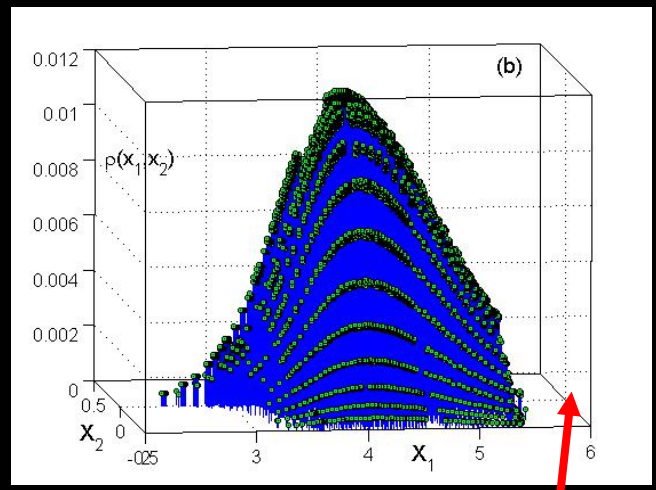
Fast moving

Transmitted density on real plane for the thin Eckart barrier surface

Arrival time 1200 a.u.



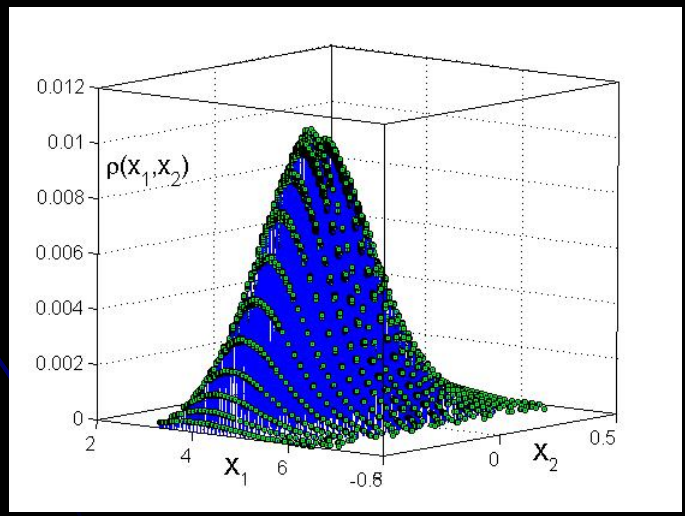
backside



side view

The front end is missing!
Extrapolate the density.

Transmitted density for the Gaussian barrier potential surface



No ‘holes’ or missing regions for the Gaussian case
The integrated density (reaction probability) is within 10% of the exact value

Questions and Directions

1. Why does low-order DPM work much better for thick barriers than for thin ones (Gaussian and Eckart)?

2. For Eckart barriers, how can we ‘fill in’ the holes formed when trajectories diffract around poles in the complex-extended potential?

3. Many DPM trajectories run just fine, but others ‘blow up’. Why?

4. Isochrones (surfaces of launch points) are difficult to locate…
   brute-force grid search is not practical in high dimensionality.

5. Is it possible to avoid the isochrone problem?
   (Yes, with non-Lagrangian ALE constant speed trajectories)

Computational Investigation of Wave Packet Scattering In the Complex Plane

A. Accurate wave packet propagation on a computational grid in z-space

Low energy initial Gaussian wave packet scatters from an Gaussian barrier

Problem: how to deal with the huge numerical values off the real-axis

Introduce both damping of functions and absorbing potentials near grid edges

Follow evolution by plotting several functions: amplitude, QMF, etc.

B. Dynamics of exact quantum trajectories: analytic approach

Thousands of QTs were propagated and plotted

Some groups of trajectories form long-lived, highly correlated structures, such as helical wrapping around stagnation curves.

