

Contents

1	Hagedorn Semiclassical Wavepackets	iv
1.1	Hagedorn's parametrization	v
1.2	Lowering and Raising operators	vii
1.2.1	Recursive relation for Hagedorn wavepackets	vii
1.3	Semiclassical dynamics	viii
1.3.1	Quadratic potential	viii
1.3.2	Cubic potentials	ix
1.3.3	General potentials	ix
1.3.4	Recursive relation for the coefficient matrix (better title)	x
1.4	Numerical/Algorithmic implementation	xvi
1.4.1	Static	xvii
1.4.2	Adaptive	xix
1.5	Approximation of F and higher order cross terms	xxii
2	Non-adiabatic transitions: avoided crossings	xxv
2.1	Change of co-ordinates	xxvii
2.2	Projecting onto Hagedorn basis	xxix
2.2.1	Different parameter set Π	xxix
2.3	d-dimensional case	xxxiii
2.3.1	Monte Carlo Integration	xxxiii
2.3.2	Taylor expansion + Gaussian Integrals	xxxv
2.3.3	One dimension - principle of stationary phase	xxxvi
	Appendices	xl

A	Transmitted Wavepacket	xli
A.1	Transmitted wavepacket	xli
A.1.1	Constant eigenvalues	xli
A.1.2	Non-constant + tilted crossings	xlii
B	Proof of concept for choosing different parameter set in the projection	xlii
2.1	One dimensional case	xlvi
3	Transmitted wavepacket recurrence relation derivation	li

To do:

- Background reading: [Hag98]
- Numerics: [FGL09]
- Tunneling paper: [GHJ10] - Are they also doing a projection? Or they have an explicit expression for the transition of Hagedorn wavepackets?
- Lubich review: [Lub08] - splitting scheme, hyperbolic cross set, Gaussian quadrature rules
- Numerical integration: [Bou17]; Gaussian quadrature rules
- Non-adiabatic transitions near avoided crossings: [BGH12] - this is a fully numerical approach, i.e. just extending the Dirac-Frenkel variational approximation.
- Propagation of wavepackets for conical intersections [FLR20]

1 Hagedorn Semiclassical Wavepackets

[./main_hagedorn.tex]subfilesamsmath

The superadiabatic formulas of equation ... allow us to effectively reduce a coupled systems of two PDEs to two uncoupled PDEs, within the error made by the approximation of the transmitted wavepacket. In this framework, the numerical challenge stems from solving the one level BO dynamics efficiently. In the literature there are various approaches for doing this, often revolving around Gaussian basis sets. **I actually do not know about any methods, this is something to look into.** In this chapter we explore an approach which is rather unknown within the Chemistry community and absent from any QMD commercial or open-source software package. It consists in evolving a set of parametrised orthogonal basis functions, known as Hagedorn wavepackets. Precisely, these are the d-dimensional equivalent of Hermite polynomials, eigenfunctions of the Harmonic oscillator [reference]. When the potential is quadratic one has to evolve only the set of parameters according to a system of ODEs. For more general potentials, one accounts for the non-quadratic remainder term by applying the Dirac-Frenkel variational principle; this leads to a set of integrals involving the non-quadratic remainder term [reference]. Our contribution consists in merging the Superadiabatic formula with this classical evolution of the Hagedorn wavepackets. In Section 1 we introduce the Hagedorn wavepackets and their properties as outlined in [Hag98]. In Section 2 we address the projection of the transmitted wavepacket back onto an Hagedorn basis. The nature of these integrals is similar to the ones encountered in accounting the non-quadratic terms of the quadratic potential.

[./main_hagedorn.tex]subfilesamsmathamssymbdefinitionamsthm,xpatchcomment[backend = biber, style = alphabetic, sorting = ynt]biblatexHagedorn.bib

1.1 Hagedorn's parametrization

To do list:

- Why is it exact for a quadratic potential (metaplectic transformation?)
- (similar to egorov theorem)
- what does $e^{-iH/t}$ do qualitatively? (exponentiation of 1st order derivative is translation)
- How does the variance of the wavepacket evolve in time? Can you draw any qualitative conclusions about when the support spreads out fast vs slow?

Consider a complex semi-classical Gaussian wavepacket in the following form

$$\varphi_0^\epsilon(\mathbf{x}) := (\pi\epsilon)^{-\frac{d}{4}} (\det \mathbf{Q})^{-\frac{1}{2}} \exp \left(\frac{i}{2\epsilon} (\mathbf{x} - \mathbf{q})^T \mathbf{P} \mathbf{Q}^{-1} (\mathbf{x} - \mathbf{q}) + \frac{i}{\epsilon} \mathbf{p}^T (\mathbf{x} - \mathbf{q}) \right) \quad (1.1)$$

Here on we will follow the notational convention as in [Bou17] to denote the set of parameters as $\Pi := (\mathbf{q}, \mathbf{p}, \mathbf{Q}, \mathbf{P})$. The matrix $\mathbb{C}^{d \times d} \ni \mathbf{C} := \mathbf{P} \mathbf{Q}^{-1}$ is complex symmetric with positive definite imaginary part. The matrices \mathbf{P} and \mathbf{Q} must satisfy further properties as enunciated in the following Lemma.

Lemma 1.1 ([Hag98]) *Two square matrices $\mathbf{Q}, \mathbf{P} \in \mathbb{C}^{d \times d}$ satisfy the following relations*

$$\begin{aligned} \mathbf{Q}^T \mathbf{P} - \mathbf{P}^T \mathbf{Q} &= 0 \\ \mathbf{Q}^* \mathbf{P} - \mathbf{P}^* \mathbf{Q} &= 2iI_d \end{aligned} \tag{1.2}$$

if and only if \mathbf{Q} and \mathbf{P} are invertible and $\mathbf{C} = \mathbf{P}\mathbf{Q}^{-1}$ is complex symmetric with positive definite imaginary part. Moreover, $\Im(\mathbf{C}) = (\mathbf{Q}\mathbf{Q}^)^{-1}$.*

(\Rightarrow) Suppose b.w.o.c. that \mathbf{Q} is not invertible. Then $\det(\mathbf{Q}) = 0 = \det(\mathbf{Q}^*)$ which leads to a contradiction when taking the determinant of both sides of the second equation in (1.2). Similarly for \mathbf{P} .

Multiplying the first equation in (1.2) by \mathbf{Q}^{-1} on the right we get $\mathbf{Q}^T \mathbf{P} \mathbf{Q}^{-1} = \mathbf{P}^T \Rightarrow \mathbf{P} \mathbf{Q}^{-1} = (\mathbf{Q}^T)^{-1} \mathbf{P}^T = (\mathbf{P} \mathbf{Q}^{-1})^T$ and therefore \mathbf{C} is complex symmetric. Show positive definiteness: $\mathbf{x}^T \Im(\mathbf{C}) \mathbf{x} = \langle \mathbf{x}, \Im(\mathbf{C}) \mathbf{x} \rangle > 0 \ \forall \ \mathbf{x} \in \mathbb{R}^n$
(\Leftarrow)

Lemma ... gives a sufficient and necessary condition for two matrices \mathbf{Q}, \mathbf{P} to yield such a matrix \mathbf{C} (?) and also a method(?) to construct two desired such matrices. **Given a complex symmetric matrix, how to decompose into such P, Q. Is there a trivial decomposition involving say the identity matrix. However, is there a decomposition which requires a least number of wavepackets such that the error in the norm (or another statistic/observable) is less than some tolerance value?**

The fact that $\Im(\mathbf{C}) = (\mathbf{Q}\mathbf{Q}^*)^{-1}$ justifies why the normalisation factor of the complex Gaussian depends on \mathbf{Q} alone. The reason why Hagedorn's

parametrisation is useful will become clearer when we will look at the dynamics. It is then useful to compare it to what one obtains when this parametrisation is not made.

1.2 Lowering and Raising operators

- Eigenvalues and eigenvectors of the d-dimensional Harmonic oscillator
- Commutator relations...
- This is background reading
- the properties of the raising and lowering operators and then used to derive a recurrence relation for

1.2.1 Recursive relation for Hagedorn wavepackets

For a given parameter set Π with matrices \mathbf{P}, \mathbf{Q} satisfying Lemma 1.1, a Schauder orthonormal basis for $L^2_{\mathbb{C}}(\mathbb{R}^d, \mathcal{B}(\mathbb{R}^d), \lambda^d)$ can be constructed with the basis vectors $\Phi := \{\varphi_k[\Pi]\}_{k \in \mathbb{N}^d}$ satisfying the following recursive relation

$$\begin{aligned} \varphi_0^\epsilon &:= \varphi_0^\epsilon[\Pi](\mathbf{x}) \\ \mathbf{Q} \left(\sqrt{k_j + 1} \varphi_{k+\langle j \rangle}^\epsilon \right)_{j=1}^d &= \sqrt{\frac{2}{\epsilon}} (\mathbf{x} - \mathbf{q}) \varphi_k^\epsilon - \overline{\mathbf{Q}} \left(\sqrt{k_j} \varphi_{k-\langle j \rangle}^\epsilon \right)_{j=1}^d \end{aligned} \quad (1.3)$$

[Exercise: derive recurrence relation](#)

1.3 Semiclassical dynamics

1.3.1 Quadratic potential

Consider a one level TDSE with a quadratic potential $V(\mathbf{x})$ and φ_k^ϵ as initial condition, for some $k \in \mathbb{N}^d$. Then, Hagedorn showed that the solution is fully described in terms of a system of ordinary differential equations for the parameter set Π , that is

$$\dot{\mathbf{q}} = \mathbf{p} \quad (1.4)$$

$$\dot{\mathbf{p}} = -\nabla V(\mathbf{q}) \quad (1.5)$$

$$\dot{\mathbf{P}} = -\nabla^2 V(\mathbf{q}) \mathbf{Q} \quad (1.6)$$

$$\dot{\mathbf{Q}} = \mathbf{P} \quad (1.7)$$

$$S(t) = \int_0^t \frac{|\mathbf{p}(s)|^2}{2} - V(\mathbf{q}(s)) ds \quad (1.8)$$

where $e^{-\frac{i}{\epsilon}S(t)}$ is a multiplicative global phase factor. **To do: understand the equation of motion and describe what happens from a qualitative perspective.**

Can it be proven using Baker-Campbell-Hausdorf formula? First for φ_0 and then generally using the properties of the raising and lowering operators as in..[reference].

Since $\left[-\frac{\epsilon^2}{2}\Delta, V\right] = \frac{\epsilon^2}{2}\Delta(V) = \frac{\epsilon^2}{2}c$ where $c \in \mathbb{R}$, the Baker-Campbell-Hausdorf formula yields

$$\begin{aligned} \varphi_0(x, t) &= e^{-\frac{i}{\epsilon}tH} \varphi_0(x, 0) \\ &= e^{i\frac{\epsilon}{2}t\Delta} e^{-\frac{i}{\epsilon}tV} e^{i\frac{\epsilon}{4}tc} \varphi_0(x, 0) \end{aligned} \quad (1.9)$$

...

1.3.2 Cubic potentials

Why can we not modify the system of ODEs describing the evolution of the parameter set Π such that the result also holds for cubic potentials? If we think of what would happen if we had a tunnelling situation, then the tunnelled wavepacket would have to be described by a different set of parameters or even a different linear combination of Hagedorn wavepackets. **this is an informal answer.** But perhaps there is a modification if the potential is a polynomial of only even degrees, but this will not be very useful from a practical perspective.

