An efficient *Space* and *Time* saving algorithm for solving Time Dependent Schrödinger Equation using the (t,t') method

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Certificate of Examination

This is to certify that the dissertation titled **An efficient "Space" and "Time" saving algorithm for solving Time Dependent Schrödinger Equation using the (t,t') method** submitted by **Mr. Alkit Gugalia (Reg. No. MS13143)** for the partial fulfillment of BS-MS dual degree programme of the Institute, has been examined by the thesis committee duly appointed by the Institute. The committee finds the work done by the candidate satisfactory and recommends that the report be accepted.

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Dated: April 16, 2018

Declaration

The work presented in this dissertation has been carried out by me with Dr. P. Balanarayan at the Indian Institute of Science Education and Research Mohali. This work has not been submitted in part or in full for a degree, a diploma, or a fellowship to any other university or institute. Whenever contributions of others are involved, every effort is made to indicate this clearly, with due acknowledgement of collaborative research and discussions. This thesis is a bonafide record of original work done by me and all sources listed within have been detailed in the bibliography.

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In my capacity as the supervisor of the candidate's project work, I certify that the above statements by the candidate are true to the best of my knowledge.

Dr. P. Balanarayan
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"If you thought that science was certain - well, that is just an error on your part."

Richard Feynman

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Abbreviations

a.u.	**a**tomic **u**nits
BEC	**B**ose **E**instein **C**ondensate
GPE	**G**ross **P**itaevskii **E**quation
KHA	**K**ramers **H**enneberger **A**pproximation
MCTDHB	**M**ulti **C**onfigurational **T**ime **D**ependent **H**artree **F**ock for **B**osons
TDSE	**T**ime **D**ependent **S**chrödinger **E**quation

Physical Constants

Charge of an electron	e	$=$	$1.602\ 176\ 62 \times 10^{-19}\ C$
Reduced Planck's constant	$\hbar$	$=$	$1.054\ 571\ 80 \times 10^{-34}\ Js$
Boltzmann constant	K_B	$=$	$1.380\ 6452 \times 10^23\ m^2kgs^2K^1$

Dedicated to my Family and Friends.

Abstract

The first part of the work focuses on the Merging dynamics of the Bose-Einstein Condensates in the presence of oscillating trap potentials. The dynamics is studied by using the Gross-Pitaevskii equation and the MCTDHB theory in the KH frame of reference. In the second part of the work, an algorithm is proposed which uses analytic expressions for block diagonalization and other trnasformations involved in the (t, t') method, thus reducing the memory storage and computational time required for performing such heavy calculations.

Chapter 1

Introduction

1.1 Bose-Einstein Condensates

Bose-Einstein Condensation is a phenomenon observed at really low temperatures when a large fraction of particles in a bosonic gas occupy the lowest energy quantum state.

Bosons are particles with integral spins and they do not follow Pauli's Exclusion principle. The uncertainty in position of a thermal momentum distribution of a particle of mass m is given by the de Broglie wavelength λ_{DB} [1].λ_{DB} can be approximated as the average de Broglie wavelength of a particle of mass m kept at a fixed temperature T. It is given by the formula:

$$\lambda_{DB} = \left(\frac{2\pi\hbar}{mK_BT} \right)^{\frac{1}{2}} \tag{1.1}$$

At very low temperatures, the interatomic separation in a gas of bosonic particles starts becoming less than λ_{DB}:

$$\frac{V}{N\lambda_{DB}^3} \leq 1 \tag{1.2}$$

where V is the volume of the gas and N is the number of particles.

When the atoms are cooled below a critical temperature T_c, the quantum mechanical wave-packets of these particles interfere with each other and they go through a process of phase transition to form the Bose-Einstein condensates[1].

1.2 Historical background

In 1925, Albert Einstein predicted a new state of matter, the Bose-Einstein Condensates. He made this prediction on the basis of a method introduced to him by S.N. Bose to derive the Planck's radiation law in a new way. Einstein extended this idea to non-interacting atoms and immediately realized that there should be a temperature below which a large fraction of the bosonic atoms would condense into the lowest energy ground state and thus form the Bose-Einstein Condensates[1]. In the mid 1930s, Fritz London[3] and Laszlo Tisza[4] put forward the possibility of the presence of the BECs being responsible for the superfluidity in liquid $^4\mathbf{He}$. In the 1980s, laser cooling was discovered which meant ultra low temperatures were now achievable and in 1995, the first BECs were observed in a system of dilute atomic alkali gases[2]. Cornell, Wieman, and Ketterle shared the 2001 Nobel prize for preparing the first BECs and studying their properties[6] [7]. Anthony J. Leggett in his review of the BECs[5] says, "Perhaps the single aspect of BEC systems that makes them most fascinating is best illustrated by the cover of Science magazine of December 22, 1995, in which the Bose condensate is declared "molecule of the year" and pictured as a platoon of soldiers marching in lockstep: every atom in the condensate must behave in exactly the same way, and this has the consequence, inter alia, that effects which are so small as to be essentially invisible at the level of a single atom may be spectacularly amplified." BECs display superconductivity and superfluidity which are prime examples of showing quantum properties on a macroscopic scale.

1.3 Formation of the BECs

The first requirement to achieve a sustained Bose-Einstein condensate is a Bosonic gas of extremely low density $10^{14} cm^{-3}$) [2]. The purpose of such low densities is to delay the process of three body collisions. The three body collisions are responsible for chemical equilibrium which, on cooling, will transform the gas into the more stable solid state. Low densities will assure that two body collisions reach kinetic equilibrium before chemical equilibrium and thus facilitate the formation of Bose-Einstein Condensation [2]. But such low densities will demand an extremely low temperature for the phase transition to occur. Thus, the cooling process must be capable of ensuring temperature in the micro to nano kelvin range. [1] This cooling is initialized by the very well know laser cooling techniques. When the gas is subjected to a laser, the scattered photons are higher in energy as compared to the incident laser field. The excess energy comes from the atoms which, as a result, start cooling down. The cooled atoms are then magnetically trapped [1]. It was observed in early experiments that the gas would stick on the surface of the container when it is cold. Harold Hess, in 1986, came up with idea of magnetic trapping which would be like a ”surfaceless container” for BEC [8]. The magnetically trapped gas is then subject to forced evaporative cooling in which the higher energetic atoms are forced out of the trap by reducing its depth. The atoms which remain adapt to their new environment and form the metastable Bose-Einstein Condensates.

1.4 Objectives and Overview

The work presented in this thesis is divided into two parts.

The first part of the thesis focuses on the study of the merging of two fragmented Bose-Einstein Condensates in the presence of an oscillating field. The trapping of BECs is one of the most fascinating phenomenon in this field[5]. R. Dum and co-workers[12], in 1998, theoretically showed the splitting dynamics of BECs in the Kramers-Hanneberger frame of reference[7]. The initial wavepacket was bound in a Harmonic Potential and the potential trap was shaken periodically to observe the fragmentation of the condensates. Reversing the Mechanism, and starting with the wavepackets of the BECs in a double well potential which experiences the oscillations of a highly intense laser. The BECs, due to this high frequency and high intensity laser reside in a potential which is effectively a single well potential. The study of this phenomenon helps develop an understanding of how the reactions of atoms can take place in intense laser fields. Two atoms are put in a trap and the trap is shaken vigorously to achieve a molecule. An analogue of the Schr*ö*dinger equation for bosons is the Gross-Pitaevskii equation. It is derived in the mean field approximation keeping in mind that the BECs have really low density. It is also assumed that all the bosons lye in the ground state of the system. The Multiconfigurational Time Dependent Hartree Method for Bosons[11] overcomes the second approximation by including the effects of the higher excited states. The merging dynamis is studied for optimal KH parameters which are calculated for a symmetric double well potential.

The second part of the thesis focuses on methods for solving the Time Dependent Schr*ö*dinger equation with time dependent hamiltonian which are encountered in the study of the interaction of matter with fields. U. Peskin and N. Moiseyev[12], proposed a brilliant method for solving the TDSE with time dependent hamiltonians which is known as the (t,t') algorithm. But, the computational complexity of this method is the need to store a floquet matrix[15] which is infinite dimensional in the space co-ordinate and one of the time co-ordinates. The algorithm proposed deals with this complexity of this matrix in efficient ways described in Chapter 3. The algorithm could form the basis of creating an algorithm which could be used to study phenomenon in the presence of multiple chromatic lasers. There is a possibility of extending it in multi dimensions as no storage space is required.

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Chapter 2

Theoretical analysis of Merging of BECs

2.1 Many Body Hamiltonian and The Gross-Pitaevskii equation

The many body hamiltonian for a system of N spinless bosonic particles is[1]:

$$\hat{H} = \sum_{i=1}^{N} \left(\frac{p_i^2}{2m} + V_0(\vec{r}_i) \right) + \frac{1}{2} \sum_{i=1}^{N} \sum_{j \neq i}^{N} V_{int}(|\vec{r}_i - \vec{r}_j|) \tag{2.1}$$

where the first term in the hamiltonian, $\frac{p_i^2}{2m}$, is the kinetic energy of the ith particle. The next term, V_0, is the external trapping potential and the final term is the inter particle interaction between the ith and the jth particles. As BECs are purely a product of the Bose-Einstein statistics, the interaction term is not necessary for explaining the process of condensation, but it is useful for explaining many of the properties associated with the condensates. This interaction term prompts us to derive a special equation for explaining the behaviour of BECs known as the Gross-Pitaevskii equation[1]. As mentioned above, any number of Bosons, unlike fermions, can occupy the same quantum state and in the limit of temperature T going to zero, almost all of the Bosons are found in the lowest energy state of the system. The eigenvectors corresponding to the lowest energy state or the ground state can be obtained by minimizing the free energy functional:[1]

$$F = E - \mu N \tag{2.2}$$

where

$$E[\Psi(\vec{r})] = \frac{\langle \Psi(\vec{r})|\hat{H}|\Psi(\vec{r})\rangle}{\langle \Psi(\vec{r})|\Psi(\vec{r})\rangle} \tag{2.3}$$

and μ is a lagrange multiplier also known as the chemical potential. Here, the variational parameter is the many body wavefunction $\Psi(\vec{r})$. The simplest form of the wavefunction can be taken to be the Hatree Fock ansatz, i.e., the product of the single particle ground states. It is also known as the Gross-Pitaevskii ansatz in the limit of T going to zero. This approximation is known as the "mean field approximation". The "mean field approximation", made for dilute systems, states that the effect felt by any given particle is the single averaged effect of all the other particles. The N-particle tensor product wavefunction is taken as:

$$|\Psi(\vec{r})\rangle = |\psi_1(\vec{r})\rangle|\psi_2(\vec{r})\rangle...|\psi_N(\vec{r})\rangle \tag{2.4}$$

where $\psi_i(\vec{r})$ is the single particle ground state wavefunction. When the normalization $\langle \Psi(\vec{r})|\Psi(\vec{r})\rangle = 1$ is considered, equation (1.9) reduces to:[1]

$$F(\Psi) = \langle \Psi(\vec{r})|\hat{H}|\Psi(\vec{r})\rangle - \mu N\langle \Psi(\vec{r})|\Psi(\vec{r})\rangle \tag{2.5}$$