Also, trajectories display hyperbolic flow as they pass near quasi-nodes

C. Approximate quantum trajectories compared with exact ones

Compare the exact QTs with approximate (DPM) QTs of various orders

Work in progress

The Scattering Problem and the Computational Grid

Initial Gaussian wave packet $\psi(z,0) = (2\beta/\pi)^{1/4} e^{-\beta(z-x_0)^2} e^{ik(z-x_0)}$

Gaussian barrier $V(z) = V_0 e^{-\alpha(z-x_b)^2}$

Parameters (au): $x_0 = 6 \quad x_b = 9 \quad \beta = 6 \quad \alpha = 4 \quad V_0 = 0.035 \quad E = V_0/4$

The computational grid: $x_{min} = 0 \quad x_{\max} = 16 \quad y_{\min} = -3 \quad y_{\max} = 2$

$N_x = 351 \quad N_y = 251 \quad N_{total} = 88,201$

Example of blow-up off of the real axis: $z = (x_0, \pm 3) \quad \psi(z) \approx 10^{23}(!)$

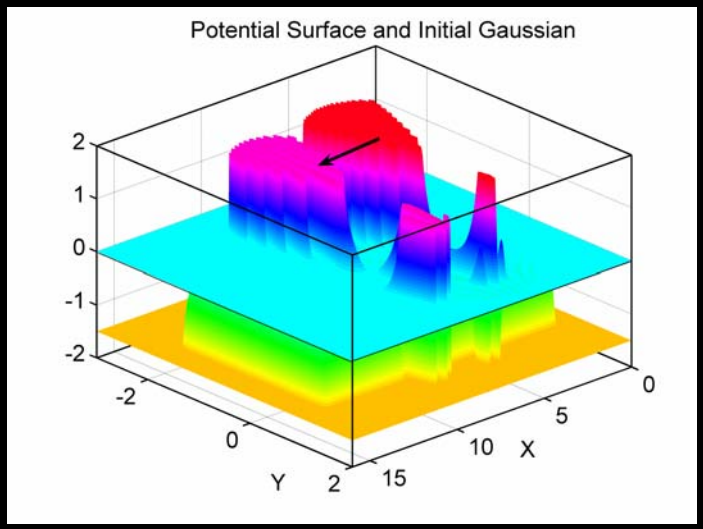
To propagate the wave packet on a finite (small) grid:

(1) Damp the initial packet and the potential along the y-direction,

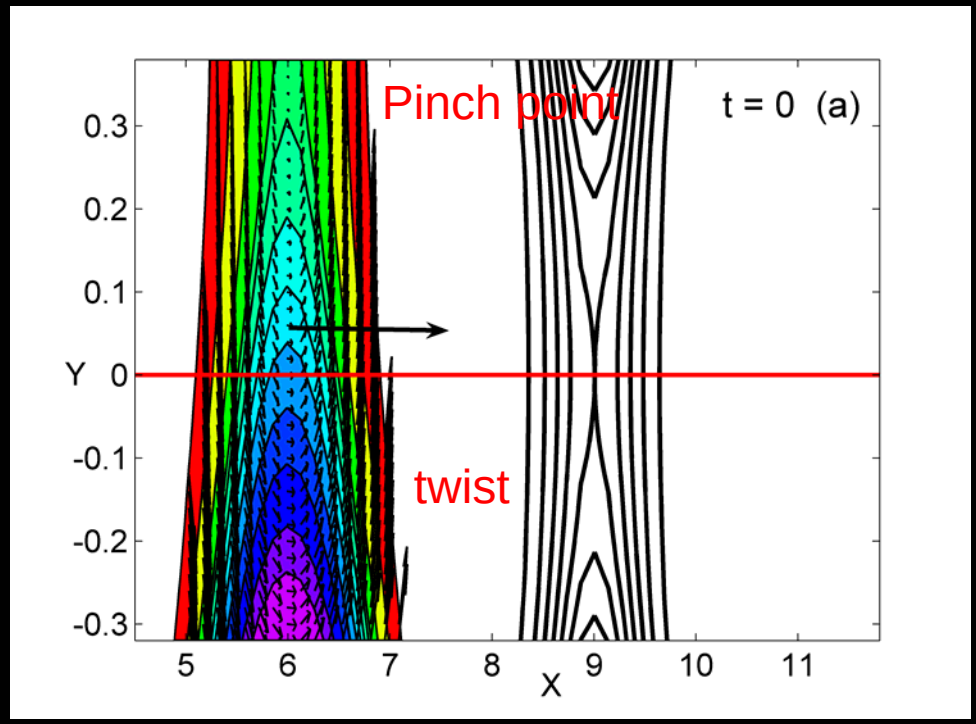
(2) Every M time steps, re-damp the wave packet along the y direction