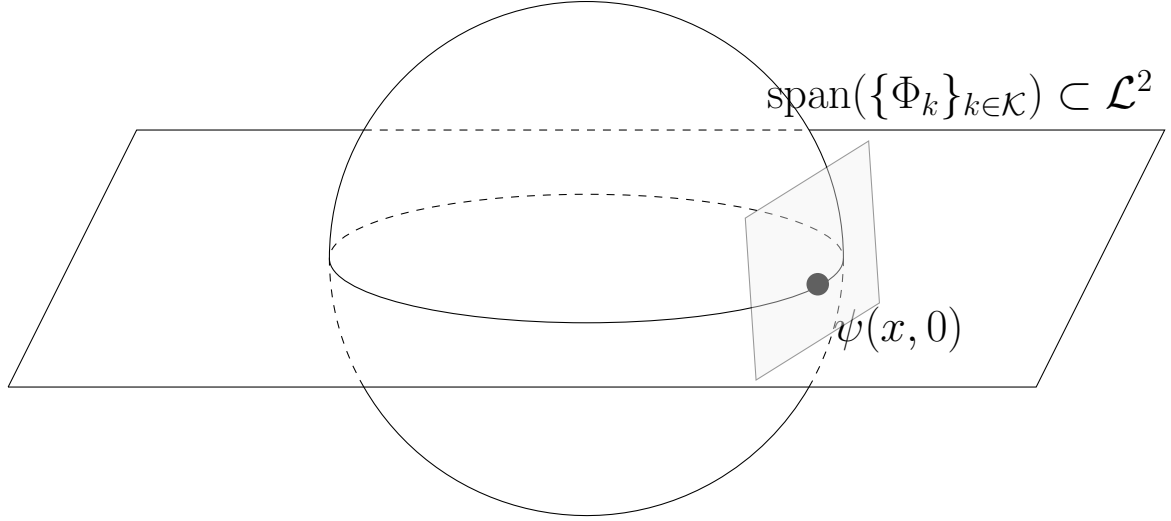
1.3.3 General potentials

Consider the quadratic and non-quadratic remainder term of the potential separately. [Use the picture to aid with the explanation](#) Now consider as initial condition a linear combination of Hagedorn wavepackets $\sum^{\mathcal{K}} \varphi_k$ while the coefficients $\mathbf{c}(t) := (c_k(t))_{k \in \mathcal{K}}$ satisfy the following differential equation

$$i\epsilon \dot{\mathbf{c}}(t) = \mathbf{F}(t)\mathbf{c}(t) \tag{1.10}$$

where $\mathbf{F}_{kl}(t) = \langle \varphi_k, W\varphi_l \rangle = \overline{\mathbf{F}_{lk}}$.

[comment on the kind of system you obtained above intuitively and how you would go about solving it efficiently. What are the properties of the block matrix F](#)



Would the approximation then have necessarily smaller norm...? only if you do the projection and do not increment the number of basis functions

I suppose one would diagonalise \mathbf{F} since it is a symmetric matrix and obtain a fully decoupled system. So what is this Kryloc space about - see Lubich

Different choices for \mathcal{K} have been investigated in [Lub08] together with the related computational challenges. A numerical scheme (order...) based on the splitting method for solving the system of equations (1.4) - (1.7) can be found in [Lub08] while an higher order method in [BG20]

1.3.4 Recursive relation for the coefficient matrix (better title)

I need to specify this somewhere but I have chosen perhaps the non standard convention of conjugating the second entry which will entail having to put the basis vectors in the second entry when doing the projection, I think the

convention is to conjugate the first entry - check it out

For non-quadratic potentials, the major computational effort arises in the computation of the (time dependent) block matrix \mathbf{F} . Its entries are d -dimensional integrals which must be solved at each time step **Check numerical scheme**. **Numerical integration - sampling from the Gaussian**

- Surely we can recycle the sampled points at time zero since we can just translate the Gaussian in the integrand from which we are sampling from to $\mathbf{q} = 0$ (and off course the translation in the potential term)
- Since the time dependence of the integral appears in the integrand only in the parameter set (for non time dependent potentials), we can also morph the sampled points according to how the covariance matrix evolves in time...?
- We still obviously need evaluation of the potential at those points but as far as the sampling goes it can be done only once...?
- Still for the potential you could also recycle the evaluation for neighbouring times?
- How can it be that Monte Carlo is dimension independent but QMC is dimension dependent but nonetheless achieves better convergence? See effective dimension in [WF03]

Is there any way to comment on $\frac{d}{dt}\langle\varphi_k, W\varphi_l\rangle$ - all it is changing are the parameters of the wavepackets - translation in position space and momentum space and dilation/contraction. One could imagine recycling the sampled

points for neighbouring times? or better if I know, how the integral involving the Gaussian evolves, can I say anything about the evolution of the higher order moments. recycling the sample points since you would also consider translating the function to zero, and then simply apply a linear transformation to the sampled points to account for the spread of the wavepacket...? Can I not apply this on the simple resulting integral...? I do not think so as you would have already evaluated the integrand? whose numerical implementation for this algorithm can soon get out of hand. What is the best that one can hope for? How would it scale with the dimension...? Is Monte Carlo independent of the dimension...? would that mean I could use the same number of points independent of the dimension? what if I do integration and then differentiation with Monte Carlo - is that justified?

In [Bou17] this has been addressed through (sparse) quadrature methods (what about QMC?) whose computational complexity is still exponential in nature ? But why? Is there a reason why quadrature methods would be better in this case and MC methods? what about you solve the d-dimensional gaussian integral - what can you say about the derivatives for the higher order terms...?

In this section, we investigate alternative numerical and asymptotic integration methods which have (?) better scaling performance?complexity?. Further, we compute the entries of \mathbf{F} recursively from knowledge of \mathbf{F}_{00} and its "higher order moments" $\langle(\mathbf{x} - \mathbf{q})\rangle$ alone. In other words, given that the Hagedorn wavepackets are polynomials of degree k we have used the linearity of the integral to compute the each monomial where the coefficients are

hidden in the multiplying matrices...(in quotes) Consequently, the problem will reduce to computing \mathbf{F}_{00} and its higher order moments.

In what follows, $W(\mathbf{x})$ denotes as always the non-quadratic remainder term of the potential matrix which we will take to be real valued. We re-write the recursive relation as **At some point I will consider translating everything to the origin**

$$\left(\sqrt{k_j + 1}\varphi_{k+\langle j \rangle}^\epsilon\right)_{j=1}^d = \sqrt{\frac{2}{\epsilon}}\mathbf{Q}^{-1}\left((\mathbf{x} - \mathbf{q})_j\varphi_k^\epsilon\right)_{j=1}^d - \mathbf{Q}^{-1}\overline{\mathbf{Q}}\left(\sqrt{k_j}\varphi_{k-\langle j \rangle}^\epsilon\right)_{j=1}^d \quad (1.11)$$

Since $k_j \geq 0 \forall j$, let \mathbf{K} denote the diagonal matrix with diagonal entries $\mathbf{K}_{jj} = k_j$ for $j \in \{1, \dots, d\}$. Then, multiplying each side of the equality by $(\mathbf{K} + \mathbf{I}_d)^{-\frac{1}{2}}$ yields

$$\begin{aligned} \left(\varphi_{k+\langle j \rangle}^\epsilon\right)_{j=1}^d &= \sqrt{\frac{2}{\epsilon}}(\mathbf{K} + \mathbf{I}_d)^{-\frac{1}{2}}\mathbf{Q}^{-1}\left((\mathbf{x} - \mathbf{q})_j\varphi_k^\epsilon\right)_{j=1}^d + \\ &\quad - (\mathbf{K} + \mathbf{I}_d)^{-\frac{1}{2}}\mathbf{Q}^{-1}\overline{\mathbf{Q}}\mathbf{K}^{\frac{1}{2}}\left(\varphi_{k-\langle j \rangle}^\epsilon\right)_{j=1}^d \end{aligned} \quad (1.12)$$

Letting $\mathbf{A} = \sqrt{\frac{2}{\epsilon}}(\mathbf{K} + \mathbf{I}_d)^{-\frac{1}{2}}\mathbf{Q}^{-1}$, $\mathbf{B} = (\mathbf{K} + \mathbf{I}_d)^{-\frac{1}{2}}\mathbf{Q}^{-1}\overline{\mathbf{Q}}\mathbf{K}^{\frac{1}{2}}$ we can re-write the above expression more concisely as

$$\left(\varphi_{k+\langle j \rangle}^\epsilon\right)_{j=1}^d = \mathbf{A}\left((\mathbf{x} - \mathbf{q})_j\varphi_k^\epsilon\right)_{j=1}^d - \mathbf{B}\left(\varphi_{k-\langle j \rangle}^\epsilon\right)_{j=1}^d \quad (1.13)$$

Multiplying both sides by $W\overline{\varphi_l}$ (a scalar valued function) and integrating over \mathbb{R}^d yields

$$\left(\langle \varphi_{k+\langle j \rangle}^\epsilon, W\varphi_l^\epsilon \rangle\right)_{j=1}^d = \mathbf{A}\left(\langle (\mathbf{x} - \mathbf{q})_j\varphi_k^\epsilon, W\varphi_l^\epsilon \rangle\right)_{j=1}^d - \mathbf{B}\left(\langle \varphi_{k-\langle j \rangle}^\epsilon, W\varphi_l^\epsilon \rangle\right)_{j=1}^d \quad (1.14)$$

Note that in order to compute $\langle \varphi_{k+\langle j \rangle}^\epsilon, W\varphi_l^\epsilon \rangle$ we need the higher order moment of the previous Hagedorn wavepacket $\langle (\mathbf{x} - \mathbf{q})_j \varphi_k^\epsilon, W\varphi_l^\epsilon \rangle$. Similarly, we can derive a recurrence relation for these higher order terms. By defining the diagonal matrix \mathbf{Y} with entries $\mathbf{Y}_{jj} = (\mathbf{x} - \mathbf{q})_j$ and multiplying both sides of equation (1.13) gives

$$\mathbf{Y}^p \left(\varphi_{k+\langle j \rangle}^\epsilon \right)_{j=1}^d = \mathbf{Y}^p \mathbf{A} \left((\mathbf{x} - \mathbf{q})_j \varphi_k^\epsilon \right)_{j=1}^d - \mathbf{Y}^p \mathbf{B} \left(\varphi_{k-\langle j \rangle}^\epsilon \right)_{j=1}^d \quad (1.15)$$

The diagonal matrix \mathbf{Y}^p does not commute with \mathbf{A} unless the latter is also diagonal and so we have that the higher order moments depend on the higher order cross term moments

$$\begin{aligned} \left((\mathbf{x} - \mathbf{q})_j^p \varphi_{k+\langle j \rangle}^\epsilon \right)_{j=1}^d &= \mathbf{Y}^p \left(\sum_{i=1}^d \mathbf{A}_{j,i} (\mathbf{x} - \mathbf{q})_i \varphi_k^\epsilon \right)_{j=1}^d - \mathbf{Y}^p \left(\sum_{i=1}^d \mathbf{B}_{j,i} \varphi_{k-\langle i \rangle}^\epsilon \right)_{j=1}^d \\ &= \left(\sum_{i=1}^d \mathbf{A}_{j,i} (\mathbf{x} - \mathbf{q})_j^p (\mathbf{x} - \mathbf{q})_i \varphi_k^\epsilon \right)_{j=1}^d - \left(\sum_{i=1}^d \mathbf{B}_{j,i} (\mathbf{x} - \mathbf{q})_j^p \varphi_{k-\langle i \rangle}^\epsilon \right)_{j=1}^d \end{aligned} \quad (1.16)$$

Now, repeating the same process as before, multiplying by the scalar $W\overline{\varphi_l}$ on both sides and integrating over \mathbb{R}^d yields

$$\begin{aligned} &\left(\langle (\mathbf{x} - \mathbf{q})_j^p \varphi_{k+\langle j \rangle}^\epsilon, W\varphi_l \rangle \right)_{j=1}^d = \\ &= \left(\sum_{i=1}^d \mathbf{A}_{j,i} \langle (\mathbf{x} - \mathbf{q})_j^p (\mathbf{x} - \mathbf{q})_i \varphi_k^\epsilon, W\varphi_l \rangle \right)_{j=1}^d - \left(\sum_{i=1}^d \mathbf{B}_{j,i} \langle (\mathbf{x} - \mathbf{q})_j^p \varphi_{k-\langle i \rangle}^\epsilon, W\varphi_l \rangle \right)_{j=1}^d \end{aligned} \quad (1.17)$$

which now leads to finding an additional recurrence relation for the cross terms/moments. Note that equation (1.14) is just equation (1.17) with $p = 0$.