Calculating each term of $\langle \Psi(\vec{r})|\Psi(\vec{r})\rangle$: The Kinetic Energy term ($\hbar = 1$):

$$\langle \Psi(\vec{r})|\sum_{i=1}^{N}\frac{p_i^2}{2m}|\Psi(\vec{r})\rangle = \langle \Psi(\vec{r})|\sum_{i=1}^{N}-\frac{\nabla_i^2}{2m}|\Psi(\vec{r})\rangle \tag{2.6}$$

$$= -\frac{1}{2m}\sum_{i=1}^{N}\iint..\int \psi^*(\vec{r}_1)\psi^*(\vec{r}_2)..\psi^*(\vec{r}_i)..\psi^*(\vec{r}_N) \tag{2.7}$$

$$\nabla_i^2\psi(\vec{r}_1)\psi(\vec{r}_2)..\psi(\vec{r}_i)..\psi(\vec{r}_N)d\vec{r}_1 d\vec{r}_2..d\vec{r}_i..d\vec{r}_N$$

As all the particles are identical, equation (1.14) can be written as[1]

$$\big\langle\Psi(\vec{r})\big|\sum_{i=1}^{N}\frac{p_i^2}{2m}\big|\Psi(\vec{r})\big\rangle = -\frac{N}{2m}\int\psi^*(\vec{r})\nabla^2\psi(\vec{r})d\vec{r} \tag{2.8}$$

Similarly, the potential energy term:[1]

$$\big\langle\Psi(\vec{r})\big|\sum_{i=1}^{N}V_0(\vec{r}_i)\big|\Psi(\vec{r})\big\rangle = N\int\psi^*(\vec{r})V_0\psi(\vec{r})d\vec{r} \tag{2.9}$$

The interaction term is a two particle operator, so the total number of permutation of coordinates will be $\frac{N(N-1)}{2}$. The interaction term is written as:[1]

$$\big\langle\Psi(\vec{r})\big|\frac{1}{2}\sum_{i=1}^{N}\sum_{j\neq i}^{N}V_{int}(|\vec{r}_i-\vec{r}_j|)\big|\Psi(\vec{r})\big\rangle = \frac{N(N-1)}{2}\int d\vec{r}\int\psi^*(\vec{r})\psi^*(\vec{r}^{'})V_{int}(|\vec{r}-\vec{r}^{'}|)\psi(\vec{r})\psi(\vec{r}^{'})d\vec{r}^{'} \tag{2.10}$$

and

$$\mu\big\langle\Psi(\vec{r})\big|\Psi(\vec{r})\big\rangle = \mu\Big(\int\psi^*(\vec{r})\psi(\vec{r})d\vec{r}\Big)^N \tag{2.11}$$

The functional $F[\psi(\vec{r})]$ can be minimized with respect to the variational parameter $\psi^*(\vec{r})$:[1]

$$\frac{\delta F}{\delta\psi^*} = 0 \tag{2.12}$$

The terms are as follows:[1]

$$\frac{\delta\big\langle\Psi(\vec{r})\big|\sum_{i=1}^{N}\frac{p_i^2}{2m}\big|\Psi(\vec{r})\big\rangle}{\delta\psi^*} = -N\frac{1}{2m}\int\delta\psi^*(\vec{r})\nabla^2\psi(\vec{r})d\vec{r} \tag{2.13}$$

$$\frac{\delta\big\langle\Psi(\vec{r})\big|\sum_{i=1}^{N}V_0(\vec{r}_i)\big|\Psi(\vec{r})\big\rangle}{\delta\psi^*} = N\int\delta\psi^*(\vec{r})V_0(\vec{r})\psi(\vec{r})d\vec{r} \tag{2.14}$$

$$\begin{aligned}&\frac{\big\langle\delta\Psi(\vec{r})\big|\frac{1}{2}\sum_{i=1}^{N}\sum_{j\neq i}^{N}V_{int}(|\vec{r}_i-\vec{r}_j|)\big|\Psi(\vec{r})\big\rangle}{\delta\psi^*} \\ &= N(N-1)\int\delta\psi^*(\vec{r})\Big(\int|\psi(\vec{r})|^2V_{int}(|\vec{r}-\vec{r}^{'}|)d\vec{r}^{'}\Big)\psi(\vec{(r})d\vec{r}\end{aligned} \tag{2.15}$$

$$\frac{\delta\langle\Psi(\vec{r})|\Psi(\vec{r})\rangle}{\delta\psi^*} = N\int \delta\psi^*(\vec{r})\psi(\vec{r})d\vec{r} \tag{2.16}$$

Combining all the terms and arranging them leads to:[1]

$$\begin{split}\frac{\delta F[\Psi(\vec{r})]}{\delta\psi^*} = N\int\Big[-\frac{1}{2m}\nabla^2\psi(\vec{r}) + V_0(\vec{r})\psi(\vec{r})\\ + (N-1)\Big(\int|\psi(\vec{r})|^2V_{int}(|\vec{r}-\vec{r}^{\prime}|)d\vec{r}^{\prime}\Big)\psi(\vec{r}) - \mu\psi(\vec{r})\Big]\delta\psi^*(\vec{r})d\vec{r} = 0\end{split} \tag{2.17}$$

It can be clearly seen that the expression inside the square brackets must go to zero. The form of the interaction potential is generally taken to be $V_{int}(|\vec{r}-\vec{r}^{\prime}|) = \frac{4\pi}{m}a_s\delta(\vec{r}-\vec{r}^{\prime})$. Here, a_s is the s-wave scattering length. With the approximation valid for large number of particles , $N-1 \simeq N$, the following equation is obtained:[1]

$$-\frac{1}{2m}\nabla^2\psi(\vec{r}) + V_0(\vec{r})\psi(\vec{r}) + N\frac{4\pi}{m}a_s|\psi(\vec{r})|^2\psi(\vec{r}) = \mu\psi(\vec{r}) \tag{2.18}$$

This equation is known as the time Gross-Pitaevskii equation. The value of a_s determines the strength of the interactions present between the Bosons. A positive a_s signifies repulsive interactions and a negative a_s signifies attractive interactions. As equation (1.25) is non-linear, the energy per particle is calculated by multiplying the equation (1.25) with $\psi^*(\vec{r})$ and integrating it over $\vec{r}$, where the value of μ is equal to $\frac{\delta\langle H\rangle_N}{\delta N}$ [2]

The Time-Dependent Gross-Pitaevskii equation can be written as:

$$i\frac{\delta\psi(\vec{r},t)}{\delta t} = -\frac{1}{2m}\nabla^2\psi(\vec{r},t) + V_0(\vec{r},t)\psi(\vec{r},t) + N\frac{4\pi}{m}a_s|\psi(\vec{r},t)|^2\psi(\vec{r},t) - \mu\psi(\vec{r},t) \tag{2.19}$$

2.2 Beyond the Mean Field Approximation

The Gross-Pitaevskii ansatz is generally used to describe the dynamics of BECs. But this approximation is only concentrates on the ground state of the system because the basis is ground state single particle wavefunctions. To observe the effects of the excited state on the system a more general approach needs to be taken.

The next step in this direction was taken by Streltsov and co-workers in 2003[5]. They introduced the multi-orbital mean field theory, also called as the Best Mean field (BMF) theory. In this they considered a many body ansatz in which they distibuted N particles in M orbitals such that n_1 particle were in orbital ϕ_1, n_2 particle were in ϕ_2 and n_m particles were in ϕ_M as:[4]

$$\Psi(\vec{r}_1, \vec{r}_2, ..., \vec{r}_n) = \phi_1(\vec{r}_1)...\phi_1(\vec{r}_{n_1})\phi_2(\vec{r}_{n_1+1})...\phi_2(\vec{r}_{n_1+n_2})...\phi_M(\vec{r}_{n_1+n_2+..+n_M}) \quad (2.20)$$

In a later work, using this approximation they showed that on splitting the BECs in presence of a time dependent barrier, the evolved state is an excited state which is very close to the ground state[6].

In 2007, they introduced the Multconfigurational time-dependent Hartree method for bosons[3]. In this approach a time dependent ansatz analogous to the configuration interactions in Electronic Structure Theory is taken. In a bosonic system with N particles and M states, the time dependent ansatz is a combination of all the possible configurations which come out of distributing N particles over M states. As bosonic particles are indistinguishable and don't follow the Pauli's exclusion principle, the total number of configurations will be $\binom{M+N-1}{N}$.

In the second quantization formalism, a time dependent permanent defined by using a complete set of orthonormal time dependent orbitals $\{\phi_k(\mathbf{r}, t)\}$ can be written as:

$$|\vec{n}; t\rangle = \frac{1}{\sqrt{n_1!n_2!n_3!...n_M!}}[a_1^\dagger(t)]^{n_1}[a_2^\dagger(t)]^{n_2}...[a_M^\dagger(t)]^{n_M}|0\rangle \quad (2.21)$$

where $|0\rangle$ is the vacuum state and $a_1^\dagger$ is the bosonic creation operator which has been discussed in the first chapter. Here $n = n_1 + n_2 + ... + n_M$, where $(n_1, n_2....., n_M)$ signify the total number of particles in each state.