(3) Use absorbing (negative imaginary) potentials near all 4 grid boundaries

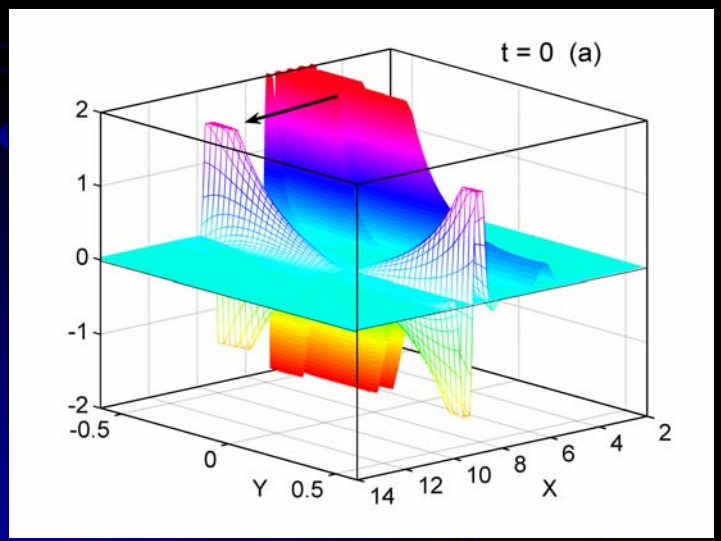
Initial time, t = 0



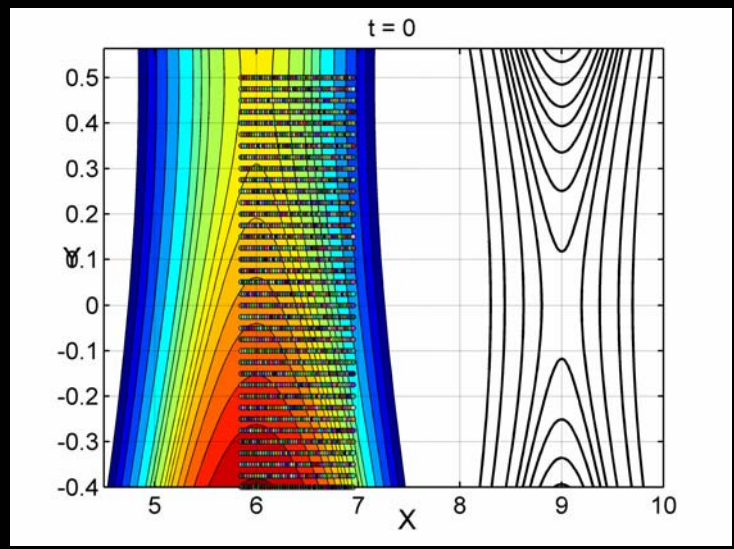
$$V_{total}(z) = V_0 e^{-\alpha(z-x_b)^2} \cdot D(y) + V_{absorb}(z)$$



Quantum momentum function : note swirl around pinch point

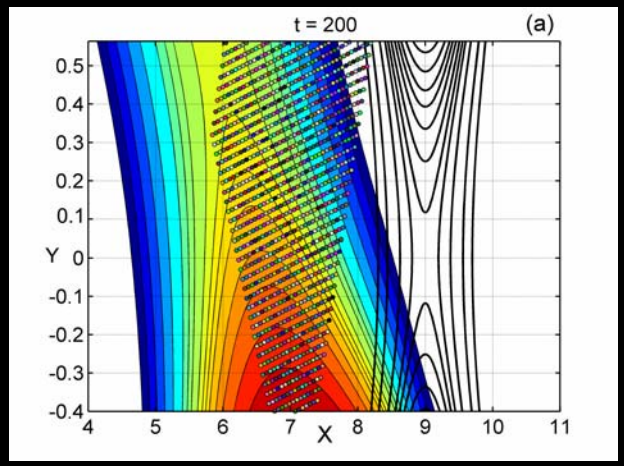


Real (wave function)