We can write out the general expression. Letting $\alpha \in \mathbb{N}^d$, we then have

$$\begin{aligned}
& \left(\left\langle (\mathbf{x} - \mathbf{q})_j^p \prod_{s=1}^d (\mathbf{x} - \mathbf{q})_s^{\alpha_s} \varphi_{k+\langle j \rangle}, W \varphi_l \right\rangle \right)_{j=1}^d = \\
& = \left(\sum_{i=1}^d A_{j,i} \left\langle (\mathbf{x} - \mathbf{q})_j^p \prod_{s=1}^d (\mathbf{x} - \mathbf{q})_s^{\alpha_s} (\mathbf{x} - \mathbf{q})_i \varphi_k, W \varphi_l \right\rangle \right)_{j=1}^d + \\
& - \left(\sum_{i=1}^d B_{j,i} \left\langle (\mathbf{x} - \mathbf{q})_j^p \prod_{s=1}^d (\mathbf{x} - \mathbf{q})_s^{\alpha_s} \varphi_{k-\langle i \rangle}, W \varphi_l \right\rangle \right)_{j=1}^d
\end{aligned} \tag{1.18}$$

We want to make clear the dependence in the computation of F_{ij} on the other ...terms/moments?

For a given index, what other indices do I need? To understand this dependence it suffices to work with the indices.

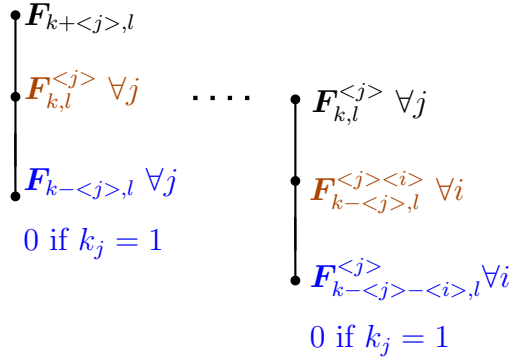
Is there an easy way I can explain this? A graph? Something else? **Simpler case:** Let $k \in \mathbb{N}^d$ and fix $l = 0_{\mathbb{N}^d}$. Looking at equation bla bla Then the intregral with $k + \langle j \rangle$ requires computation of only the first term in equation ... bla bla ... since the second term will have been already computed since we are doing this recursively.

It makes sense just by arguing as them being polynomials, also recall that you would have had computed already the second term and then what you want to say is that for a given index α , what cross terms do you need to re-compute, which ones you need to recycle and so on I am 100% sure there is a way to work through it in terms of indices alone... I wonder if we can think of the algorithm just in terms of indeces then Note that we have kept l fixed above, the reason being that we can use the con-

jugate symmetry property to fill in the other entries. This is to be argued better but ultimately, it should be clear that we can construct the block matrix \mathbf{F} from knowledge of \mathbf{F}_{00} and its higher order terms alone.

[./main_{agedorn.tex}]subfilesamsmathpgfplotscompat = 1.11

1.4 Numerical/Algorithmic implementation



the direction should not matter w.r.t.
direction?

Figure 1: recursion implementation

NOTE THAT THE SECOND TERM IS ZERO FOR K=0 I
STILL ACTUALLY NEED TO MAKE SURE THAT THIS APPROACH
WOULD WORK WITH EVOLUTION OF THE COEFFICIENTS, WHICH
I AM ACTUALLY NOT SURE OF AT THE MOMENT: ALSO ARE YOU
SURE APPLYING THE RECURSION RELATION WOULD NOT GIVE
YOU JUST ANOTHER WAY OF OBTAINING THE COEFFICIENTS OF

THE POLYNOMIALS? There are two approaches that we can consider with regards to the numerical implementation: **static** and **adaptive**. In the static approach the basis set is fixed at all times while for the adaptive approach basis vectors are added until the L2 norm is within some prescribed tolerance of 1. We investigate the latter approach together where the index set is the hyperbolic cross set **To do**:

- Understand why the hyperbolic is a better choice...
- Everything should be in [Lub08] including proof

At the moment we only report the definition. The hyperbolic multi-index set $\mathcal{K}(K) := \left\{ k \in \mathbb{N}^d : \prod_{n=1}^d (1 + k_n) \leq K \right\}$ with $K \in \mathbb{N}$. Because of the recurrence relation it makes sense to consider the cube (Lubich proposes hyperbolic set...)

1.4.1 Static

Here, we fix the basis set $\mathcal{K} \subset \mathbb{N}$ throughout the dynamics. This means that each time step we need to build the block matrix.

In the case of a static basis set throughout the dynamics, we can consider pre-computing all higher order cross terms moments involving φ_{00} or compute them as we go along when needed. The latter approach is definitely the one to take in the adaptive case.

Filling the first diagonal entry of \mathbf{F} is straightforward

$$\mathbf{F} = \begin{bmatrix} F_{00} & \begin{bmatrix} \dots \end{bmatrix} \\ \begin{bmatrix} \dots \end{bmatrix} & \begin{bmatrix} \dots \end{bmatrix} \dots \\ \begin{bmatrix} \dots \end{bmatrix} & \begin{bmatrix} \dots \end{bmatrix} \\ \dots & \dots \end{bmatrix} \quad (1.19)$$

Then, we can proceed filling \mathbf{F} along the columns, i.e. fix $l = 0$, which is in analogy to moving along the axes on the n-dimensional lattice through the recurrence relation. Let $k = (0, \dots, 0)$ and $\langle j \rangle = (0, \dots, 1, \dots, 0)$ In order to compute $F_{00+\langle j \rangle 0}$ I need $\langle (x - q)_j \varphi_0, W \varphi_0 \rangle$, the first moment.

$$\mathbf{F} = \begin{bmatrix} F_{00} & \begin{bmatrix} \dots \end{bmatrix} \\ \begin{bmatrix} F_{00+\langle j \rangle 0} \end{bmatrix}_{j=1}^d & \begin{bmatrix} \dots \end{bmatrix} \dots \\ \begin{bmatrix} \dots \end{bmatrix} & \begin{bmatrix} \dots \end{bmatrix} \\ \dots & \dots \end{bmatrix} \quad (1.20)$$

We can now move one step more along the axes of the d-dimensional lattice to compute $F_{00+\langle j \rangle 0+\langle j \rangle 0}$ where we now need the moments $\langle (\mathbf{x} - \mathbf{q})_j \varphi_1, W \varphi_0 \rangle_{j=1}^d$ which need the cross terms moments $\langle (\mathbf{x} - \mathbf{q})_j (\mathbf{x} - \mathbf{q})_i \varphi_0, W \varphi_0 \rangle$ and need to figure out more generally about $F_{00+\sum_{i=1}^n \langle j \rangle 0}$ for some n .

$$\mathbf{F} = \begin{bmatrix} F_{00} & \begin{bmatrix} \dots \end{bmatrix} \\ \begin{bmatrix} F_{00+\langle j \rangle 0} \end{bmatrix}_{j=1}^d & \begin{bmatrix} \dots \end{bmatrix} \\ \begin{bmatrix} F_{00+\langle j \rangle 0+\langle j \rangle 0} \end{bmatrix}_{j=1}^d & \begin{bmatrix} \dots \end{bmatrix} \\ \dots & \dots \end{bmatrix} \quad (1.21)$$

Since the block matrix is Hermitian/self-adjoint and we have built the first column, we have implicitly built the first row.

$$\mathbf{F} = \begin{bmatrix} F_{00} & \left[\overline{F_{00+\langle j \rangle 0}} \right]_{j=1}^d \\ \left[F_{00+\langle j \rangle 0} \right]_{j=1}^d & \left[\cdots \right] \\ \left[F_{00+\langle j \rangle 0 + \langle j \rangle 0} \right]_{j=1}^d & \left[\cdots \right] \\ \cdots & \cdots \end{bmatrix} \quad (1.22)$$

Now we can fill in the 2nd column

$$\mathbf{F} = \begin{bmatrix} F_{00} & \left[\overline{F_{00+\langle 1 \rangle 0}} & \overline{F_{00+\langle 2 \rangle 0}} & \cdots & \overline{F_{00+\langle d \rangle 0}} \right] \\ \left[F_{00+\langle 1 \rangle 0} \right] & \left[F_{01+\langle 1 \rangle 0} \right] & & & \\ \left[F_{00+\langle 2 \rangle 0} \right] & \left[F_{01+\langle 2 \rangle 0} \right] & & & \\ \cdots & \cdots & & & \cdots \\ \left[F_{00+\langle d \rangle 0} \right] & \left[F_{01+\langle d \rangle 0} \right] & & & \\ \left[F_{10+\langle 1 \rangle 0} \right] & \left[F_{11+\langle 1 \rangle 0} \right] & & & \\ \left[F_{10+\langle 2 \rangle 0} \right] & \left[F_{11+\langle 2 \rangle 0} \right] & & & \\ \cdots & \cdots & & & \\ \left[F_{10+\langle d \rangle 0} \right] & \left[F_{11+\langle d \rangle 0} \right] & & & \\ \cdots & \cdots & & & \end{bmatrix} \quad (1.23)$$

1.4.2 Adaptive

Fixing the basis set throughout the dynamics might not be such a good idea especially one case about preserving conserved quantities such as the norm within some prescribed tolerance $\mathcal{TO}\mathcal{L}$. One may consider a larger set than necessary at time zero to begin with but requires some educated guessing about how many basis function are necessary. It would more convenient to consider an adaptive approach.

1. Decompose initial condition at time zero by augmenting the set \mathcal{K} with basis functions until $|\sum_{k \in \mathcal{K}} |c_k|^2 - 1| \leq \mathcal{TO}\mathcal{L}$
2. When updating coefficients/projecting the transmitted wavepacket repeat at the same process as in 1 perhaps filling up the set using the unit cube or this hyperbolic set.

Example: 1d

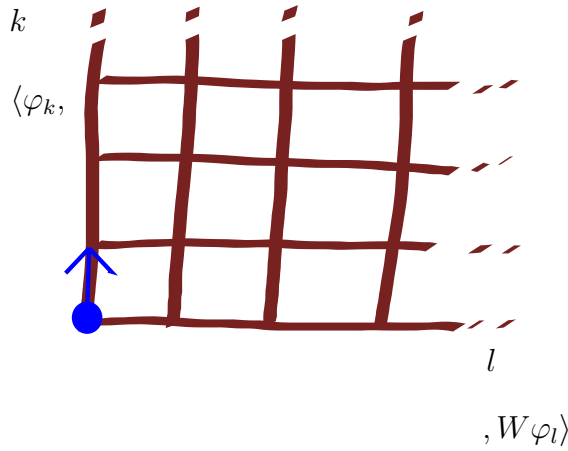


Figure 2: Integral recursion: d=1

(the blue denotes the things we have to compute) (emphasise that one of the terms in the recursion relation will have always been pre-computed - but do we use it anywhere else)

Also other than the moments you will have access to all the results since you need to then exponentiate the matrix

Let us first start by fixing $l = 0$ and understand how the recursion for $\langle \varphi_k, W\varphi_0 \rangle$ unfolds so as to better the pattern (also translate so that $q = 0$).

$\langle \varphi_0, W\varphi_0 \rangle$ (given)

$\langle \varphi_1, W\varphi_0 \rangle \leftarrow \langle x\varphi_0, W\varphi_0 \rangle$ (i.e. the first "moment" of the previous one)

$\langle \varphi_2, W\varphi_0 \rangle \leftarrow \langle \varphi_0, W\varphi_0 \rangle, \langle x\varphi_1, W\varphi_0 \rangle \leftarrow \langle x^2\varphi_0, W\varphi_0 \rangle$

$\langle \varphi_3, W\varphi_0 \rangle \leftarrow \langle \varphi_1, W\varphi_0 \rangle, \langle x\varphi_2, W\varphi_0 \rangle \leftarrow \langle x\varphi_0, W\varphi_0 \rangle, \langle x^2\varphi_1, W\varphi_0 \rangle \leftarrow \langle x^3\varphi_0, W\varphi_0 \rangle$

$\langle \varphi_k, W\varphi_0 \rangle \leftarrow (\text{already computed stuff}) \langle x^k\varphi_0, W\varphi_0 \rangle$

Still to answer the question of whether I use the moments at any other stage

— use Hermitian property...

presumably $k=0$ involves x^2 moment - check - already computed

This means I need to focus only on the integrals involving φ_0

Now it obviously does make perfect sense. But then is there really an advantage...? because essentially you are computing the integral of a polynomial against a gaussian but the polynomial is simplified essentially.. I suppose it would depend on whether you can approximate these integral more easily since you do know the "polynomials" in your integral...