The assumption of the total wavefunction in terms of the time dependent permanent can be written as:[3]

$$\Psi(t) = \sum_{\vec{n}} C_{\vec{n}}(t)|\vec{n}; t\rangle \quad (2.22)$$

where $C_{\vec{n}}t$ is known as the expansion coefficients. In the limit of the number of states $M \rightarrow \infty$, this ansatz describes the system exactly and the permanents span the entire bosonic hilbert space.

The two sets of independent parameters, the expansion coefficients $C_{\vec{n}}t$ and the single particle orbitals $\phi_k(\vec{r},t)$ are studied over time to determine the properties of the system. The equations concerning their motion are derived using the time-dependent variational principle. The functional action can be written in terms of the lagrangian as:

$$S[x_1, x_2] = \int_{t1}^{t2} \mathcal{L}\{x_1, x_2\} dt \tag{2.23}$$

so, for time-dependent Schrodinger equation:

$$\begin{aligned} S[\{C_{\vec{n}}(t)\}, \{\phi_k(\vec{r},t)\}] &= \int \Big\{ \langle \Psi(t)|\hat{H} - i\frac{\partial}{\partial t}|\Psi(t)\rangle & (2.24) \\ &\quad - \sum_{k=1,j=1}^{M} \alpha_{k,j}(t)\{\langle \phi_k|\phi_j\rangle - \lambda_{k,j}\Big\} dt & (2.25) \end{aligned}$$

Now, the functional action is minimized with respect to the two independent parameters, the expansion coefficients and the single particle orbitals. Solving the equations gives two equations of motion.

Firstly, the expectation term in the above equation can be written as:

$$\langle \Psi(t)|\hat{H} - i\frac{\partial}{\partial t}|\Psi(t)\rangle = \sum_{\vec{n}} \sum_{\vec{n}'} C^*_{\vec{n}}[C_{\vec{n}'}\langle \vec{n};t|\hat{H} - i\frac{\partial}{\partial t}|\vec{n};t\rangle - \langle \vec{n};t|\vec{n};t\rangle i\frac{\partial C_{\vec{n}'}}{\partial t}] \tag{2.26}$$

Using the Orthogonality relation, the functional variation with respect to the expansion coefficients $\{C_{\vec{n}}(t)\}$

$$\frac{\partial S}{\partial C^*_{\vec{n}}(t)} = 0 \tag{2.27}$$

gives[3]

$$\sum_{\vec{n}'} \langle \vec{n};t|\hat{H} - i\frac{\partial}{\partial t}|\vec{n}';t\rangle C_{\vec{n}'} = i\frac{\partial C_{\vec{n}}}{\partial t} \tag{2.28}$$

This is the first equation of motion in a matrix form[3]

$$\mathbf{H}(t)\mathbf{C}(t) = i\frac{\partial \mathbf{C}(t)}{\partial t} \tag{2.29}$$

To write the hamiltonian in a form dependent on the orbitals, the second quantization formalism is used where the operators can be expressed using the creation and annhilation operators. The one particle operators can be written as:

$$\hat{h} - i\frac{\partial}{\partial t} = \sum_k \sum_q a_k^{'} a_q \left[h_{kq} - \left(i\frac{\partial}{\partial t} \right)_{kq} \right] \tag{2.30}$$

where

$$\hat{h} = \sum_{i=1}^{n} \left(\frac{p_i^2}{2m} + V_0(\vec{r}_i) \right) \tag{2.31}$$

and the two particle operator is defined as:

$$\frac{1}{2} \sum_i^n \sum_{j \neq i}^n V(|\vec{r}_i - \vec{r}_j|) = \frac{1}{2} \sum_{k,s,q,l} a_k^\dagger a_s^\dagger a_q a_l V_{ksql} \tag{2.32}$$

The next step involves writing the expectation value in a new form using definitions of one particle and two particle reduced density matrix elements, $\rho_{kq} = \langle \Psi(t) | a_k^\dagger a_q | \Psi(t) \rangle$ and $\rho_{ksql} = \langle \Psi(t) | a_k^\dagger a_s^\dagger a_q a_l | \Psi(t) \rangle$ [3]

$$\begin{aligned} \langle \Psi(t) | \hat{H} - i\frac{\partial}{\partial t} | \Psi(t) \rangle &= \sum_{k=1}^{M} \sum_{q=1}^{M} \rho_{kq} \left[h_{kq} - \left(i\frac{\partial}{\partial t} \right)_{kq} \right] && (2.33) \\ & + \frac{1}{2} \sum_{k=1}^{M} \sum_{s=1}^{M} \sum_{q=1}^{M} \sum_{l=1}^{M} \rho_{ksql} V_{ksql} - i \sum_{\vec{n}} C_{\vec{n}}^* \frac{\partial C_{\vec{n}}}{\partial t} && (2.34) \end{aligned}$$

Now, minimizing the functional action with respect to the orbitals:

$$\frac{\partial S}{\partial \phi_k^*(\vec{r}, t)} = 0 \tag{2.35}$$

would yield the following equation [3]

$$\sum_{q=1}^{M} \left[\rho_{kq} \left(\hat{h} - i\frac{\partial}{\partial t} \right) + \sum_{s=1}^{M} \sum_{l=1}^{M} \rho_{ksql} \hat{V}_{sl} \right] |\phi_q\rangle = \sum_{j=1}^{M} \alpha_{kj} |\phi_j\rangle \tag{2.36}$$

The value of the lagrange multiplier can be obtained by multiplying this equation from the left with $\langle\phi_j|$. the value is again substituted into the equation and then the equation is transformed using the invariance property of the ansatz ($\langle\phi_k|\frac{\partial\phi_q}{\partial t}\rangle = 0$) and the final equation describing the motion of the orbitals is achieved[3]

$$i|\frac{\partial\phi_j}{\partial t}\rangle = \hat{\mathbf{p}}\left[\hat{h}|\phi_j\rangle + \sum_{k=1}^{M}\sum_{s=1}^{M}\sum_{q=1}^{M}\sum_{l=1}^{M}(\rho(t))^{-1}_{jk}\rho_{ksql}\hat{V}|\phi_q\rangle\right] \tag{2.37}$$

where $\hat{\mathbf{p}}$ is a projection operator which ensures that the changes of the orbitals in time are always in the orthogonal space to the orbitals.[3]

$$\hat{\mathbf{p}} = 1 - \sum_{i=1}^{M}|\phi_i\rangle\langle\phi_i| \tag{2.38}$$

The unitary transformation ($\langle\phi_k|\dot{\phi}_q\rangle = 0$) also simplifies the matrix elements of eq. (2.28) to $\langle\hat{n};t|\hat{H}|\vec{n};t\rangle$

Thus, two coupled equations of motions have been obtained. These equations are exact equations are exact representation of the system in the limit of the number of states going to infinity. Thus, a better approximation is achieved as compared to the Gross-Pitaevskii equation.

2.3 Atoms in an intense laser field

When an atom is subjected to a light beam, the ionization probability of the electron increases with the increase in the intensity of the beam. This phenomenon occurs at low intensities where the field can be taken as a small perturbation. But in the presence of highly intense lasers the atoms tend to stabilize rather than ionize which goes against common intuition. At such high intensities the atom becomes "supersaturated"[11] with photons and the electrons see a continuous electric field instead of quanta of photons. So when studying the high intensity regime the atom is taken as a quantum mechanical entity and the field is treated classically. This approach is known as the semiclassical approach. The electrons oscillate in the presence of the electric field and their attachment with nucleus (binding energy) weakens. But instead of ionizing they tend to stabilize and reside in a ”dressed state” which has a very high kinetic energy. The probability density in this state is distorted because of the electrons are far away from the nucleus and are having a large kinetic energy. The process of stabilization is largely due to the presence of the oscillating field. The binding energy plays a little role.

W. C. Henneberger, in 1968, suggested a new frame of reference, based on a work of Kramers. This reference frame came to be know as the Kramers-Henneberger reference frame[7] and was later explored by many groups. In this regime, the frame of reference is moving with the oscillating electron. The stationary electron is hit by an oscillating nucleus but most of the time the nucleus is far away from the electron because of the high intensity of the field and the collision time is very small if high frequency is taken. As a result, ionization probability decreases and atomic stabilization is achieved. The electron eventually gains enough momentum to undergo ionization but prolonged stabilization can be seen. The electron in this reference frame sees a time averaged potential which is known as the KH potential and the electron remains bound in this potential. This frame of reference can be described mathematically by a set of transformations which are discussed in this subsequent sections.

2.3.1 Atom-Field equation

The Schröinger equation describing the interaction of an electron with an electromagnetic field is written in the radiation gauge as($\hbar = 1$):

$$\left(-\frac{1}{2}[\vec{\nabla} - i\vec{A}(\vec{r},t)]^2 + V_{scalar}(\vec{r},t) + V_{binding}(\vec{r}) \right)\Psi = i\frac{\delta\Psi}{\delta t} \tag{2.39}$$

where $\vec{V}(\vec{r},t)$ and $V_{scalar}(\vec{r},t)$ are the vector and scalar potentials associated with the field, $V_{binding}$ is the binding potential of the atom. The vector and scalar potentials are gauge dependent quantities. They change across different gauge transformations to keep the total system invariant under these transformations. The quantities which are independent of these transformations are the electric field, $\vec{E} = -\nabla V_{scalar} - \frac{\delta \vec{A}}{\delta t}$, and magnetic field $\vec{B} = \nabla \times \vec{A}$.

The Schrödinger equation can be simplified by using the dipole approximation. As the size of atom is small as compared to the wavelength of the electromagnetic wave, the expansion of the vector potential can be truncated by ignoring the higher terms. Mathematically the dipole approximation is written as $\vec{k}.\vec{r} << 1$ where $\vec{k} = 2\pi/\lambda$. The vector potential is[13]

$$\begin{aligned} \vec{A}(\vec{r}+\vec{r}_i,t) &= \vec{A}(t)exp\{i\vec{k}.(\vec{r}+\vec{r}_i)\} && (2.40) \\ &\simeq \vec{A}(t)exp(i\vec{k}.\vec{r}_i) && (2.41) \end{aligned}$$

Now, the Schrödinger equation after applying the dipole approximation and the conditions of the radiation gauge can be written as:

$$\left(-\frac{1}{2}[\nabla - i\vec{A}(\vec{r}_i,t)]^2 + V_{binding}(\vec{r})\right)\Psi = i\frac{\delta\Psi}{\delta t} \tag{2.42}$$

The transformation

$$\Psi = exp\{i\vec{A}(\vec{r}_i,t).\vec{r})\chi(\vec{r},t) \tag{2.43}$$

converts the Schrödinger equation into a very familiar form

$$\left(\frac{\vec{p}^2}{2} + V_{binding}(\vec{r}) - \vec{r}.\vec{E}(\vec{r}_i,t)\right)\chi(\vec{r},t) = i\frac{\delta\chi(\vec{r},t)}{\delta t} \tag{2.44}$$

where $\vec{E} = -\dot{\vec{A}}$.