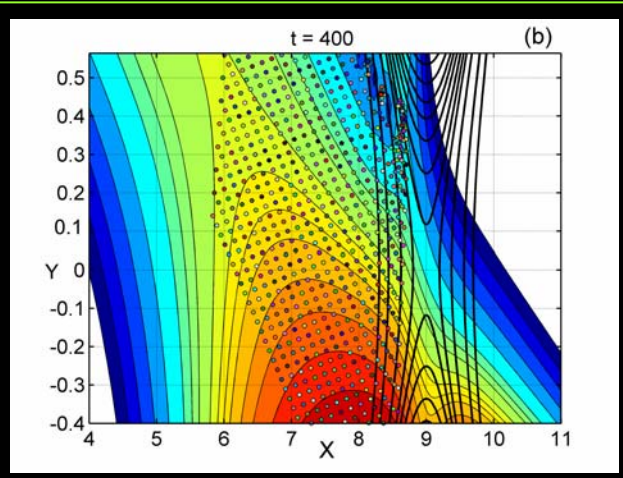


45 x 43 grid of 1935 quantum trajectories

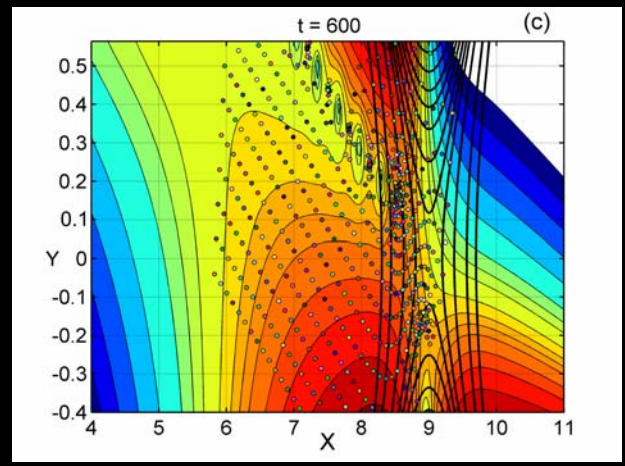
Evolution of exact quantum trajectories



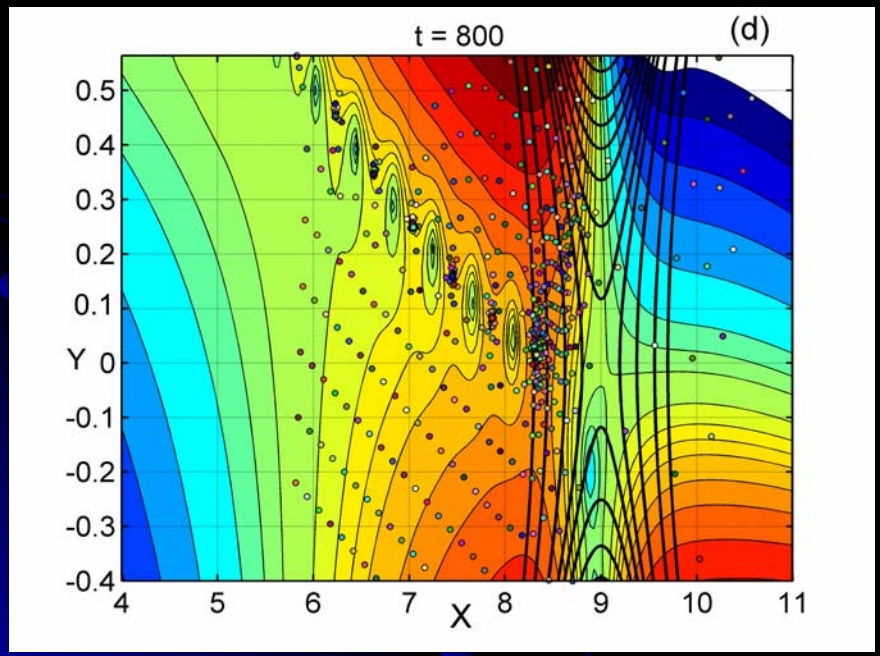
Shearing deformation



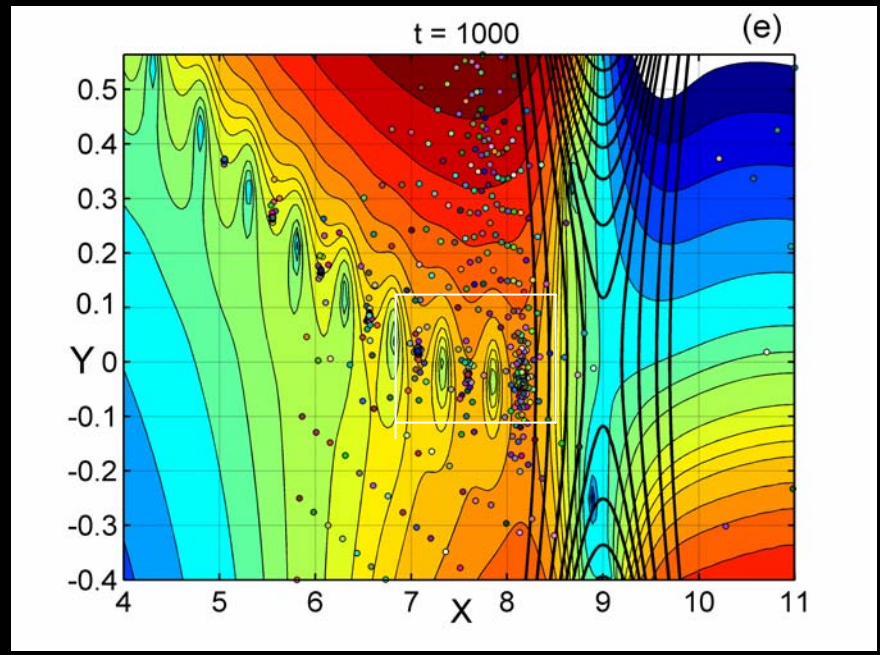
Clustering begins in upper right



Quasi-nodes form



Quasi-nodes and clusters move toward lower left



Quasi-nodes cross real-axis: ‘observer’ detects ‘interference oscillations’

The quantum momentum function and the Polya vector field