Essentially all it is is that we do know the monomials explicitly, all that has be done really is "hide" the constants and the rescaling (I mean they are polynomials)

1.5 Approximation of \mathbf{F} and higher order cross terms

Is the inner product invariant to translations...?

Hence, we are interested in computing the following integrals involving φ_0^ϵ .

Translate center to origin: $\mathbf{y} = \mathbf{x} - \mathbf{q}$

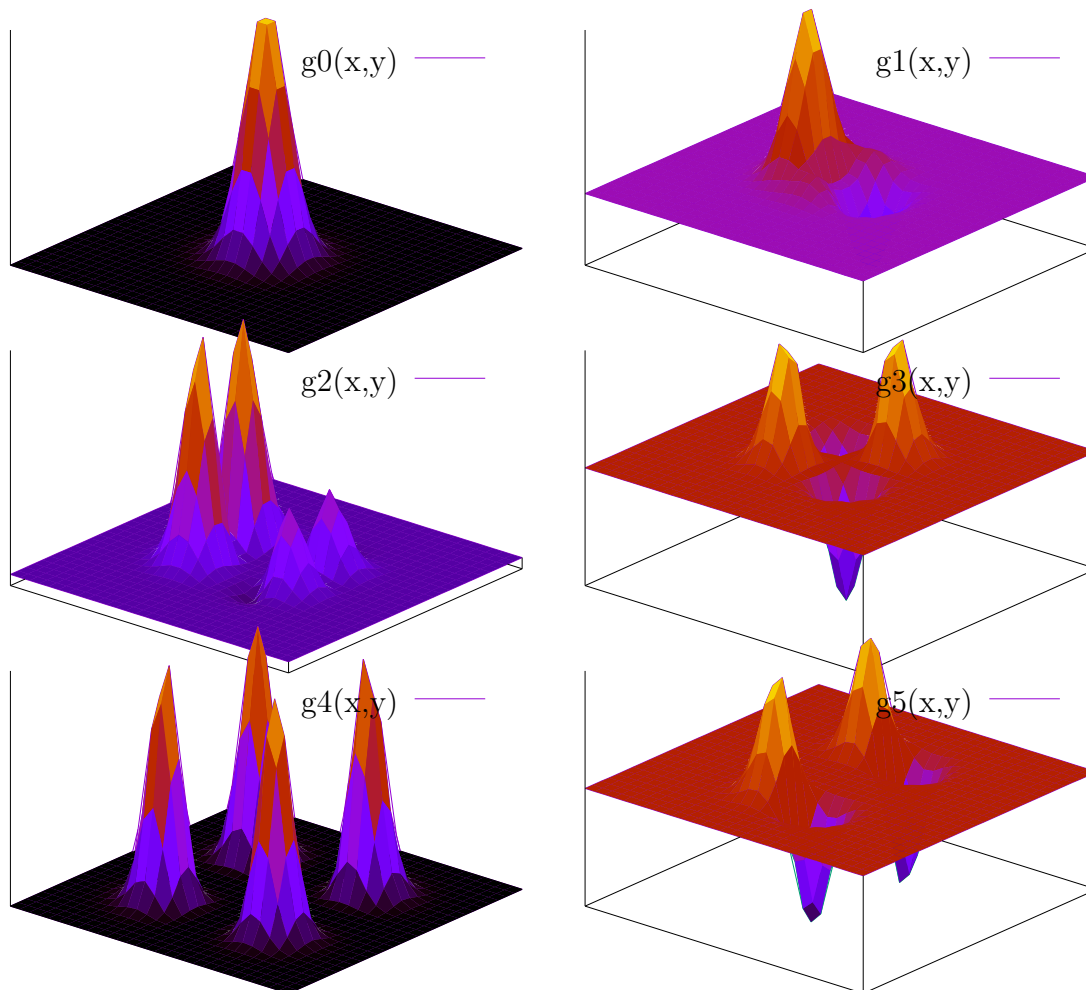
$$\left\langle \prod_{i=1}^d \mathbf{y}_i^{\alpha_i} \varphi_0^\epsilon, W(\mathbf{y} + \mathbf{q}) \varphi_0^\epsilon \right\rangle = \int_{\mathbb{R}^d} \prod_{i=1}^d \mathbf{y}_i^{\alpha_i} W(\mathbf{y} + \mathbf{q}) |\varphi_0^\epsilon|^2 d\mathbf{y} \quad (1.24)$$

Questions: Consider the non-quadratic remainder $W(\mathbf{x})$ of a conical intersection crossing? (I do not think any problems arise if W has a spike singularity - this would be crossing is reached)

- Suppose $\mathbb{N}^d \ni \alpha = 0_{\mathbb{N}^d}$. Then, for W (what assumptions are needed for the Laplace's method?) nice, the integrand is concentrated around \mathbf{q} with variance...?
- But how does the integral behave as we consider higher moments? (plot...) In particular, does the constant in the error using Laplace's method depend on the index..? You would think it would, so what about consider differentiation but then do I really want to do that... Perhaps, it is easier this way as you would be just looking at product rule rather than introducing a delta and differentiating with respect to it
- Could it be stretched more in one direction than in another?
- Does the approximation get worse with the order of the moments...
- Other options: sequence of one dimensional integration, fake derivative, and therefore only one integral..

- More precisely, will the constant in the error get worse?

Integrand



Question: One can approximate the integral using Laplace's method but would you diagonalise or not? Diagonalisation requires computation of eigenvalues which might not be so convenient. where α is an index of powers which, if the sub-basis is fixed will be at most $|\alpha| = \sum_{i=1}^d \alpha_i < f(|\mathcal{K}|)$

Laplace's method would require quite a few number of terms ? in order to give a good error. But derivatives may be expensive to compute? Also, wouldn't the number of terms grow factorially with the dimension when we have composition of differential operators?

Alternative: Consider the integral in (1.25). One solution can be as follows:

$$\begin{aligned} & \int_{\mathbb{R}^d} \prod_{i=1}^d \mathbf{y}_i^{\alpha_i} W(\mathbf{y} + \mathbf{q}) e^{-\frac{1}{\epsilon} \mathbf{y}^T \Im(\mathbf{C}) \mathbf{y}} d\mathbf{y} \\ &= \sqrt{\frac{(2\pi)^d}{\det \Im(\mathbf{C})}} \exp \left[\frac{\epsilon}{4} \right] \end{aligned} \tag{1.25}$$

For certain $u(\mathbf{x})_s$, checking the conditions, one can solve the integral as follows

$$h \tag{1.26}$$

Recall the parameters depend on time. Could this be more efficient than numerical integration? Does not look like it?

2 Non-adiabatic transitions: avoided crossings

Need to discuss [GHJ10] which is about applying a similar method but in the context of tunneling

Still need to look at the work of Hagedorn for gap size shrinking with $\sqrt{\epsilon}$ but also the the work by Olivier using normal local forms in the context of avoided crossings. Hagedorn wavepacket dynamics in the context of avoided crossings were also investigated by Bourquin et al. in [BGH12] The authors extend the one level algorithm outlined in [Lub08] to the multilevel case: the potential matrix $V(\mathbf{x})$ is splitted into a diagonal quadratic term and the non-quadratic remainder, with the non-adiabatic coupling terms occupying the off-diagonal entries. The same dirac-Frenkel variational principle reported in Section ... can be applied to yield a similar set of equations for the update of the coefficients as in equation (...) but now it also involves the coupling terms. Details of the numerics regarding the computation of these integrals can be found in [Bou17]. Here we consider incorporating the formulae for the transmitted wavepackets outlined in section ... in this methodology. Hence, we need to integrate only the one level dynamics. A similar approach was also considered in [BGH12] by implementing the one dimensional transition formula derived in [HJ05] through JWKB analysis but with no concrete implementation given the impracticity of the formula (analytic continuation - needs to be explained better). eventually will need a comparison of the nu-

merical results beside a description of advantages/disadvantages Since the transmitted wavepacket is expressed in momentum space, we might as well work in momentum space, and so take $x \mapsto \xi$, so that we do not worry about pre-factors resulting from the Fourier Transform of Hagedorn wavepackets. We will now denote the parameter set as $\hat{\Pi} := \{p, q, P, Q\}$ as a reminder that we are working in momentum space.

Consider the following initial condition corresponding to one of the adiabatic subspaces

$$\hat{\psi}^{\pm}(\xi, 0) := \sum_{k \in \mathcal{K}} c_k^{\pm}(0) \varphi_k^{\epsilon}[\hat{\Pi}^{\pm}(0)](\xi) \quad (2.1)$$

where $c_k^{\pm} \in \mathbb{C}$ $\xi \in \mathbb{R}^d$ and the index set $\mathcal{K} \subset \mathbb{N}^d$.

How do we detect the avoided crossing?

Let t_c denote the time at which the avoided crossing is detected. The wavepacket at time t_c is given by

$$\hat{\psi}^{\pm}(\xi, t_c) = \exp\left(-\frac{i}{\epsilon} S^{\pm}(t_c)\right) \sum_{k \in \mathcal{K}} c_k^{\pm}(t_c) \varphi_k^{\epsilon}[\hat{\Pi}^{\pm}(t_c)](\xi) \quad (2.2)$$

The projection onto a Hagedorn basis set at the avoided crossing for the transmitted wavepacket requires the following steps:

1. **Change of basis/co-ordinates** for the Hagedorn wavepackets such that a coordinate axis is parallel to $p(t_c)$, the momentum of the wavepacket at the time of the avoided crossing (and keep same orientation)
2. **Compute the new parameter set** for p, P, Q p should be enough

How to detect an avoided crossing in high dimension?

3. Project the transmitted wavepackets onto the new basis set.

[./main_hagedorn.tex]subfilesamsmath

2.1 Change of co-ordinates

In order to apply the one dimensional transition formula we need to rotate the co-ordinate axes such that one co-ordinate lies parallel to p_c . Hence, we want a matrix R satisfying

- $Rp_c = (\|p_c\|, 0, \dots, 0)$ - align momentum vector with one axis
- $R^T = R^{-1}$ (orthogonality)
- $\det(R) = 1$ (keep the same orientation of the basis)

We construct it by applying the Gram-Schmidt process to the vector $\frac{p_c}{\|p_c\|}$, i.e. we form an orthonormal basis $\{r_1 = \frac{p_c}{\|p_c\|}, r_2, \dots, r_d\}$. Stacking this vectors into a matrix R as rows yields the desired rotation matrix R , that is

$$R = \begin{bmatrix} \frac{p_c^T}{\|p_c\|} \\ r_2^T \\ \dots \\ r_d^T \end{bmatrix} \quad (2.3)$$

If $\det(R) = -1$ then multiply its last column by -1 . (There is also another way of doing it called Householder reflection which is cheaper although not sure in what it consists yet)

Applying the change of coordinates to the wavepacket at the crossing yields

$$\hat{\psi}^\pm(\tilde{\xi}, t_c) = \exp\left(-\frac{i}{\epsilon} S^\pm(t_c)\right) \sum_{k \in \mathcal{K}} c_k^\pm(t_c) \varphi_k^\epsilon[\hat{\Pi}^\pm(t_c)](R^T \tilde{\xi}) \quad (2.4)$$

Since the φ_k^ϵ 's obey recurrence relation (1.3) we only need to consider the change of coordinates for the zeroth order wavepacket (1.1)

$$\begin{aligned}
\varphi_0^\epsilon[\hat{\Pi}](\tilde{\xi}) &\propto \exp \left(\frac{i}{2\epsilon} (R^T \tilde{\xi} - p)^T Q P^{-1} (R^T \tilde{\xi} - p) + \frac{i}{\epsilon} q^T (R^T \tilde{\xi} - p) \right) \\
&= \exp \left(\frac{i}{2\epsilon} (R^T (\tilde{\xi} - Rp))^T Q P^{-1} R^T (\tilde{\xi} - Rp) + \frac{i}{\epsilon} q^T R^T (\tilde{\xi} - Rp) \right) \\
&= \exp \left(\frac{i}{2\epsilon} (\tilde{\xi} - Rp)^T R Q P^{-1} R^T (\tilde{\xi} - Rp) + \frac{i}{\epsilon} (Rq)^T (\tilde{\xi} - Rp) \right) \\
&= \exp \left(\frac{i}{2\epsilon} (\tilde{\xi} - Rp)^T R Q (RP)^{-1} (\tilde{\xi} - Rp) + \frac{i}{\epsilon} (Rq)^T (\tilde{\xi} - Rp) \right)
\end{aligned} \tag{2.5}$$

where we have simply applied transpose rules for each equality. Perhaps expected, a rotation of the co-ordinate axes yields a new parameter set $\hat{\Pi}' = \{Rp, Rq, RP, RQ\}$ for the family of Hagedorn wavepackets. Indeed, the matrices RP and RQ still satisfy the symplectic properties of Lemma 1.1 since

$$\begin{aligned}
(RP)^T(RQ) - (RQ)^T(RP) &= \\
P^T R^T RQ - Q^T R^T RP &= \\
P^T Q - Q^T P &= 0
\end{aligned} \tag{2.6}$$

and similarly

$$\begin{aligned}
(RP)^*(RQ) - (RQ)^*(RP) &= \\
P^* R^* RQ - Q^* R^* RP &= \\
P^* R^T RQ - Q^* R^T RP &= \\
P^* Q - Q^* P &= 2iI_d
\end{aligned} \tag{2.7}$$

With these new set of parameters we can then use the recurrence relation of equation (1.3) to yield the higher order wavepackets.