In this equation, the $\vec{r}.\vec{E}(\vec{r}_i,t)$ term represents the dipole interaction term. A set of transformations can now be applied to this equation to take it to the Kramers-Hanneberger frame[9].

2.3.2 Kramers-Hanneberger transformation

The time-dependent dipole term $\vec{r}.\vec{E}(\vec{r_i},t)$ needs to be removed from the equation (2.46). The transformation required to remove it can be derived by writing

$$\chi(\vec{r},t) = U_1\phi(\vec{r},t) \tag{2.45}$$

in the equation (2.46). The equation now becomes($\hbar = 1$):

$$i\vec{\phi}(\vec{r},t)\frac{\delta U_1}{\delta t} + iU_1\frac{\delta\phi(\vec{r},t)}{\delta t} = \left(\frac{\vec{p}^2}{2} + V_{binding}(\vec{r})\right)U_1\phi(\vec{r},t) - \vec{r}.\vec{E}(\vec{r_i},t)U_1\phi(\vec{r},t) \tag{2.46}$$

From this equation it can be seen that

$$i\phi(\vec{r},t)\frac{\delta U_1}{\delta t} = -\vec{r}.\vec{E}(\vec{r_i},t)U_1\phi(\vec{r},t) \tag{2.47}$$

Solving this equation gives the first transformation:

$$U_1 = exp\left(i\vec{r}\int_0^t \vec{E}(\vec{r_i},t)\delta t\right) \tag{2.48}$$

Similarly the second transformation to the wavefunction can be obtained

$$U_2 = exp\left(i\nabla\int_0^{t'} \delta t' \int_0^t \vec{E}(\vec{r},t)\delta t\right) \tag{2.49}$$

Equation (2.50) and (2.51) transforms the hamiltonian into a form in which the potential becomes time dependent. This time dependent potential is known as the KH potential and this frame of reference is called the KH frame of reference.

$$\left(\frac{\vec{p}^2}{2} + V(\vec{r} - \int \vec{A}(\vec{r_i},t)\delta t)\right)\Psi^{KH} = i\frac{\delta\Psi^{KH}}{\delta t} \tag{2.50}$$

The following expression is generally used:

$$\int \vec{A}(\vec{r_i},t)\delta t = \alpha(t) \tag{2.51}$$

where $\alpha(t) = \alpha_0 cos(\omega t)$. The term α decides the amplitude of the oscillations and ω is the frequency of the field. Here

$$\alpha = \frac{\epsilon}{\omega^2} \tag{2.52}$$

where ϵ is the field intensity.

This indicates that the field parameters, intensity and frequency, must be chosen carefully in order to achieve stabilization. The value of α is key to observe the phenomenon of stabilization.

The time dependent potential can be expanded into a time-independent zeroth order term and time dependent higher order terms.

$$V(\vec{r} - \alpha cos(\omega t)) = V_0^{KH} + \sum_{n=1}^{\infty} V_n^{KH} cos(n\omega t) \tag{2.53}$$

where

$$V_0^{KH} = \frac{1}{2\pi}\int_0^{2\pi} V(\vec{r} - \alpha cos(\tau))d(\tau) \tag{2.54}$$

$$V_n^{KH} = \frac{1}{2\pi}\int_0^{2\pi} V(\vec{r} - \alpha cos(\tau))cos(n\tau)d(\tau) \tag{2.55}$$

The zeroth order time independent term is the effective potential experienced by the electron. The higher order time-dependent terms are fast oscillating which can be taken as a perturbation at low frequencies and equal to zero at high frequencies. The zeroth order potential is a stabilizing potential and the terms reponsible for ionization are eliminated.

2.3.3 Zeroth-order KH calculations for Merging Dynamics

The Zeroth order potential term for a symmetrical double well potential of the form $Ax^4 - Bx^2$ is:

$$\begin{aligned} V_0^{KH} &= \frac{1}{2\pi}\int_0^{2\pi} A(x - \alpha cos(\tau))^4 - B(x - \alpha cos(\tau))^2 d\tau && (2.56)\\ &= \frac{1}{2\pi}\left[A(\frac{3}{4}\alpha^2\pi x^2 + 6\alpha^2\pi x^2 + 2\pi x^4) - B\pi(\alpha^2 + 2x^2)\right] && (2.57) \end{aligned}$$

The merging caluclations are done at $A = 1a.u.$ and $B = 5a.u.$. The potential V_0^{KH} at different values of *alpha* are plotted. The ideal α for which stabilization occurs on top of the barrier is $\alpha = 2.581190$.

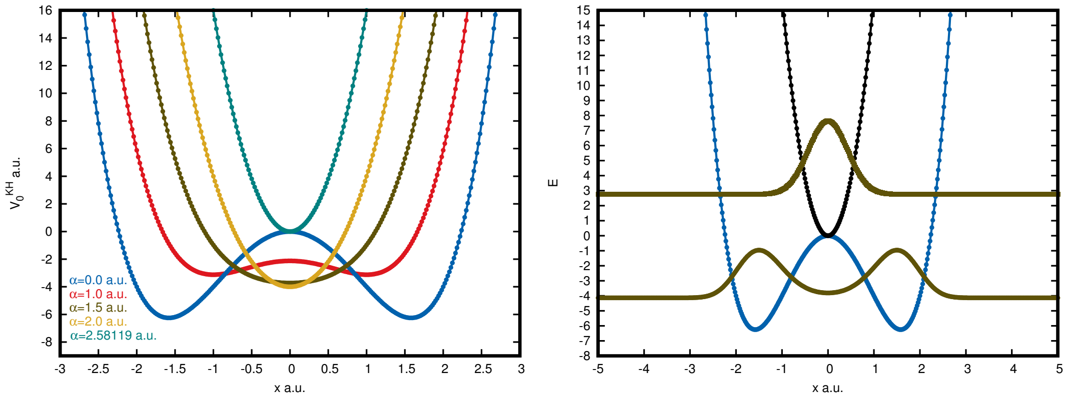

FIGURE 2.1: (a) V_0^{KH} at different α values. (b)Stabilization of the wavefunction on top of the time averaged KH potential.

The variation of the value of α with the variation in the values of A and B is also observed. At B=5, α decreases with the increasing A value and at A=1, α increases with the increasing value of B.

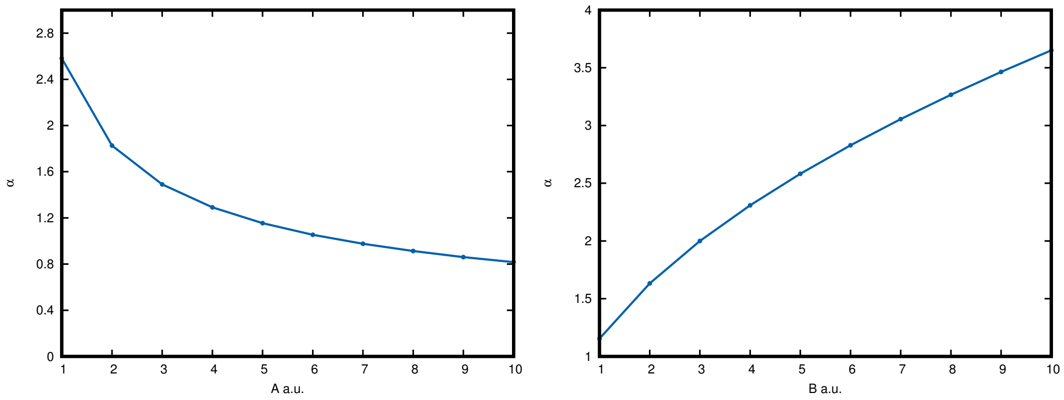

FIGURE 2.2: (a)Variation of α with A at B=5 (b) Variation of α with B at A=5.

2.4 Merging of the BECs

The BECs formed through the cooling method discussed in the first chapter have a finite lifetime. The loss of the BECs can be attributed to the presence of the 3 body inelastic collisions within the gas and 2 body elastic collision with the gas present in the background of the trap.[15] A really simple approach to having a long lived BEC is introducing a newly formed BEC confined in a trap and merging it with the already existing one. A continuous BEC source is also helpful in the generation of atomic beams[14]. Chikkatur and co-workers, in 2002, showed that two independently trapped Bose-Einstein condensates could be merged into one by bringing them closer together[16].

A lot of work has been done towards the study of the fragmentation of the BECs both theoretically and experimentally. In 1998, R. Dum and co-workers[12], studied this phenomenon using the KH regime. They started with a wave-packet initially trapped in a harmonic potential and then this potential was periodically shaken. They used the Gross-Pitaevskii equation to describe the system.

$$i\frac{\delta\psi}{\delta t} = [-\nabla^2 + V(\vec{r} + \alpha(t)\hat{z}) + \frac{4\pi a}{m_{a.u.}}N|\psi|^2]\psi \tag{2.58}$$

where[12]

$$V(\vec{r}) = \begin{cases} \frac{m}{2}(\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2), & V(\vec{r}) \leq V_{cut-off}, \\ V_{cut-off} & V(\vec{r}) > V_{cut-off} \end{cases} \tag{2.59}$$

and[12]

$$\alpha(t) = \begin{cases} \alpha_0 sin^2(\frac{\pi}{2}\frac{t}{t_{on}} sin(\omega_{laser}t), & 0 \leq t \leq t_{on} \\ \alpha_0 sin(\omega_{laser}t), ton \geq t_{on} \end{cases} \tag{2.60}$$

where t_{on} is the time for which the laser pulse is switched on and after that a continuous laser is being shined on the system.