Quantum momentum function, QMF $p(z) = \frac{\mathrm{h}}{i}\frac{1}{\psi(z)}\frac{\partial\psi(z)}{\partial z} = (p_r(z), p_i(z))$

Polya vector field
Conjugate field $P(z) = p^*(z) = (p_r(z), -p_i(z))$

Streamlines near nodes and stagnation points

		QMF	Polya field
Near node	$\psi(z) = 0$	Hyperbolic*	Circular
Near stagnation point	$\frac{\partial\psi(z)}{\partial z} = 0$	Circular	Hyperbolic

Vortices: C. C. Chou and R. E. Wyatt, JCP 128, 234106 (2008)
Streamlines: C. C. Chou and R. E. Wyatt, JCP, submitted

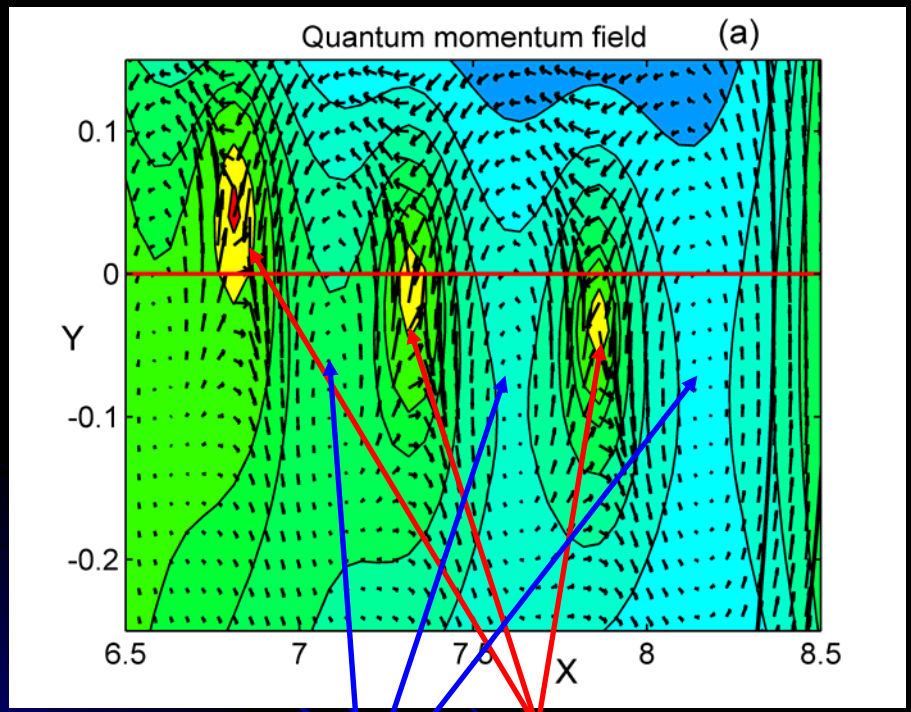
* Compare with vortices in real space: circular flow around node