[./main_hagedorn.tex]subfilescomment[backend = biber, style = alphabetic, sorting = ynt]biblatexHagedorn.bib

2.2 Projecting onto Hagedorn basis

Upon detection of the avoided crossing and application of the transmission formula, the transmitted wavepacket $\hat{\psi}^\mp$ needs to be projected back onto a Hagedorn basis set $\Phi[\hat{\Pi}'] = \{\varphi_l[\hat{\Pi}']\}_{l \in \mathcal{L}}$ in order for it to be evolved away from the crossing using Hagedorn dynamics. Since $\Phi[\hat{\Pi}']$ is an orthonormal set, the closest point in its linear span to the transmitted wavepacket is given by

$$P_\Phi \hat{\psi}^\mp := \sum_{l \in \mathcal{L}} \left\langle \frac{\hat{\psi}^\mp}{\|\hat{\psi}^\mp\|}, \varphi_l[\hat{\Pi}'] \right\rangle \varphi_l[\hat{\Pi}'] \quad (2.8)$$

and it is unique since a linear subspace is convex. The coefficients $a_l := \left\langle \frac{\hat{\psi}^\mp}{\|\hat{\psi}^\mp\|}, \varphi_l[\hat{\Pi}'] \right\rangle$ are known as Fourier coefficients. The parameters $\hat{\Pi}$ for the new sub-basis set may take different values than the ones for the incoming wavepacket so as to minimise the number of Fourier coefficients a_l needed to represent the transmitted wavepacket. This is further discussed in the next subsection.explain better

2.2.1 Different parameter set Π

Constant eigenvalues - do it for the general case

Given the considerations outlined in the previous subsection we consider a different parameter set for the Hagedorn basis corresponding to the transmitted wavepacket. More precisely we change the entry of the momentum parameter p corresponding to the direction of motion of the wavepacket at

the crossing, $\{p, q, P, Q\} \mapsto \{p', q, P, Q\}$ where $p'_i = p_i \forall 1 < i \leq d$ while

$$\begin{aligned} p'_1 &= \langle \xi \hat{\psi}^\mp(\xi_1), \hat{\psi}^\mp(\xi_1) \rangle \\ &= \dots \end{aligned} \tag{2.9}$$

The Fourier coefficients are then given by

$$\begin{aligned} a_l &= \left\langle \frac{\hat{\psi}^\mp}{\|\hat{\psi}^\mp\|}, \varphi_l[\hat{\Pi}'] \right\rangle \\ &= \frac{1}{\|\hat{\psi}^\mp\|} \left\langle \sum_{k \in \mathcal{K}} c_k^\pm(t_c) f(\xi_1, \nu(\xi_1); \delta) \varphi_k^\epsilon[\hat{\Pi}_{t_c}^\pm](\tilde{\xi}), \varphi_l^\epsilon[\hat{\Pi}'](\xi) \right\rangle \\ &= \frac{1}{\|\hat{\psi}^\mp\|} \sum_{k \in \mathcal{K}} c_k^\pm(t_c) \left\langle f(\xi_1, \nu(\xi_1); \delta) \varphi_k^\epsilon[\hat{\Pi}_{t_c}^\pm](\tilde{\xi}), \varphi_l^\epsilon[\hat{\Pi}'](\xi) \right\rangle \end{aligned} \tag{2.10}$$

where

$$f(\xi_1, \nu(\xi_1); \delta) = \exp\left(-\frac{i}{\epsilon} S^\pm(t_c)\right) \sin\left(\frac{\pi\gamma}{2}\right) \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) \exp\left[-\frac{q_c}{\epsilon} |\xi_1 - \nu(\xi_1)|\right]$$

and

$$\tilde{\xi} = (\nu(\xi_1), \xi_2, \dots, \xi_d)$$

It is actually convenient to define

$$a_{kl} := \left\langle f(\xi_1, \nu(\xi_1); \delta) \varphi_k^\epsilon[\hat{\Pi}_{t_c}^\pm](\tilde{\xi}), \varphi_l^\epsilon[\hat{\Pi}'](\xi) \right\rangle$$

so that

$$a_l = \frac{1}{\|\hat{\psi}^\mp\|} \sum_{k \in \mathcal{K}} c_k^\pm(t_c) a_{kl}$$

Following the same approach as for the update of the coefficients stemming from the variational approximation, computation of the coefficients requires solving integrals of the following form

$$kd \tag{2.11}$$

Under the integral sign, the product of the two Gaussians is given by

$$\begin{aligned}
&= \exp \left[\frac{i}{2\epsilon} (\tilde{\xi} - p)^T Q P^{-1} (\tilde{\xi} - p) + \frac{i}{\epsilon} q^T (\tilde{\xi} - p) - \frac{q_c}{\epsilon} |\xi_1 - \nu(\xi_1)| \right] \\
&\quad \exp \left[\frac{i}{2\epsilon} (\xi - p')^T Q P^{-1} (\xi - p') + \frac{i}{\epsilon} q^T (\xi - p') \right] d\xi
\end{aligned} \tag{2.12}$$

We can simplify the argument in the exponential term further. Consider the real and imaginary matrix decomposition $Q P^{-1} = R + iI$ where R and I are symmetric since $Q P^{-1}$ is, and let

$$\begin{aligned}
\tilde{a} &= [\nu(\xi_1) - p_1, 0, \dots, 0] & a &= [\xi_1 - p'_1, 0, \dots, 0] & b &= [0, \xi_2 - p_2, \dots, \xi_d - p_d]
\end{aligned} \tag{2.13}$$

. Then,

$$\begin{aligned}
& \frac{i}{2\epsilon}(\tilde{a} + b)^T(R + iI)(\tilde{a} + b) + \frac{i}{\epsilon}q^T(\tilde{a} + b) - \frac{q_c}{\epsilon}|\xi_1 - \nu(\xi_1)| + \\
& \frac{i}{2\epsilon}(a + b)^T(R + iI)(a + b) + \frac{i}{\epsilon}q^T(a + b) = \\
& - \frac{1}{2\epsilon}[(\tilde{a} + b)^T I(\tilde{a} + b) + (a + b)^T I(a + b) + 2q_c|\xi_1 - \nu(\xi_1)|] \\
& + \frac{i}{2\epsilon}[(\tilde{a} + b)^T R(\tilde{a} + b) - (a + b)^T R(a + b) + 2q^T(\tilde{a} - a)] = \\
& - \frac{1}{2\epsilon}[\tilde{a}^T I \tilde{a} + a^T I a + 2b^T I b + (\tilde{a}^T + a^T)Ib + b^T I(\tilde{a} + a) + 2q_c|\xi_1 - \nu(\xi_1)|] \\
& + \frac{i}{2\epsilon}[\tilde{a}^T R \tilde{a} - a^T R a + (\tilde{a}^T - a^T)Rb + b^T R(\tilde{a} - a) + 2q_1(\tilde{a}_1 - a_1)] = \\
& - \frac{1}{2\epsilon}[\tilde{a}^T I \tilde{a} + a^T I a + 2b^T I b + 2(\tilde{a}^T + a^T)Ib + 2q_c|\xi_1 - \nu(\xi_1)|] \\
& + \frac{i}{2\epsilon}[\tilde{a}^T R \tilde{a} - a^T R a + 2(\tilde{a}^T - a^T)Rb + 2q_1(\tilde{a}_1 - a_1)] = \\
& - \frac{1}{2\epsilon}\left[I_{11}(\tilde{a}_1^2 + a_1^2) + 2b^T I b + 2(\tilde{a}_1 + a_1) \sum_{j=2}^d I_{1,j}b_j + 2q_c|\xi_1 - \nu(\xi_1)|\right] \\
& + \frac{i}{2\epsilon}\left[R_{11}(\tilde{a}_1^2 - a_1^2) + 2(\tilde{a}_1 - a_1) \sum_{j=2}^d R_{1,j}b_j + 2q_1(\tilde{a}_1 - a_1)\right] = \\
& - \frac{1}{2\epsilon}\left[I_{11}(\tilde{a}_1^2 + a_1^2) + 2b^T I b + 2(\tilde{a}_1 + a_1) \sum_{j=2}^d I_{1,j}b_j + 2q_c|\xi_1 - \nu(\xi_1)|\right] \\
& + \frac{i}{2\epsilon}\left[R_{11}(\tilde{a}_1^2 - a_1^2) + 2(\tilde{a}_1 - a_1) \left(q_1 + \sum_{j=2}^d R_{1,j}b_j\right)\right]
\end{aligned} \tag{2.14}$$

We investigate different attempts to solving the integral for the Fourier coefficients. For each approach there is a convenient way of re-writing the integral.

The integral representation of a_{kl} is given by

$$\begin{aligned}
a_{kl} &\propto \int_{\mathbb{R}^d} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^\pm](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \times \\
&\exp \left\{ -\frac{1}{2\epsilon} \left[I_{11}(\tilde{a}_1^2 + a_1^2) + 2b^T I b + 2(\tilde{a}_1 + a_1) \sum_{j=2}^d I_{1,j} b_j + 2q_c |\xi_1 - \nu(\xi_1)| \right] \right\} \\
&\exp \left\{ \frac{i}{2\epsilon} \left[R_{11}(\tilde{a}_1^2 - a_1^2) + 2(\tilde{a}_1 - a_1) \left(q_1 + \sum_{j=2}^d R_{1,j} b_j \right) \right] \right\} d\xi \\
&= \int_{\mathbb{R}^{d-1}} \exp \left\{ -\frac{1}{\epsilon} [b^T I b] \right\} \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^\pm](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \\
&\times \exp \left\{ -\frac{1}{2\epsilon} \left[I_{11}(\tilde{a}_1^2 + a_1^2) + 2(\tilde{a}_1 + a_1) \sum_{j=2}^d I_{1,j} b_j + 2q_c |\xi_1 - \nu(\xi_1)| \right] \right\} \\
&\exp \left\{ \frac{i}{2\epsilon} \left[R_{11}(\tilde{a}_1^2 - a_1^2) + 2(\tilde{a}_1 - a_1) \left(q_1 + \sum_{j=2}^d R_{1,j} b_j \right) \right] \right\} d\xi_1 d\xi_{d-1}
\end{aligned} \tag{2.15}$$

The proportionality constant is $\exp\left(-\frac{i}{\epsilon} S^\pm(t_c)\right) \sin\left(\frac{\pi\gamma}{2}\right)$ **Have the resulting integral here**

We also note that I is real symmetric and so diagonalizable

2.3 d-dimensional case

Solution of the inner most nasty integral in ξ_1 may then allow us to solve the remaining $d - 1$ dimensional integral explicitly

2.3.1 Monte Carlo Integration

(Ignore global phase factor for the moment)

We find it convenient to re-write a_{kl} in terms of complex coefficients as follows