The condensates got distorted and formed a fragmented state in the presence of the pulse, feeling the effects of the time averaged oscillations.

The opposite phenomenon is studied where two fragmented condensates initially placed in a symmetric double well potential are taken. The trapping potential of the condensates is then shaken vigorously by applying a pulse of the form mentioned in eq. (2.60). This dynamics is studied using both the Gross-Pitaevskii equation and the **MCTDHB** [3] method. It is observed that due to the time averaged effects of the fast oscillating potential, the initially fragmented BECs form a single merged BEC of greater density confined in a time averaged potential.

The calulations are performed using the **MCTDHB** [3] package as follows:

- A system of $N = 100$ weakly interacting bosons is taken, with interaction strength $\lambda_0 = -0.002 a.u.$.
- A symmetric double well potential of the form $Ax^4 - Bx^2$ is initially applied, where $A = 1$ and $B = 5$.
- The grid is taken from $x = -10 a.u.$ to $x = 10 a.u.$ with number of points $nx = 201$.
- Relaxation of the system is performed for time interval $t = 10 a.u.$ to obtain an initial stable wavepacket.
- The time dependent potential is of the form:

$$V(x,t) = \begin{cases} (x - \alpha_0 sin^2(\frac{\pi}{2}\frac{t}{100}) sin(25t))^4 - 5(x - \alpha_0 sin^2(\frac{\pi}{2}\frac{t}{100}) sin(25t))^2, t \leq t_{on} = 100 \\ (x - \alpha_0 sin(25t))^4 - 5(x - \alpha_0 sin(25t))^2, t \geq t_{on} = 100 \end{cases} \tag{2.61}$$

- Time propagation calculations are done for two values of α_0, first value is $\alpha_0 = 2.3245 a.u.$ and the other value is calculated through Zeroth order KH calculations which is $\alpha_0 = 2.581190 a.u.$. Here, $\omega = 25 a.u.$ and $t_{on} = 100 a.u.$. Merging was observed in both the cases.
- In the Gross-Pitaevskii framework (M=1) all the particles are in the ground state. For number of states (M=2) the particles are equally divided into the two states initially. Some crude time-propagation calculations were initially done by Abhijeet Roy[17] at N=1000 bosons and $\alpha_0 = 4.3245$.
- Results are shown for $\alpha_0 = 2.581190$ at different time intervals in the figure

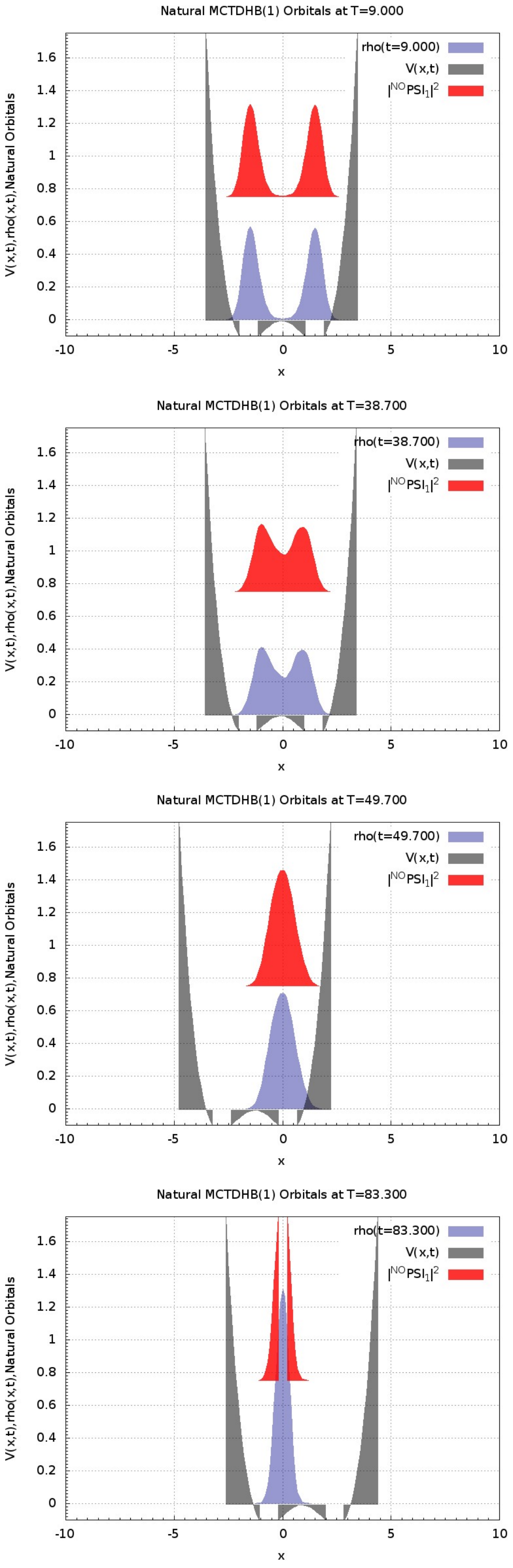

FIGURE 2.3: Merging of condensates for M=1 or GPE at $\alpha_0 = 2.581190 a.u.$

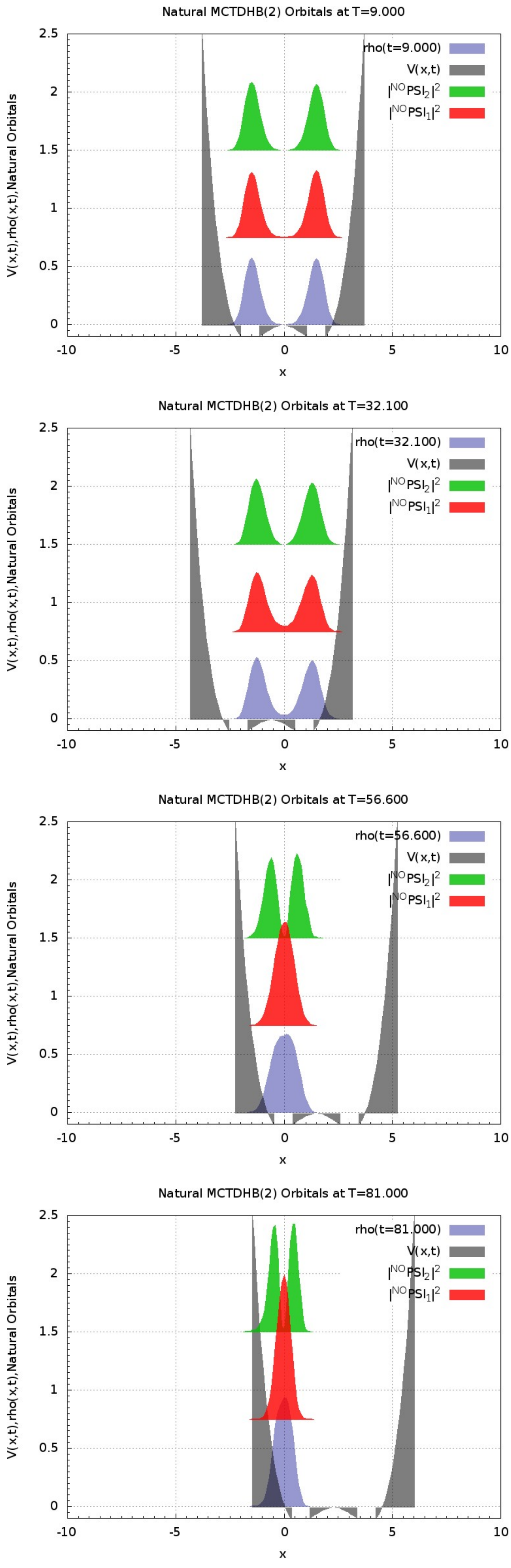

FIGURE 2.4: Merging of condensates for M=2 states at $\alpha_0 = 2.581190 a.u.$

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Chapter 3

Implementation of the (t,t') algorithm for time-dependent Hamiltonians

3.1 Quantum Adiabatic Theorem

An adiabatic process can be defined as a gradual change in the external surroundings of a system so as to allow the system to perform a smooth change of a degree of freedom as opposed to an abrupt change which can lead to erratic and chaotic behaviour. The energy is not conserved during an adiabatic change.

For a quantum system which remains non-degenerate for a time interval, the Time Dependent Schr$\ddot{o}$dinger equation (TDSE) can be written as:

$$i\hbar\frac{\partial\psi(\vec{r},t)}{\partial t} = \hat{H}(\vec{r},t)\psi(\vec{r},t) \tag{3.1}$$

The wavefunction can be transformed using a time dependent unitary operator U as[2]:

$$\psi(\vec{r},t) = U(t)\phi(\vec{r},t) \tag{3.2}$$

If eq. (3.2) is substituted into eq. (3.1), one obtains:

$$i\hbar U(t)\frac{\partial\phi(\vec{r},t)}{\partial t}+i\hbar\phi(\vec{r},t)\frac{\partial U(t)}{\partial t}=\hat{H(t)}(\vec{r},t)U(t)\phi(\vec{r},t) \tag{3.3}$$

Left multiplying the above equation with $U^{-1}(t)$ gives:

$$i\hbar\frac{\partial\phi(\vec{r},t)}{\partial t}+i\hbar\phi(\vec{r},t)U^{-1}(t)\frac{\partial U(t)}{\partial t}=\hat{H}_T(\vec{r},t)\phi(\vec{r},t) \tag{3.4}$$

If a gradually varying Hamiltonian $H(t)$ is taken in the beginning, then U(t) will also vary slowly with time and hence the second term on the L.H.S. of eq. (3.4) will go to zero. Here, $\hat{H}_T(\vec{r},t)$ is a transformed operator given by $\hat{H}_T(\vec{r},t)=U^{-1}\hat{H}(\vec{r},t)U$ The equation can now be written as:

$$i\hbar\frac{\partial\phi(\vec{r},t)}{\partial t}=\hat{H}_T(\vec{r},t)\phi(\vec{r},t) \tag{3.5}$$

Thus it can be seen that if the initial wavefunction is an eigenfunction of the Hamiltonian, it will remain its eigenfunction during the evolution process albeit the Hamiltonian is slowly varying. This is known as the quantum adiabatic theorem[1].