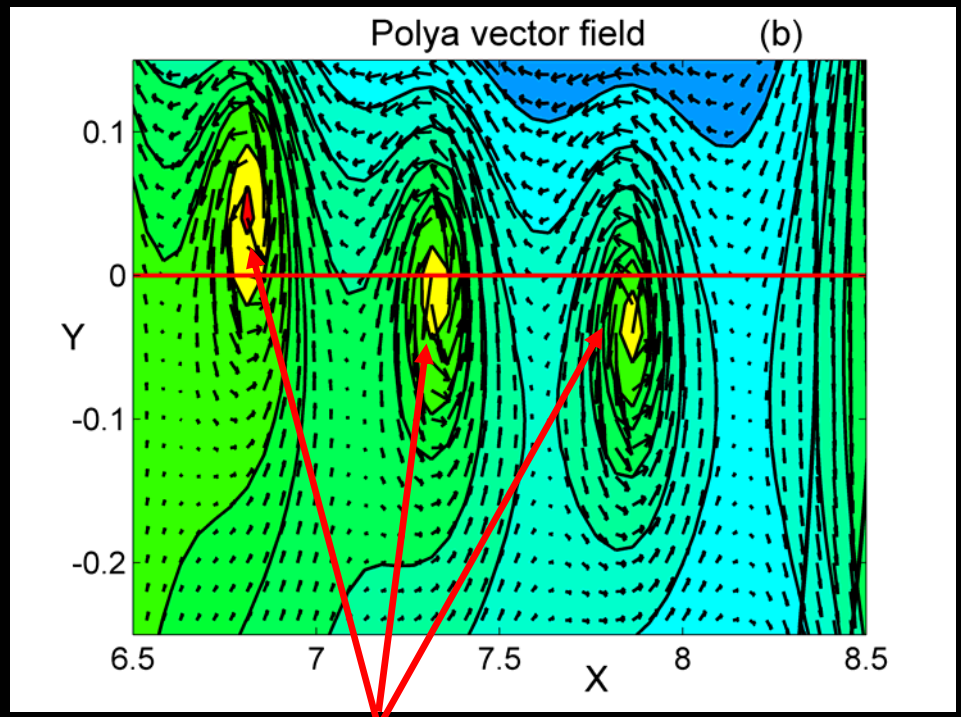
Quantum momentum field and Polya vector field

t=1000 a.u.

$$p(z,t) = \frac{\mathrm{h}}{i}\frac{1}{\psi(z,t)}\frac{\partial\psi(z,t)}{\partial z} = (p_r(z,t),\, p_i(z,t))$$