(we will use this form in other parts of the projection section)

$$\begin{aligned}
a_{kl} &= \sin\left(\frac{\pi\gamma}{2}\right) \int_{\mathbb{R}^{d-1}} \exp\left[-\frac{1}{\epsilon} b^T I b\right] \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^{\pm}](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \times \\
&\quad \exp\left[-\frac{1}{2\epsilon} (\alpha \tilde{a}_1^2 + \bar{\alpha} a_1^2 + \beta \tilde{a}_1 + \bar{\beta} a_1 + 2q_c |\xi_1 - \nu(\xi_1)|)\right] d\xi_1 d\xi_{d-1} \\
&= \int_{\mathbb{R}^{d-1}} \exp\left[-\frac{1}{\epsilon} b^T I b\right] \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^{\pm}](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \times \\
&\quad \exp\left[-\frac{1}{2\epsilon} \left(\alpha(\nu(\xi_1) - p_1)^2 + \bar{\alpha}(\xi_1 - p'_1)^2 + \beta(\nu(\xi_1) - p_1) + \bar{\beta}(\xi_1 - p'_1) \right.\right. \\
&\quad \left.\left. + 2q_c |\xi_1 - \nu(\xi_1)|\right)\right] d\xi_1 d\xi_{d-1}
\end{aligned} \tag{2.16}$$

We spot a Gaussian term in the integral which would be suitable for numerical integration via MC.

$$\begin{aligned}
a_{kl} &= \int_{\mathbb{R}^{d-1}} \exp\left[-\frac{1}{\epsilon} b^T I b\right] \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^{\pm}](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \times \\
&\quad \exp\left[-\frac{1}{2\epsilon} \left(\alpha(\nu(\xi_1) - p_1)^2 + \beta(\nu(\xi_1) - p_1) + \bar{\beta}(\xi_1 - p'_1) + iR_{11}(\xi_1 - p'_1)^2 + 2q_c |\xi_1 - \nu(\xi_1)|\right)\right] \\
&\quad \exp\left[-\frac{I_{11}}{2\epsilon} (\xi_1 - p'_1)^2\right] d\xi_1 d\xi_{d-1}
\end{aligned} \tag{2.17}$$

where

$$\alpha = I_{11} - iR_{11} \quad \beta = -i2q_1 + 2 \sum_{j=2}^d (I_{1,j} - iR_{1,j}) b_j \tag{2.18}$$

We can use a Monte Carlo estimator so that

$$\begin{aligned}
a_{kl} = & \lim_{N \rightarrow \infty} \sum_{n=0}^N \text{sgn}(\xi_n) \Theta(\xi_n^2 - 4\delta) \left(1 + \frac{\xi_n}{\nu(\xi_n)} \right) \\
& \exp \left[-\frac{1}{2\epsilon} \left(\alpha(\nu(\xi_n) - p_1)^2 + iR_{11}(\xi_n - p'_1)^2 + 2q_c |\xi_n - \nu(\xi_n)| \right) \right] \times \\
& \int_{\mathbb{R}^{d-1}} P_k[\hat{\Pi}_{t_c}^{\pm}](\nu(\xi_n), \xi_2, \dots, \xi_d) \overline{P_l[\hat{\Pi}'](\xi_n, \xi_2, \dots, \xi_d)} \\
& \exp \left[-\frac{1}{\epsilon} (b^T I b) \right]
\end{aligned} \tag{2.19}$$

To do:

- integral is incomplete but we get the idea
- will loose efficiency of samples when checking $\Theta(\dots)$
- there is some normalisation constant missing
- inner integral should be solvable since I is diagonalizable

2.3.2 Taylor expansion + Gaussian Integrals

We will consider this approach once we have an expression for the k^{th} order polynomials. For the moment jump to the section on one dimensional integration which consists in the same approach and not much will change for this situation since we will be able to solve the remaining $d - 1$ integrals exactly.

2.3.3 One dimension - principle of stationary phase

Could think of solvin inner integral using an asymptotic expansion and then see whether the resulting $d - 1$ integral could be solved exactly...?

In this case we want to re-write the integral in a form such that the imaginary term in the exponent is explicit. For $d = 1$, the polynomials reduce to Hermite polynomials. The integral is of the form

$$\int_{\mathbb{R}} f(\xi; \delta) \exp \left[\frac{i}{2\epsilon} g(\xi; \delta) \right] d\xi \quad (2.20)$$

where

$$\begin{aligned} f(\xi; \delta) &= \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)} \right) p_k[\Pi](\nu(\xi)) p_l[\Pi'](\xi) \\ &\quad \exp \left[-\frac{1}{2\epsilon} (2I_{11}\xi_1^2 + 2\xi_1(-I_{11}p'_1 + \sum_{j=2}^d I_{1,j}b_j) + 2\nu(\xi_1)(-p_1I_{11} + \sum_{j=2}^d I_{1,j}b_j)) \right] \\ &\quad \exp \left[I_{11}(-4\delta + p_1^2 + p_1'^2) - 2(p_1 + p'_1) \left(\sum_{j=2}^d I_{1,j}b_j \right) + q_c |\xi_1 - \nu(\xi_1)| \right] \\ g(\xi; \delta) &= 2\nu(\xi_1) \left(\sum_{j=2}^d R_{1,j}b_j + q_1 - R_{11}p_1 \right) - 2\xi_1 \left(\sum_{j=2}^d R_{1,j}b_j + q_1 - R_{11}p'_1 \right) \\ &\quad R_{11}(-4\delta + p_1^2 - p_1'^2) + 2 \left(q_1 + \sum_{j=2}^d R_{1,j}b_j \right) (-p_1 + p'_1) \end{aligned} \quad (2.21)$$

If you approximate $\nu(\xi_1)$ then you would be able to say something about the frequency of the oscillations

For $d = 1$, the polynomials reduce to the Hermite polynomials We can re-write the oscillator more succintly as

$$g(\xi_1; \delta) = \alpha \nu(\xi_1) + \beta \xi_1 + \gamma$$

where

$$\begin{aligned}
\alpha &= 2(q_1 + \sum_{j=2}^d R_{1,j} b_j - R p_1) \\
\beta &= 2(-q_1 - \sum_{j=2}^d R_{1,j} b_j + R p'_1) \\
\gamma &=
\end{aligned} \tag{2.22}$$

and $g(\xi)$ has a stationary point on $(2\sqrt{\delta}, \infty)$ at ξ_1^* that solves

$$\begin{aligned}
\frac{\alpha \xi_1}{\sqrt{\xi_1^2 - 4\delta}} + \beta &= 0 \\
\Leftrightarrow \xi_1^* &= \sqrt{\frac{-4\beta^2 \delta}{\alpha^2 - \beta^2}}
\end{aligned} \tag{2.23}$$

where $\alpha^2 - \beta^2 \leq 0$ with equality when

The stationary point is non-degenerate so should be able to re-write in quadratic form via a change of variables

The frequency depends on the ration δ/ϵ Change of variables...? $f(\xi)$ is not smoothed but can be smoothed...?

$$\begin{aligned}
&\int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) g(\xi_1, \nu(\xi_1)) \times \\
&\quad \exp \left[-\frac{1}{2\epsilon} (\alpha \tilde{a}_1^2 + \bar{\alpha} a_1^2 + \beta \tilde{a}_1 + \bar{\beta} a_1 + 2q_c |\xi_1 - \nu(\xi_1)|) \right] d\xi_1
\end{aligned} \tag{2.24}$$

References

- [Hag98] George A Hagedorn. “Raising and Lowering Operators for Semi-classical Wave Packets”. In: *Annals of Physics* 269.1 (Oct. 1998), pp. 77–104. ISSN: 00034916. DOI: [10.1006/aphy.1998.5843](https://doi.org/10.1006/aphy.1998.5843). URL: <https://linkinghub.elsevier.com/retrieve/pii/S0003491698958432> (visited on 03/27/2021).
- [WF03] Xiaoqun Wang and Kai-Tai Fang. “The Effective Dimension and Quasi-Monte Carlo Integration”. In: *Journal of Complexity* 19.2 (Apr. 2003), pp. 101–124. ISSN: 0885064X. DOI: [10.1016/S0885-064X\(03\)00003-7](https://doi.org/10.1016/S0885-064X(03)00003-7). URL: <https://linkinghub.elsevier.com/retrieve/pii/S0885064X03000037> (visited on 06/02/2021).
- [HJ05] George A. Hagedorn and Alain Joye. “Determination of Non-Adiabatic Scattering Wave Functions in a Born-Oppenheimer Model”. In: *Annales Henri Poincaré* 6.5 (Oct. 2005), pp. 937–990. ISSN: 1424-0637, 1424-0661. DOI: [10.1007/s00023-005-0231-y](https://doi.org/10.1007/s00023-005-0231-y). URL: <http://link.springer.com/10.1007/s00023-005-0231-y> (visited on 03/05/2021).
- [Lub08] Christian Lubich. *From Quantum to Classical Molecular Dynamics: Reduced Models and Numerical Analysis*. In collab. with European Mathematical Society. Zurich Lectures in Advanced Mathematics. Zürich, Switzerland: European Mathematical Society, 2008. ISBN: 978-3-03719-067-8.
- [BGT09] Volker Betz, Benjamin D. Goddard, and Stefan Teufel. “Superadiabatic Transitions in Quantum Molecular Dynamics”. In: *Proceed-*

- ings of the Royal Society A: Mathematical, Physical and Engineering Sciences* 465.2111 (Nov. 8, 2009), pp. 3553–3580. ISSN: 1364-5021, 1471-2946. DOI: [10.1098/rspa.2009.0337](https://doi.org/10.1098/rspa.2009.0337). URL: <https://royalsocietypublishing.org/doi/10.1098/rspa.2009.0337> (visited on 03/28/2021).
- [FGL09] Erwan Faou, Vasile Gradinaru, and Christian Lubich. “Computing Semiclassical Quantum Dynamics with Hagedorn Wavepackets”. In: *SIAM Journal on Scientific Computing* 31.4 (Jan. 2009), pp. 3027–3041. ISSN: 1064-8275, 1095-7197. DOI: [10.1137/080729724](https://doi.org/10.1137/080729724). URL: <http://epubs.siam.org/doi/10.1137/080729724> (visited on 03/06/2021).
- [GHJ10] V. Gradinaru, G. A. Hagedorn, and A. Joye. “Tunneling Dynamics and Spawning with Adaptive Semiclassical Wave Packets”. In: *The Journal of Chemical Physics* 132.18 (May 14, 2010), p. 184108. ISSN: 0021-9606, 1089-7690. DOI: [10.1063/1.3429607](https://doi.org/10.1063/1.3429607). URL: <http://aip.scitation.org/doi/10.1063/1.3429607> (visited on 04/08/2021).
- [BGH12] Raoul Bourquin, Vasile Gradinaru, and George A. Hagedorn. “Non-Adiabatic Transitions near Avoided Crossings: Theory and Numerics”. In: *Journal of Mathematical Chemistry* 50.3 (Mar. 2012), pp. 602–619. ISSN: 0259-9791, 1572-8897. DOI: [10.1007/s10910-011-9827-2](https://doi.org/10.1007/s10910-011-9827-2). URL: <http://link.springer.com/10.1007/s10910-011-9827-2> (visited on 03/27/2021).

- [Bou17] Raoul Bourquin. “Numerical Algorithms for Semiclassical Wavepackets”. In: 2017. DOI: [10.3929/ethz-b-000183094](https://doi.org/10.3929/ethz-b-000183094).
- [BGH19] V. Betz, B. D. Goddard, and Tim Hurst. “Nonadiabatic Transitions in Multiple Dimensions”. In: *SIAM Journal on Scientific Computing* 41.5 (Jan. 2019), B1011–B1033. ISSN: 1064-8275, 1095-7197. DOI: [10.1137/18M1188756](https://doi.org/10.1137/18M1188756). URL: <https://epubs.siam.org/doi/10.1137/18M1188756> (visited on 03/28/2021).
- [BG20] Sergio Blanes and Vasile Gradinaru. “High Order Efficient Splittings for the Semiclassical Time-Dependent Schrödinger Equation”. In: *Journal of Computational Physics* 405 (Mar. 2020), p. 109157. ISSN: 00219991. DOI: [10.1016/j.jcp.2019.109157](https://doi.org/10.1016/j.jcp.2019.109157). URL: <https://linkinghub.elsevier.com/retrieve/pii/S0021999119308629> (visited on 05/20/2021).
- [FLR20] Clotilde Fermanian-Kammerer, Caroline Lasser, and Didier Robert. *Propagation of Wave Packets for Systems Presenting Codimension 1 Crossings*. Aug. 25, 2020. arXiv: [2001.07484](https://arxiv.org/abs/2001.07484) [math-ph]. URL: <http://arxiv.org/abs/2001.07484> (visited on 06/28/2021).