3.2 Split Operator Method

The split operator method is an approximate method which is used for numerically solving the TDSE. If the Time dependent Schrödinger equation is of the form ($\hbar=1$):

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\hat{H}(\vec{r})\Psi(\vec{r},t) \tag{3.6}$$

where $\hat{H}(\vec{r})$ is a time independent hamiltonian, the unitary transform for time-evolution is given by:

$$U(t)=e^{-i\hat{H}(\vec{r})t} \tag{3.7}$$

The Hamiltonian can be written as the sum of a kinetic Energy operator and the potential energy operator. The time evolution of the wavefunction is then described by the following equation:

$$\Psi(\vec{r},t) = e^{-i(\hat{T}(\vec{r})+\hat{V}(\vec{r}))t}\Psi(\vec{r},0) \tag{3.8}$$

Since, the kinetic energy and potential energy operators do not commute,

$$e^{-i(\hat{T}(\vec{r})+\hat{V}(\vec{r})t} \neq e^{-i\hat{T}(\vec{r})t}e^{-i\hat{V}(\vec{r})t}. \tag{3.9}$$

The Baker-Campbell-Hausdorff formula states that[13]:

$$e^{\hat{x}}e^{\hat{y}} = exp(\hat{x}+\hat{y}+\frac{1}{2}[\hat{x},\hat{y}]+\frac{1}{12}[\hat{x},[\hat{x},\hat{y}]]-\frac{1}{12}[\hat{y},[\hat{y},\hat{x}]]+....) \tag{3.10}$$

In 1982, Feit and Fleck [4]suggested an approximation method for solving this equation which is known as the split operator method. Using another variant of the BCH formula,

$$e^{\hat{x}}e^{\hat{y}}e^{\hat{z}} = exp(\hat{x}+\hat{y}+\hat{z}+\frac{1}{2}[\hat{x},\hat{y}]+\frac{1}{2}[\hat{y},\hat{z}]+\frac{1}{2}[\hat{x},\hat{z}]+\frac{1}{12}[[\hat{x},\hat{y}]+\hat{x}+\hat{y}+\hat{z}]+...) \tag{3.11}$$

,the evolution operator can be written as:

$$e^{-i\frac{\hat{V}(\vec{r})}{2}t}e^{-i\hat{T}(\vec{r})t}e^{-i\frac{\hat{V}(\vec{r})}{2}t} = e^{-i(\frac{\hat{V}(\vec{r})}{2}+\hat{T}(\vec{r})+\frac{\hat{V}(\vec{r})}{2})t}+O(t^3) \tag{3.12}$$

Here the two kinetic energy operators commute with each other and the error is of the order of $O(t^3)$, which is better as compared to equation (3.9) where the error is of the order of $O(t^2)$.

But this method is more useful in the case of time independent Hamiltonians because the evolution operator of the time dependent Hamiltonian can't be written in this way. Although, this method is used for time dependent Hamiltonians, there are other approximations which are better and discussed in further sections.

3.3 The time-ordering operator

The Time evolution of the wavefunction of a TDSE with a time dependent Hamiltonian cannot be described easily like the TDSE with a time-independent Hamiltonian operator (eq. 3.8).

In comparison with eq. (3.8), the most suitable form seems to be ($\hbar = 1$):

$$\Psi(\vec{r}, t) = e^{-i(\int_0^t \hat{H}(\vec{r},t')dt')}\Psi(\vec{r}, 0) \tag{3.13}$$

but this equation is not exact. The time evolution operator in the interval $(0 < t < t_f)$ must follow a chronological order

$$U(t_f, 0) = U(t_f, t)U(t, 0) \tag{3.14}$$

but as the operator $e^{-i\int_0^t \hat{H}(\vec{r},t')dt'}$ does not commute at two different instances of time

$$e^{-ii\int_t^{t_f} \hat{H}(\vec{r},t')dt' - i\int_0^t \hat{H}(\vec{r},t')dt'} \neq e^{-i\int_t^{t_f} \hat{H}(\vec{r},t')dt'}e^{-i\int_0^t \hat{H}(\vec{r},t')dt'} \tag{3.15}$$

One way to overcome this is to write the wavefunction in a Taylor expansion[2]:

$$\begin{aligned}\Psi(\vec{r}, t) &= \Psi(\vec{r}, 0) - i\int_0^t \hat{H}(\vec{r}, t')dt'\Psi(\vec{r}, 0) \qquad (3.16)\\ &-\frac{1}{2}\int_0^t\int_0^t \hat{H}(\vec{r}, t')\hat{H}(\vec{r}, t'')dt'dt''\Psi(\vec{r}, 0) + ... \qquad (3.17)\end{aligned}$$

But here also the chronological order of operations is violated. Thus, a new operator known as the time ordering operator is introduced which imposes the condition that the operator must act on the wavefunction in a certain order.

$$\Psi(\vec{r}, t) = \hat{T}e^{-i(\int_0^t \hat{H}(\vec{r},t')dt')}\Psi(\vec{r}, 0) \tag{3.18}$$

where $\hat{T}$ is the time-ordering operator.

3.4 Floquet Method for time periodic Hamiltonians

The Floquet method is used for solving the TDSE when the Hamiltonian matrix is time periodic, i.e.

$$\hat{H}(t) = \hat{H}(t+T) \tag{3.19}$$

where T is the time period of the Hamiltonian. Periodic Hamiltonians can be encountered while studying the atom in the presence of a continuous oscillating field. This method was introduced by Shirley in 1965[5]. In this method, the time dependent hamiltonian is converted into an infinite dimensional time independent Hamiltonian using a complete basis set in both the space co-ordinate and time which was suggested by H. Sambe[8] in 1973. The solution to the time dependent Schr$\ddot{o}$dinger equation with time periodic Hamiltonian is given by the Floquet ansatz ($\hbar = 1$):

$$\Psi_k(\vec{r}, t) = e^{-iE_k t}\phi_k(\vec{r}, t) \tag{3.20}$$

Here, $\phi(\vec{r}, t)$ is also time-periodic and E_k is a diagonal matrix whose elements are known as characteristic exponents.

Substituting eq. (3.20) into the TDSE yields ($\hbar = 1$):

$$\left\{\hat{H}(\vec{r}, t) - i\frac{\partial}{\partial t}\right\}\phi_k(\vec{r}, t) = E_k\phi_k(\vec{r}, t) \tag{3.21}$$

$\left\{\hat{H}(\vec{r}, t) - i\frac{\partial}{\partial t}\right\}$ is known as the Floquet Hamiltonian denoted by $\hat{H}_F(\vec{r}, t)$ and $\phi_k(\vec{r}, t)$ is known as the Floquet eigenstate. To calculate the time independent eigenvalues E_k, equation (3.25) is left multiplied by $\phi_k(\vec{r}, t)$ and integrated over position space.

$$E_k = \int_{-\infty}^{\infty} \phi_k(\vec{r}, t)\left\{\hat{H}(\vec{r}, t) - i\frac{\partial}{\partial t}\right\}\phi_k(\vec{r}, t)d^3r \tag{3.22}$$

The wavefunction can be rewritten as ($\hbar = 1$):

$$\Psi_k = e^{-i(E_k + n\omega)t}e^{in\omega t}\phi_k(\vec{r}, t) \tag{3.23}$$

A Hilbert space (R $\oplus$ T) is taken which is extended in both space and time. The Eigenfunctions can be written as a product of a Fourier vector $|n\rangle$ and an atomic eigenstate[2]

$$|\alpha n\rangle\rangle = |\alpha\rangle|n\rangle \tag{3.24}$$

The Fourier vector gives the equation[2]

$$\langle t|n\rangle = e^{in\omega t} \tag{3.25}$$

The matrix elements can be calculated as:[2]

$$\langle\langle\alpha n|H_F(\vec{r},t)|\beta m\rangle\rangle = \hat{H}(\vec{r},t)^{n-m}_{\alpha\beta} + n\omega\delta_{\alpha\beta}\delta_{nm} \tag{3.26}$$

It can be seen that the matrix formed will be a diagonal matrix due to the orthogonality of the Hamiltonian and the presence of δ terms with the $n\omega$ term.

In the presence of an oscillating potential term $e^{i\omega t}$ the Floquet hamiltonian with three Floquet channels can be written as a block tridiagonal matrix:

$$\hat{H}_f = \begin{bmatrix} E+2\omega & D & 0 & 0 & 0 \\ D & E+\omega & D & 0 & 0 \\ 0 & D & E & D & 0 \\ 0 & 0 & D & E-\omega & D \\ 0 & 0 & 0 & D & E-2\omega \end{bmatrix} \tag{3.27}$$

The elements of the Floquet matrix can be written as:[2]

$$E_{\alpha\beta} = \langle\langle\alpha 0\beta 1|\hat{H}(\vec{r},t)|\beta 0\alpha 1\rangle\rangle = \frac{1}{T}\int_0^T \langle\alpha|\hat{H}(\vec{r},t)|\beta\rangle dt = \langle\alpha|\bar{H}(\vec{r})|\beta\rangle \tag{3.28}$$

where $\bar{H}(\vec{r})$ is a time averaged hamiltonian.

$$D_{\alpha\beta} = \frac{1}{T}\int_0^T \langle\alpha|\hat{H}(\vec{r},t)|\beta\rangle e^{i\omega t} dt \tag{3.29}$$

The presence of the $e^{i\omega t}$ term discards the blocks with $|n-m| > 1$ [2].

3.5 (t,t') method for Time Dependent Schr$\ddot{o}$dinger equation

In 1993, Uri Peskin and Nimrod Moiseyev[6], gave a new formalism to solve the TDSE which generalized the Floquet theorem used for time periodic Hamiltonians to time dependent Hamiltonians. This new method is known as the (t,t') method. In 1974, J. Howland[9] built up on the idea of H. Sambe (1973) and gave a Hilbert space spanning both the space and time coordinate for any Hamiltonian which is time dependent. This idea was picked up by Peskin and Nimrod and they introduced a new co-ordinate t' in the extended Hilbert space. This space is now constituted of a complete set of basis functions in both the space co-ordinate r and the time co-ordinate t'. This expansion of the Hilbert space allows the time-dependent Hamiltonian to be written as an infinite dimensional time independent Hamiltonian. Thus, evolution operators in this time independent form do not require a time ordering operator because their chonological ordering is now not required as the Hamiltonian is time-independent.