$$P(z,t) = (p_r(z,t), -p_i(z,t))$$





Quasi-nodes: hyperbolic flow

Stagnation points: circular flow

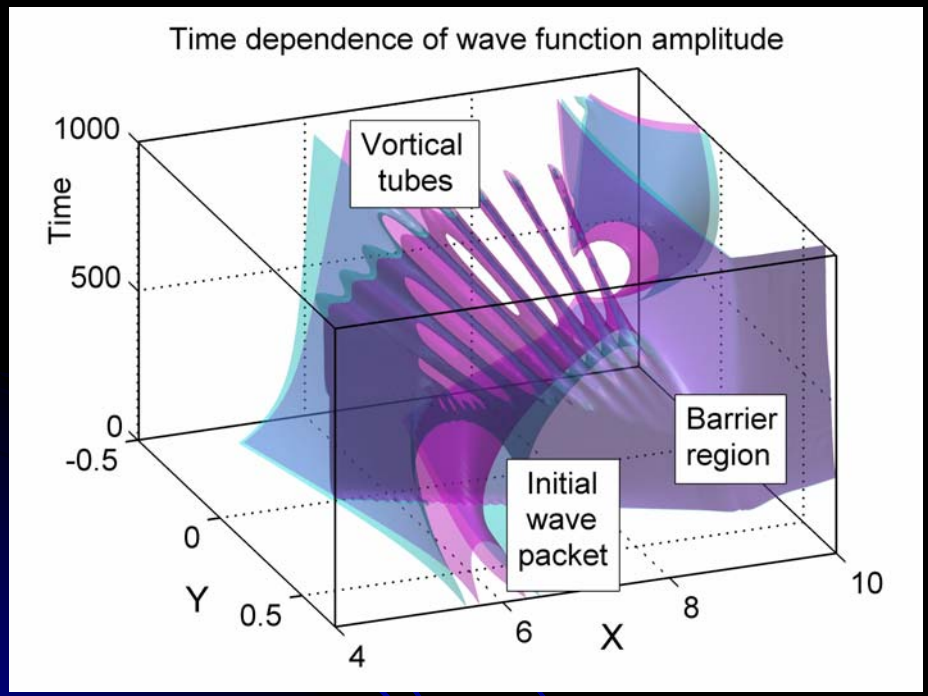
Quasi-nodes: circular flow

Note: The Polya vectors are always parallel to contours of the Born density.

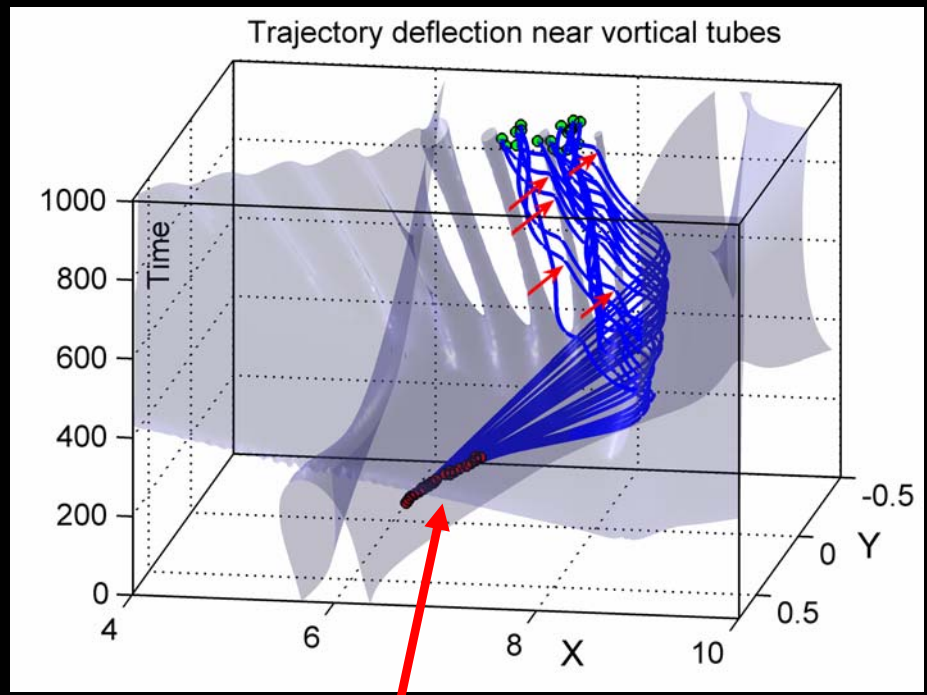
Vortical tubes* and hyperbolic quantum trajectories

* Vortical tubes were introduced into classical hydodynamics by Helmholtz in 1858

Isosurfaces of wave function amplitude show formation and persistence of tubes



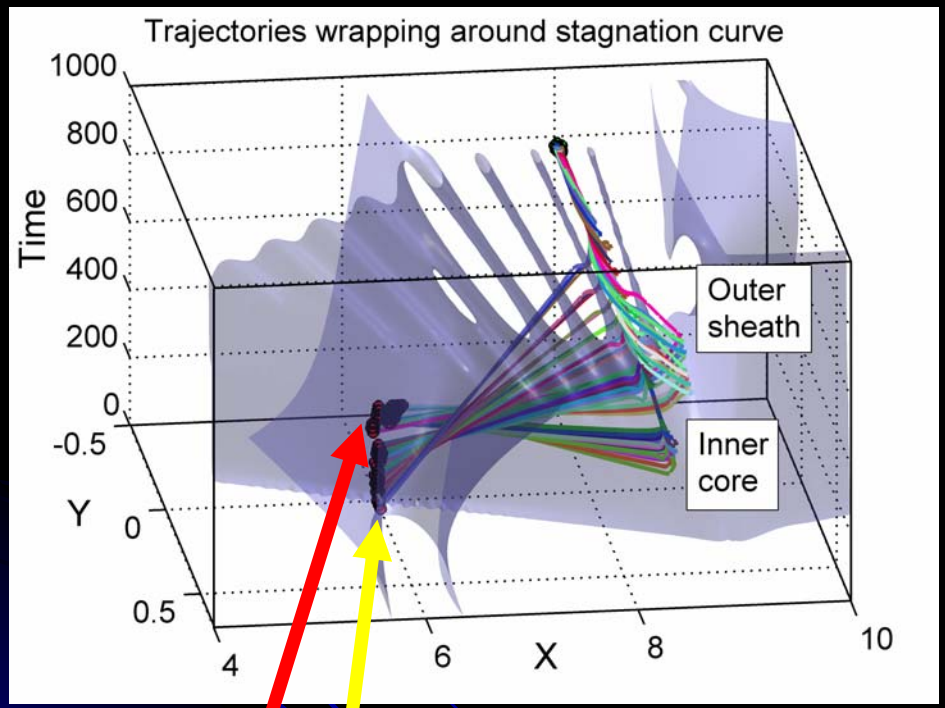
Trajectories experience 'hyperbolic indentations' as they pass near the tubes