Appendices

Some Appendix

A Transmitted Wavepacket

[./main_hagedorn.tex]subfilesamsmath

A.1 Transmitted wavepacket

We can now apply the transmission formula of equation ... for the two cases of constant and non-constant eigenvalues

A.1.1 Constant eigenvalues

The transmitted wavepacket is given by [BGT09]

$$\begin{aligned}
\hat{\psi}^\mp(\xi, t_c) &= \exp\left(-\frac{i}{\epsilon}S^\pm(t_c)\right) \sin\left(\frac{\pi\gamma}{2}\right) \times \\
&\quad \text{sgn}(\xi_1)\Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) \exp\left(-\frac{q_c}{\epsilon}|\xi_1 - \nu(\xi_1)|\right) \times \\
&\quad \sum_{k \in \mathcal{K}} c_k^\pm(t_c) \varphi_k^\epsilon[\hat{\Pi}^\pm(t_c)](\nu(\xi_1), \xi_2, \dots, \xi_d) \\
&= \exp\left(-\frac{i}{\epsilon}S^\pm(t_c)\right) \sin\left(\frac{\pi\gamma}{2}\right) \times \\
&\quad \text{sgn}(\xi_1)\Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) \exp\left(-\frac{q_c}{\epsilon}|\xi_1 - \nu(\xi_1)|\right) \times \\
&\quad \varphi_0^\epsilon[\hat{\Pi}^\pm(t_c)](\nu(\xi_1), \xi_2, \dots, \xi_d) \sum_{k \in \mathcal{K}} c_k^\pm(t_c) p_k(\nu(\xi_1), \xi_2, \dots, \xi_d)
\end{aligned} \tag{A.1}$$

- At least for Gaussian wavepackets with "large enough" momentum, the effect of the cut-off function should be "negligible" as a result of the exponential decay. Is this still the case as the order of the Hagedorn wavepacket increases...? The variance of φ_k^ϵ scales with k ...

A.1.2 Non-constant + tilted crossings

The transmitted wavepacket is given by [BGH19]

$$\begin{aligned} \hat{\psi}^\mp(\xi, t_c) = & \exp\left(-\frac{i}{\epsilon} S^\pm(t_c)\right) \times \\ & \Theta(\xi_1^2 - 4\delta) \frac{\nu(\xi_1) + \xi_1}{2|\nu(\xi_1)|} \exp\left(-\frac{\tau_c}{2\delta\epsilon} |\xi_1 - \nu(\xi_1)|\right) \exp\left(-\frac{i\tau_r}{2\delta\epsilon} (\xi_1 - \nu(\xi_1))\right) \times \\ & \sum_{k \in \mathcal{K}} c_k^\pm(t_c) \varphi_k^\epsilon[\hat{\Pi}^\pm(t_c)](\nu(\xi_1), \xi_2, \dots, \xi_d) \end{aligned} \tag{A.2}$$

Is there an actual mismatch between the formulas for the constant eigenvalue and the general case? That is, why does the sin prefactor disappear for the general case? Also, I would think the formula is invariant to the direction of the jump between levels..? The derivation was done from up to down but intuitively thing should not change...?

B Proof of concept for choosing different parameter set in the projection

Given that we can obtain the results numerically, it is questionable whether this is needed or not Here we try to determine/justify whether it is at all numerically convenient to change the parameters $\hat{\Pi}$ for the transmitted wavepacket and thus reduce the number of coefficients needed to represent the wavepacket in the Hagedorn basis. This is motivated by the knowledge we have regarding the momentum shift.

Since this is only a proof of concept, let us consider the simplest complex

Gaussian wavepacket for our incoming wavepacket with $P = 1, Q = i$. Furthermore, we are interested in the dependence of the Fourier coefficients with respect to a relative shift δ in the mean momentum of the transmitted wavepacket and so we find it convenient to consider deviations δ from $p = 0$. We also consider the location of the avoided crossing to be at $q = 0$ presumably for the dynamics one can always relocate the avoided crossing to zero by translating the center of mass - however this would then now work for on the fly dynamics? and thus removing oscillatory terms from the integral. More precisely we are interested in computing $c_k(\delta) = \langle \varphi_0^\epsilon[\delta, 0, 1, i](\xi), \varphi_k^\epsilon[0, 0, 1, i](\xi) \rangle$. The φ_k 's for this choice of parameters can be generated from the recurrence relation (1.3) in momentum space If I make an ansatz about the polynomial gaussian relation, it should give a recurrence relation for the polynomials only I would guess that one can do this by first doing the translation to $p = 0$ and then re-introducing the translation after the tabulation

$$\begin{aligned}
\varphi_0^\epsilon(\xi) &= (\pi\epsilon)^{-\frac{1}{4}} \exp\left(-\frac{\xi^2}{2\epsilon}\right) \\
\varphi_{k+1}^\epsilon &= \sqrt{\frac{2}{\epsilon(k+1)}} \xi \varphi_k^\epsilon - \sqrt{\frac{k}{k+1}} \varphi_{k-1}^\epsilon \\
&= \dots \\
\varphi_1^\epsilon(\xi) &= \sqrt{\frac{2}{\epsilon}} \xi \varphi_0^\epsilon, \quad \varphi_2^\epsilon(\xi) = \left(\sqrt{\frac{2}{\epsilon^2}} \xi^2 - \frac{1}{\sqrt{2}}\right) \varphi_0^\epsilon \\
\varphi_3^\epsilon(\xi) &= \left(\frac{2}{\sqrt{3}\epsilon^3} \xi^3 - \frac{3}{\sqrt{3}\epsilon} \xi\right) \varphi_0^\epsilon, \quad \varphi_4^\epsilon(\xi) = \left(\frac{1}{\epsilon^2} \sqrt{\frac{2}{3}} \xi^4 - \frac{\sqrt{6}}{\epsilon} \xi^2 + \sqrt{\frac{3}{8}}\right) \varphi_0^\epsilon
\end{aligned} \tag{B.1}$$

If we let $a_n(\delta) = \langle \varphi_0^\epsilon[\delta, 0, 1, i](\xi), \xi^n \varphi_0^\epsilon[0, 0, 1, i](\xi) \rangle$ we can then re-write the coefficients more succinctly as

$$\begin{aligned} c_1(\delta) &= \sqrt{\frac{2}{\epsilon}} a_1(\delta), \quad c_2(\delta) = \sqrt{\frac{2}{\epsilon^2}} a_2(\delta) - \frac{1}{\sqrt{2}} a_0(\delta) \\ c_3(\delta) &= \frac{2}{\sqrt{3\epsilon^3}} a_3(\delta) - \frac{3}{\sqrt{3\epsilon}} a_1(\delta), \quad c_4(\delta) = \frac{1}{\epsilon^2} \sqrt{\frac{2}{3}} a_4(\delta) - \frac{\sqrt{6}}{\epsilon} a_2(\delta) + \sqrt{\frac{3}{8}} \end{aligned} \quad (\text{B.2})$$

The polynomials **need to check...** would they have had complex coefficients if we had a complex valued P , in higher dimension are they linear combinations of monomials or they have cross terms? are the Hermite polynomials but I still need to get the coefficients right...? Given the form the wavepackets we only need to consider the integral for a monomial ξ^n as a prefactor,

$$\begin{aligned} a_n(\delta) &= (\pi\epsilon)^{-\frac{1}{2}} \int_{\mathbb{R}} \xi^n \exp \left[-\frac{1}{2\epsilon} (\xi^2 + (\xi - \delta)^2) \right] d\xi \\ &= (\pi\epsilon)^{-\frac{1}{2}} \int_{\mathbb{R}} \xi^n \exp \left[-\frac{1}{2\epsilon} (2\xi^2 - 2\xi\delta + \delta^2) \right] d\xi \\ &= (\pi\epsilon)^{-\frac{1}{2}} \exp \left[-\frac{\delta^2}{2\epsilon} \right] \int_{\mathbb{R}} \xi^n \exp \left[-\frac{1}{\epsilon} (\xi^2 - \xi\delta) \right] d\xi \\ &= (\pi\epsilon)^{-\frac{1}{2}} \exp \left[-\frac{\delta^2}{2\epsilon} \right] \int_{\mathbb{R}} \epsilon^n \frac{d^n}{d\delta^n} \exp \left[-\frac{1}{\epsilon} (\xi^2 - \xi\delta) \right] d\xi \\ &= (\pi\epsilon)^{-\frac{1}{2}} \exp \left[-\frac{\delta^2}{2\epsilon} \right] \epsilon^n \frac{d^n}{d\delta^n} \int_{\mathbb{R}} \exp \left[-\frac{1}{\epsilon} (\xi^2 - \xi\delta) \right] d\xi \\ &= (\pi\epsilon)^{-\frac{1}{2}} (\pi\epsilon)^{\frac{1}{2}} \epsilon^n \exp \left[-\frac{\delta^2}{2\epsilon} \right] \frac{d^n}{d\delta^n} \exp \left[\frac{\delta^2}{4\epsilon} \right] \\ &= \epsilon^n \exp \left[-\frac{\delta^2}{2\epsilon} \right] \frac{d^n}{d\delta^n} \exp \left[-\frac{(i(2\epsilon)^{-\frac{1}{2}}\delta)^2}{2} \right] \\ &= \epsilon^n \exp \left[-\frac{\delta^2}{2\epsilon} \right] \exp \left[\frac{\delta^2}{4\epsilon} \right] (-i)^n (2\epsilon)^{-n/2} H_n \left(i \frac{\delta}{\sqrt{2\epsilon}} \right) \\ &= \epsilon^n \exp \left[-\frac{\delta^2}{4\epsilon} \right] (-i)^n (2\epsilon)^{-n/2} H_n \left(i \frac{\delta}{\sqrt{2\epsilon}} \right) \end{aligned}$$

(B.3)

where we have used the a known identity for the Gaussian integral and the probabilists' Hermite polynomials $H_e(x) = \dots$

- $a_0(\delta) = c_0(\delta)$
- $a_0(0) = c_0(0) = 1$ as expected
- Checked the first few but the following plot will also serve as a sanity check
- This makes clearer Hagedorn and Joye's claim that the leading order is a Gaussian
- If we consider the c_n 's it does seem as if the dependence on the higher order basis vectors is in ascending order as we increase δ up to a certain point
- It is intuitively clear why it depends on the ratio between the momentum shift and the variance $\mathcal{O}(\epsilon)$
- The coefficients are real because we have chosen $q = 0$
- We know that the mean momentum shift is at least $2\sqrt{\delta}$ from conservation of energy

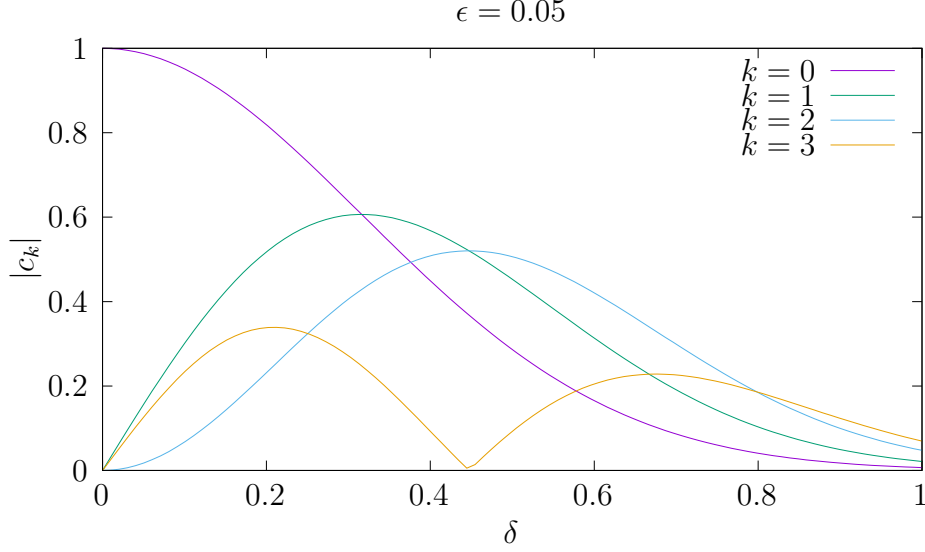


Figure 3: c_k is the coefficient corresponding to the projected Gaussian onto the k^{th} Hagedorn wavepacket.