Consider a Time Dependent Schr$\ddot{o}$dinger equation ($\hbar = 1$):

$$i\frac{\partial \Psi(\vec{r},t)}{\partial t} = \hat{H}(\vec{r},t)\Psi(\vec{r},t) \tag{3.30}$$

The solution to the time dependent Schr$\ddot{o}$dinger equation in the new scheme can be written as:[6]

$$\Psi(\vec{r},t) = \Psi(\vec{r},t',t)\big|_{t=t'} \tag{3.31}$$

The equation of time evolution of the system:

$$\Psi(\vec{r},t',t) = e^{-i\hat{H}_F(\vec{r},t')(t-t_0)}\Psi(\vec{r},t',t) \tag{3.32}$$

where $\hat{H}_F(\vec{r},t)$ is the Floquet hamiltonian in the coordinate t'.

$$\hat{H}_F(\vec{r},t') = \hat{H}(\vec{r},t') - i\frac{\partial}{\partial t'} \tag{3.33}$$

The starting point of the derivation of this equation is to differentiate the above equation with respect to t and multiply i to both sides. The equation becomes:

$$i\frac{\partial\Psi(\vec{r},t',t)}{\partial t} = \hat{H}_F(\vec{r},t')e^{-i\hat{H}_F(\vec{r},t')(t-t_0)}\Psi(\vec{r},t',t) \tag{3.34}$$

The right hand side of the equation can be written as:

$$\hat{H}(\vec{r},t')e^{-i\hat{H}_F(\vec{r},t')(t-t_0)}\Psi(\vec{r},t',t) = -i\frac{\partial\Psi(\vec{x},t',t)}{\partial t'} + \hat{H}(\vec{r},t)\Psi(\vec{r},t',t) \tag{3.35}$$

Combining equation (3.39) and (3.40), one gets[6]

$$i\left(\frac{\partial}{\partial t'} + \frac{\partial}{\partial t}\right)\Psi(\vec{r},t',t) = \hat{H}(\vec{r},t')\Psi(\vec{r},t',t) \tag{3.36}$$

Over the contour where $t = t'$, equation (3.41) converts into the time-dependent Schrödinger equation which was taken in the beginning,

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t} = \hat{H}(\vec{r},t)\Psi(\vec{r},t) \tag{3.37}$$

The initial wavefunction can be expanded in a basis of fourier functions on the t' co-ordinate

$$\Psi_i(\vec{r},t',0) = \sum_{n=-\infty}^{\infty} e^{in\omega t'}\psi(\vec{r}) \tag{3.38}$$

If the basis functions in t' are $\phi(t')$ and the basis functions in r are $\phi(\vec{r})$ then the term

$$\langle\langle\phi(t'),\phi(\vec{r})|\Psi_i(t)\rangle\rangle = \frac{1}{T}\int_0^T dt'\int_{-\infty}^{\infty} d\vec{r}\phi(t')\phi(\vec{r})\Psi_i(t) = \Psi_i(\vec{r},t',t) \tag{3.39}$$

and

$$\Psi_i(\vec{r},t',t) = [e^{-i\hat{H}_F(\vec{r},t')t}\Psi_i(\vec{r},t',0)] \tag{3.40}$$

The Floquet matrix can be written as:

$$\hat{H}_F(\vec{r},t') = -\frac{1}{2}\frac{\partial^2}{\partial r^2} + V(\vec{r},t') - i\frac{\partial}{\partial t'} \tag{3.41}$$

To remove the t' co-ordinate the Floquet matrix is averaged out over this co-ordinate in the Fourier basis expansion. The Floquet matrix elements are the same as those obtained in the previous section.

The Time Evolution of the system can now be written in a form in which only the propagation of the wavefunction corresponding to the n=0 Floquet channel needs to be studied as the initial wavefunction is expanded in the fourier basis[6]:

$$\Psi(\vec{r},t) = \sum_{n=-\infty}^{\infty} e^{in\omega t}[e^{-i\hat{H}_f(\vec{r})t}\Psi_i(\vec{r},t_0)]_n \tag{3.42}$$

This Floquet Matrix is an infinite order matrix in both space and time co-ordinate. Thus, storage of such a large matrix and time needed to evaluate the exponential of the matrix using the Taylor series are both very large. Thus, this is a really difficult task computationally. In the next section, a method is provided to reduce the complexity of the problem both in space and time using certain analytical expressions and approximations.

3.6 A Memory and Time saving Algorithm for (t,t') method

If the Time Dependent Schrödinger equation of an atom kept in an oscillating field with frequency ω is considered ($\hbar = 1$):

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t} = [-\frac{\nabla^2}{2} + V(\vec{r}) + \vec{r}.\vec{\epsilon}]\Psi(\vec{r},t) \tag{3.43}$$

Here

$$H_0 = -\frac{\nabla^2}{2} + V(\vec{r}) \tag{3.44}$$

$$D = \frac{\vec{\epsilon}.\vec{r}}{2} \tag{3.45}$$

The TDSE becomes

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t} = [H_0 + 2D]\Psi(\vec{r},t) \tag{3.46}$$

Now the Floquet type Hamiltonian can be written in a block tridiagonal form as:

$$[H_f] = [H_0 + n\omega]_{n,n'} + [D]_{n,n'\pm 1} \tag{3.47}$$

The H_f matrix for 3 floquet channels is written as:

$$[H_f]_{n_f\times n_x, n_f\times n_x} = \tag{3.48}$$

$$\begin{bmatrix} [H_0+2\omega]_{n_x\times n_x} & [D]_{n_x\times n_x} & 0 & 0 & 0 \\ [D]_{n_x,n_x} & [H_0+\omega]_{n_x,n_x} & [D]_{n_x,n_x} & 0 & 0 \\ 0 & [D]_{n_x,n_x} & [H_0]_{n_x,n_x} & [D]_{n_x,n_x} & 0 \\ 0 & 0 & [D]_{n_x,n_x} & [H_0-\omega]_{n_x,n_x} & [D]_{n_x,n_x} \\ 0 & 0 & 0 & [D]_{n_x,n_x} & [H_0-2\omega]_{n_x,n_x} \end{bmatrix}$$

Where the inner matrices are of the order of (n_x, n_x). The total Floquet matrix is of the order of $(n_f \times n_x, n_f \times n_x)$, n_f being the Floquet matrix dimension. If both $\{n, n_x\}$ $\rightarrow \infty$, the Floquet matrix will become very difficult to store. Also, the exponential expansion of such a large matrix will require a lot of time for calculations as with the number of iterations for convergence of the expansion $(n \times n_x)^3$ number of operations add up.

The first step is to separate the number matrix from the Floquet matrix.

$$[H_f] = [H_{\mathcal{F}}] + [H_{num}] \tag{3.49}$$

which will look like

$$[H_f] = \begin{bmatrix} [H_0] & [D] & 0 & 0 & 0 \\ [D] & [H_0] & [D] & 0 & 0 \\ 0 & [D] & [H_0] & [D] & 0 \\ 0 & 0 & [D] & [H_0] & [D] \\ 0 & 0 & 0 & [D] & [H_0] \end{bmatrix} + \begin{bmatrix} [2\omega] & 0 & 0 & 0 & 0 \\ 0 & [\omega] & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & [-\omega] & 0 \\ 0 & 0 & 0 & 0 & [-2\omega] \end{bmatrix} \tag{3.50}$$

Now, the block tridiagonal matrix $H_{\mathcal{F}}$ has a certain symmetry which can be used to diagonalize it using Coulson's transformations[10]. The analytic expressions of the unitary matrices are obtained which effectively reduces the storage space to only the (n_x, n_x) order matrices as a full diagonalization is not required.

$$H_{\mathcal{F}} = U^{\dagger} H_d U \tag{3.51}$$

The diagonal matrix H_d is of the form:

$$H_d = \tag{3.52}$$
$$\begin{bmatrix} [H_0 + \vec{r}.\vec{\epsilon}cos(\tau_1)] & 0 & 0 & 0 & 0 \\ 0 & [H_0 + \vec{r}.\vec{\epsilon}cos(\tau_2)] & 0 & 0 & 0 \\ 0 & 0 & [H_0 + \vec{r}.\vec{\epsilon}cos(\tau_3)] & 0 & 0 \\ 0 & 0 & 0 & [H_0 + \vec{r}.\vec{\epsilon}cos(\tau_4)] & 0 \\ 0 & 0 & 0 & 0 & [H_0 + \vec{r}.\vec{\epsilon}cos(\tau_5)] \end{bmatrix}$$

where $tau_k = \frac{k\pi}{n_f+1}$, k=1,2,...n_f. The Unitary matrix elements are given as

$$U_{ij} = \sqrt{\frac{2}{n_f+1}} sin(i\tau_j) \tag{3.53}$$

The unitary matrix is written as:

$$[U] = \begin{bmatrix} U_{11}[I]_{nx.nx} & U_{12}[I]_{nx,nx} & U_{13}[I]_{nx,nx} & U_{14}[I]_{nx,nx} & U_{15}[I]_{nx,nx} \\ U_{21}[I]_{nx.nx} & U_{22}[I]_{nx,nx} & U_{23}[I]_{nx,nx} & U_{24}[I]_{nx,nx} & U_{25}[I]_{nx,nx} \\ U_{31}[I]_{nx.nx} & U_{32}[I]_{nx,nx} & U_{33}[I]_{nx,nx} & U_{34}[I]_{nx,nx} & U_{35}[I]_{nx,nx} \\ U_{41}[I]_{nx.nx} & U_{42}[I]_{nx,nx} & U_{43}[I]_{nx,nx} & U_{44}[I]_{nx,nx} & U_{45}[I]_{nx,nx} \\ U_{51}[I]_{nx.nx} & U_{52}[I]_{nx,nx} & U_{53}[I]_{nx,nx} & U_{54}[I]_{nx,nx} & U_{55}[I]_{nx,nx} \end{bmatrix} \tag{3.54}$$

where $[I]$ is the identity matrix. Here, for n floquet channels only an (n_f, n_f) matrix needs to be made as many of the element are zeroes of repetition of themselves.