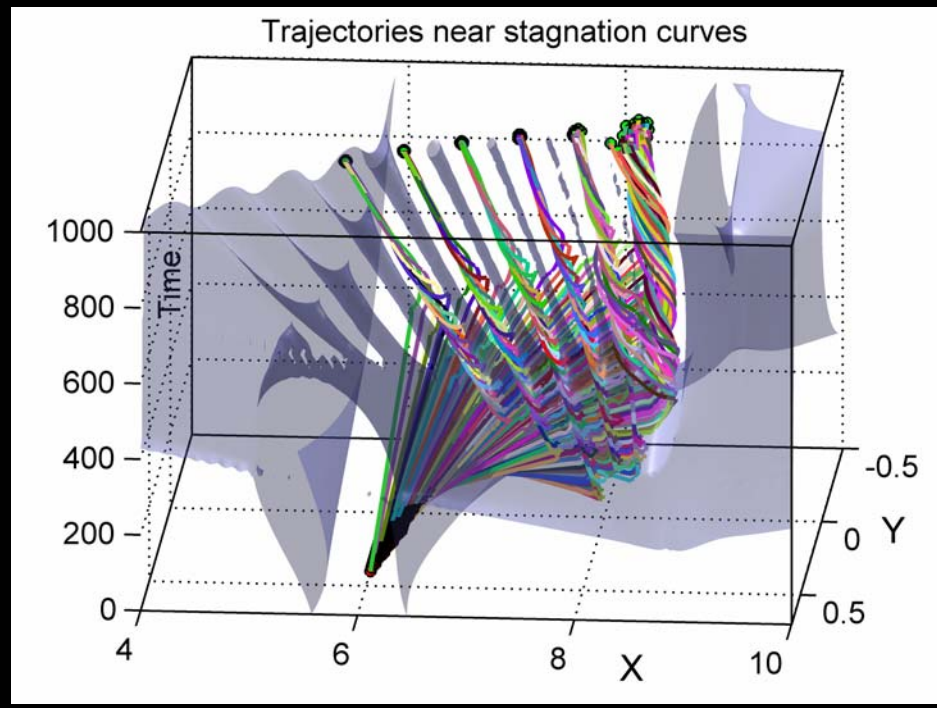


Launch points for trajectories

Trajectory wrapping around stagnation curves

Stagnation curves and vortical tubes alternate with each other.





The first trajectories to arrive wrap tightly to form the inner core. The late arrivals must be content to form the outer sheath. As time proceeds, the trajectories form a helical wrapping around the stagnation curve.

Trajectories from different launch points end up wrapping around the same stagnation curve… this is a type of long-range correlation.

Summary

1. This is the first study in the complex plane of 'exact' wave packet propagation for a barrier scattering problem.

2. Special care was exercised to handle the very large values of the analytically extended initial wave packet and the scattering potential.

3. Important dynamical feature: formation of a string of quasi-nodes and stagnation points above the real-axis. These gradually move away from the barrier region and cross over the real-axis.

4. Dynamics of exact quantum trajectories: Thousands of QTs were propagated and plotted. Some groups of trajectories form long-lived, highly correlated structures, such as helical wrapping around stagnation curves. Also, trajectories display hyperbolic paths when they pass near nodes or quasi-nodes.