2.1 One dimensional case

(Ignore global phase factor for the moment) In one dimension the a_{kl} becomes

$$\begin{aligned}
 a_{kl} = & \sin\left(\frac{\pi\gamma}{2}\right) \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) P_k[\hat{\Pi}_{t_c}^{\pm}](\tilde{\xi}) \overline{P_l[\hat{\Pi}'](\xi)} \times \\
 & \exp\left[-\frac{1}{2\epsilon} \left(\alpha(\nu(\xi_1) - p_1)^2 + \bar{\alpha}(\xi_1 - p'_1)^2 + \beta(\nu(\xi_1) - p_1) + \bar{\beta}(\xi_1 - p'_1) \right. \right. \\
 & \left. \left. + 2q_c|\xi_1 - \nu(\xi_1)|\right)\right] d\xi_1
 \end{aligned} \tag{2.4}$$

where

$$\alpha = I_{11} - iR_{11} \quad \beta = -i2q_1 \tag{2.5}$$

and (see attached written notes)

$$\begin{aligned}
P_k[\Pi](x)\varphi_0 &= \sqrt{k!} \left(\frac{i}{2\epsilon}\right)^k \sum_{\substack{j=0 \\ j \equiv k \pmod{2}}}^k \frac{(i\epsilon Q^* P^*/2)^{(k-j)/2}}{j! \left(\frac{k-j}{2}\right)!} \times \\
&\times \left(\sum_{r=0}^j \binom{j}{r} (P^*(x-q) + Q^*p)^r (Q^*i\epsilon d_x)^{j-r} \right) \varphi_0
\end{aligned} \tag{2.6}$$

so that

$$\begin{aligned}
P_k[\Pi](x) &= \sqrt{k!} \left(\frac{i}{2\epsilon}\right)^k \sum_{\substack{j=0 \\ j \equiv k \pmod{2}}}^k \frac{(i\epsilon Q^* P^*/2)^{(k-j)/2}}{j! \left(\frac{k-j}{2}\right)!} \times \\
&\times \left(\sum_{r=0}^j \binom{j}{r} (P^*(x-q) + Q^*p)^r (Q^*i\epsilon)^{j-r} \sum_{l=0}^{\lfloor j-r \rfloor} \sum_{s=0}^{j-r-2l} \binom{j-r-2l}{s} \right) \\
&\times (i\epsilon P Q^{-1}(x-q))^{j-r-2l-s} \left(\frac{i}{\epsilon} p\right)^s (i\epsilon P Q^{-1})^l \frac{(j-r)!}{l!(j-r-2l)!} 2^{-l}
\end{aligned} \tag{2.7}$$

- there is an extra parity condition on j which you are missing
- Polynomial has degree k and so we would need at most $k+1$ terms in the summation. So how can we go about simplifying the expression further, perhaps using properties of P, Q from Lemma 1.1 and a bit of manipulation
- The expression can most likely be simplified further - for the moment we will just going to use it to see if it is at least correct
- verify expression reduces to Hermite polynomial for particular parameter values

- Plot result from expression against one from recurrence relation
- Start by considering $k = 0$ for ease so only one substitution
- You should be able to evaluate the second term recursively

Before attempting any simplification we will start by considering the a_{0l} 's with $\hat{\Pi}' = \{P = 1, Q = i, q = 0, p = p'\}$ for which

Example

In certain sense we can already comment on the dependence of the ... based on the proof concept in one of previous sections

$$\begin{aligned}
P_l[\hat{\Pi}'](\xi) &= \sqrt{k!} \left(\frac{i}{2\epsilon} \right)^k \sum_{\substack{j=0 \\ j \equiv k \pmod{2}}}^k \frac{(\epsilon/2)^{(k-j)/2}}{j! \left(\frac{k-j}{2} \right)!} \times \sum_{r=0}^j \binom{j}{r} \epsilon^j i^j \left(-\frac{1}{\epsilon} (\xi - p') \right)^r \\
&\times \left(\sum_{l=0}^{\lfloor j-r \rfloor} \left(-\frac{1}{\epsilon} (\xi - p') \right)^{j-r-2l} \left(-\frac{1}{\epsilon} \right)^l 2^{-l} \frac{(j-r)!}{l! (j-r-2l)!} \right) \\
&= \sqrt{k!} \left(\frac{1}{2\epsilon} \right)^k \sum_{\substack{j=0 \\ j \equiv k \pmod{2}}}^k \frac{(\epsilon/2)^{(k-j)/2}}{\left(\frac{k-j}{2} \right)!} \epsilon^j i^{j+k} \times \sum_{r=0}^j \frac{1}{r!} \\
&\times \sum_{l=0}^{\lfloor j-r \rfloor} (-2\epsilon)^{-l} \frac{1}{l! (j-r-2l)!} \left(-\frac{1}{\epsilon} (\xi - p') \right)^{j-2l}
\end{aligned} \tag{2.8}$$

Perhaps it is easier to consider the cases where k is even or odd

The i dependence cancels since k, j have the same parity

since this should reduce to the expression for the Hermite polynomials except

some ϵ With regards to the integral, let us first consider the case $k = 0$

$$a_{0l} = \int_{\mathbb{R}} \text{sgn}(\xi_1) \Theta(\xi_1^2 - 4\delta) \left(1 + \frac{\xi_1}{\nu(\xi_1)}\right) \overline{P_l[\hat{\Pi}'](\xi)} \times \exp \left[-\frac{1}{2\epsilon} \left((\nu(\xi_1) - p_1)^2 + (\xi_1 - p'_1)^2 + 2q_c |\xi_1 - \nu(\xi_1)| \right) \right] d\xi_1 \quad (2.9)$$

As we have argued the polynomials will be real valued in this case.

If we Taylor expand $\tilde{a}_1 = \nu(\xi_1) - p_1$ about p'_1 to first order we have,

$$\begin{aligned} \tilde{a}_1 &= \text{sgn}(\xi_1) \sqrt{\xi_1^2 - 4\delta} - p_1 = \text{sgn}(\xi_1) \sum_{n=0} \dots \\ &= \text{sgn}(\xi_1) \left(\sqrt{p_1'^2 - 4\delta} - p_1 + \frac{p'_1}{\sqrt{p_1'^2 - 4\delta}} (\xi_1 - p'_1) + \dots \right) \end{aligned}$$

i.e. of the form $\alpha + \beta(\xi_1 - p'_1)$ the remainder can also be formulated as an integral - which one is most convenient and assume the wavepacket's momentum is large and positive . Further, with expanding $\nu(\xi_1)$ using the binomial theorem

$$\begin{aligned} \nu(\xi_1) &= \text{sgn}(\xi_1) \sqrt{\xi_1^2 - 4\delta} = \xi_1 \sqrt{1 - \frac{4\delta}{\xi_1^2}} = \xi_1 \sum_{n=0}^{\infty} \dots \\ &= \xi_1 \left(1 - \frac{2\delta}{\xi_1^2} - \frac{2\delta^2}{\xi_1^4} + \dots \right) = \xi_1 - \frac{2\delta}{\xi_1} - \frac{2\delta^2}{\xi_1^3} + \dots \end{aligned}$$

yields - “contribution to integral comes from positive axis” - **is the integration known over half domain**

$$\begin{aligned}
a_{0l} &\approx 2 \exp \left[-\frac{1}{2\epsilon}(\alpha^2 + p'_1 - p_1 - \alpha) \right] \int_{\mathbb{R}} \overline{P_l[\hat{\Pi}'](\xi)} \times \\
&\quad \exp \left[-\frac{1}{2\epsilon} \left((\beta^2 + 1)(\xi_1 - p'_1)^2 + (2\alpha\beta + \beta + 1)(\xi_1 - p'_1) \right) \right] d\xi_1 \\
&= 2 \exp \left[-\frac{1}{2\epsilon}(\alpha^2 + p'_1 - p_1 - \alpha) \right] \int_{\mathbb{R}} \overline{P_l[\hat{\Pi}'](u + p'_1)} \times \\
&\quad \exp \left[-\frac{1}{2\epsilon}(\gamma u^2 + \zeta u) \right] du \\
&= 2 \exp \left[-\frac{1}{2\epsilon}(\alpha^2 + p'_1 - p_1 - \alpha) \right] \sqrt{k!} \left(\frac{1}{2\epsilon} \right)^k \sum_{\substack{j=0 \\ j \equiv k \pmod{2}}}^k \frac{(\epsilon/2)^{(k-j)/2}}{\left(\frac{k-j}{2}\right)!} \epsilon^j i^{j+k} \times \sum_{r=0}^j \frac{1}{r!} \\
&\quad \times \sum_{l=0}^{\lfloor j-r \rfloor} (-2\epsilon)^{-l} \frac{1}{l!(j-r-2l)!} \left(-\frac{1}{\epsilon} \right)^{j-2l} \int_{\mathbb{R}} u^{j-2l} \exp \left[-\frac{1}{2\epsilon}(\gamma u^2 + \zeta u) \right] du
\end{aligned} \tag{2.10}$$

The integral above can be solved as

$$\begin{aligned}
&\int_{\mathbb{R}} -(2\epsilon)^{j-2l} \frac{d^{j-2l}}{d\zeta} \exp \left[-\frac{1}{2\epsilon}(\gamma u^2 + \zeta u) \right] du \\
&= -(2\epsilon)^{j-2l} \frac{d^{j-2l}}{d\zeta} \int_{\mathbb{R}} \exp \left[-\frac{1}{2\epsilon}(\gamma u^2 + \zeta u) \right] du \\
&= -(2\epsilon)^{j-2l} \sqrt{\frac{\pi}{\gamma}} \frac{d^{j-2l}}{d\zeta} \exp \left[\frac{\zeta^2}{4\gamma} \right]
\end{aligned} \tag{2.11}$$

and the derivatives once again give rise to Hermite polynomials

I still need to get round to test it

I can always consider smoothing the cut-off function with a bump function and take the limit outside...?

Change of variables – Asymptotic expansion via stationary phase principle This would work only in 1d as there is no stationary point in $d \geq 1$

Changing range of integration

Consider

$$\int_{2\sqrt{\delta}}^{\infty} f(\xi) \exp \left[-\frac{i}{2\epsilon} () \right] d\xi \quad (2.12)$$

3 Transmitted wavepacket recurrence relation derivation

We have

$$\begin{aligned} & \left(\langle (\tilde{\xi} - \mathbf{q})_j^p f(\tilde{\xi}) \tilde{\varphi}_{k+\langle j \rangle}, \varphi_l \rangle \right)_{j=1}^d = \\ & = \left(\sum_{i=1}^d \mathbf{A}_{j,i} \langle (\tilde{\xi} - \mathbf{q})_j^p (\tilde{\xi} - \mathbf{q})_i f(\tilde{\xi}) \tilde{\varphi}_k, \varphi_l \rangle \right)_{j=1}^d - \left(\sum_{i=1}^d \mathbf{B}_{j,i} \langle (\tilde{\xi} - \mathbf{q})_j^p f(\tilde{\xi}) \tilde{\varphi}_{k-\langle i \rangle}, \varphi_l \rangle \right)_{j=1}^d \end{aligned} \quad (3.1)$$

and then proceeding in a similar manner as before for the higher order cross terms. Now, let k be fixed and l vary giving

$$\begin{aligned} & \left(\langle f(\tilde{\xi}) \tilde{\varphi}_k, (\xi - \mathbf{q})_j^p \varphi_{l+\langle j \rangle} \rangle \right)_{j=1}^d = \\ & = \left(\sum_{i=1}^d \mathbf{A}_{j,i} \langle f(\tilde{\xi}) \tilde{\varphi}_k, (\xi - \mathbf{q})_j^p (\xi - \mathbf{q})_i \varphi_l \rangle \right)_{j=1}^d - \left(\sum_{i=1}^d \mathbf{B}_{j,i} \langle f(\tilde{\xi}) \tilde{\varphi}_k, (\xi - \mathbf{q})_j^p \varphi_{l-\langle i \rangle} \rangle \right)_{j=1}^d \end{aligned} \quad (3.2)$$

and then for the higher moments similarly.