The Floquet matrix is now of the form:

$$H_f = U^\dagger H_d U + H_{num} \tag{3.55}$$

As the number matrix and the $H_{\mathcal{F}}$ do not commute, the split operator approximation is applied to take the exponential of the Floquet matrix.

$$e^{-iH_f t} = e^{-i(H_{\mathcal{F}}+H_{num})t} \tag{3.56}$$

$$e^{-iH_f t} = e^{-iH_{num}t/2} e^{-iU^\dagger H_d U t} e^{-iH_{num}t/2} \tag{3.57}$$

which can be written as:

$$e^{-iH_f t} = e^{-iH_{num}t/2} U^\dagger e^{-iH_d t} U e^{-iH_{num}t/2} \tag{3.58}$$

The exponential of the number matrix is trivial to calculate. It can be seen that

$$(e^{-iH_{num}t/2} U^\dagger)^T = U e^{-iH_{num}t/2} \tag{3.59}$$

which further reduces our storage space and number of calculations. The next part deals with the calculation of the exponential of the block diagonal matrix H_d Using equation(3.49) the split operator method each element of the matrix H_d can be written as:

$$(H_d)_{ii} = e^{-i(V(\vec{r})+\vec{r}.\vec{\epsilon}cos(\tau_i))t/2} e^{-i(-\nabla^2/2)t} e^{-i(V(\vec{r})+\vec{r}.\vec{\epsilon}cos(\tau_i))t/2} \tag{3.60}$$

Another transformation is needed to convert the exponential of the kinetic energy into a diagonal form. So equation (3.65) can now be written as:

$$(H_d)_{ii} = e^{-i(V(\vec{r})+\vec{r}.\vec{\epsilon}cos(\tau_i))t/2} U_1^\dagger e^{-i(-p^2/2)t} U_1 e^{-i(V(\vec{r})+\vec{r}.\vec{\epsilon}cos(\tau_i))t/2} \tag{3.61}$$

where $-\frac{p^2}{2}$ is the kinetic energy Hamiltonian in the fourier basis with $p_k = \frac{2\pi k \Delta k}{L}$, $k = \{-\infty, \infty\}$. In this algorithm the wavefunction is Fast Fourier transformed into the momentum space and after multiplying with the exponential of the kinetic energy matrix it is back transformed into the position space.

$$(H_d)_i = e^{-i(V(\vec{r})+\vec{r}.\vec{e}cos(\tau_i))t/2}[fft]e^{-i(-p^2/2)t}[ifft]e^{-i(V(\vec{r})+\vec{r}.\vec{e}cos(\tau_i))t/2} \tag{3.62}$$

where ifft → inverse Fast Fourier transform, and fft → Forward Fast Fourier transform. These transforms were performed using the **FFTW** package.

Every matrix is acted one by one upon the initial wavefunction. So, every step has a maximum order of operation count of N^2 as the complexity is only matrix vector multiplication.

The full algorithm can be described as:

$$\Psi(r,t) = e^{-in\omega\Delta t}U^{\dagger}\begin{bmatrix} e^{-i(H_d)_1\Delta t} & 0 & 0 \\ 0 & e^{-i(H_d)_2\Delta t} & 0 \\ 0 & 0 & e^{-i(H_d)_3\Delta t} \end{bmatrix} Ue^{-in\omega\Delta t}(\Psi(r,t_0)_n)$$

$$\Psi(r,t) = e^{in\omega t'}[\Psi(r,t)]_0, \Psi(r,t) = \Psi(r,t',t)|_{t=t'}$$

3.7 Testing of the algorithm

The code is written in **FORTRAN90** [3] and **LAPACK** [1] and **FFTW**[2] libraries are used.

3.7.1 Forced Harmonic Oscillator

The Forced Harmonic Oscillator equation is given by:

$$H(r) = -\frac{1}{2m}\frac{\partial^2}{\partial r^2} + \frac{m\omega^2}{2}r^2 - rEcos(\alpha t) \tag{3.63}$$

When the initial solution of this equation is a gaussian wavepacket. It remains a Gaussian Wavepacket throughout time.

Here, Error bounds are calculated against a gaussian wavepacket suggested by Heller[12] with the parameters taken from David Lauvergnat [11] and co-workers' paper.

The Gaussian wavepacket is of the form:[12]

$$gwp(r,t) = exp(i.a_t(r-r_t)^2 + i.p_t(r-r_t) + i.\phi_t) \tag{3.64}$$

Let $d = (\omega^2 - \alpha^2)$ where the width

$$\alpha_t = i\frac{m\omega}{2} \tag{3.65}$$

the impulsion

$$p_t = -\frac{E}{d}[\alpha sin(\alpha t) - \omega sin(\omega t)] \tag{3.66}$$

The average position

$$r_t = \frac{E}{md}[cos(\alpha_t) - cos(\omega t)] \tag{3.67}$$

The phase

$$\phi_t = iln\left[\sqrt[4]{\frac{m\omega}{\pi}}\right] + t\left(\frac{E^2}{4md} - \frac{\omega}{2}\right) \tag{3.68}$$

$$+\frac{E^2}{8\alpha md}(8\alpha^2 cos(\omega t)sin(\alpha t) + (\omega^2 - 3\alpha^2)sin(2\alpha t) - 2\alpha\omega sin(2\omega t)) \tag{3.69}$$

The initial parameters taken from the same paper[11] are: m=10000 a.u. , E= 0.01 a.u., $\alpha = 11/1000 a.u.$ and propagation was done for 40000 a.u.. ($r_0 = 0, p_0 = 0, \alpha_0 = i\frac{m\omega}{2}$ and $\phi_0 = -i\sqrt[4]{\frac{m\omega}{\pi}}$.

The analytical wavepacket Ψ_E was propagated and the error in the overlap with the (t,t') wavepacket was calculated.

$$err_{overlap} = 1 - \langle\Psi|\Psi_E\rangle \tag{3.70}$$

Firstly the error was calculated for different number of Floquet channels at different time steps to check the convergence with respect to number of Floquet channels. The grid was taken from [-4.0:4.0] with number of points in $x = 1001$.(Fig 3.1)

These figures show that irrespective of the time step the error converges with the same number of Floquet channels, around, $N_F = 7$.

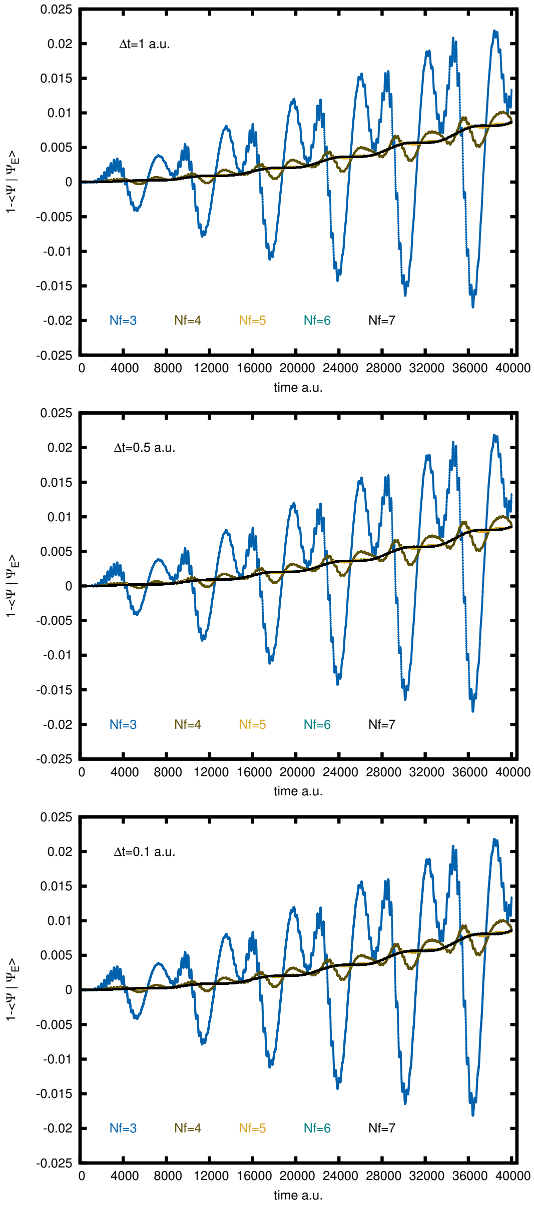

Figure 3.1: Error in the overlap different Δt with different number of floquet channels.

So, now the error in the overlap is calculated by keeping the number of Floquet channels fixed and expanding the basis in x. Larger grid with grid size $\Delta x = 0.004$ are taken. The number of Floquet channels are fixed to $N_F = 8$. figure (3.2).

Quantitative analysis of error is done by calculating the Euclidean Norm and The Max Norm.

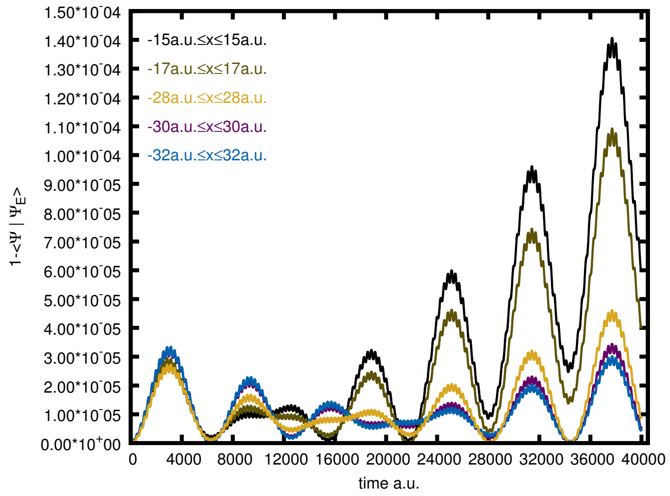

FIGURE 3.2: Error in the overlap with respect to change in the grid size.

The Euclidean Norm is given by

$$||x||_{euc} = \sqrt{\sum_{1}^{N} x_i^2} \tag{3.71}$$

The Max Norm is:

$$||x||_{max} = max(x_1, x_2, x_3, ..., x_N) \tag{3.72}$$

Grid size	Euclidean Norm	Max Norm
[-15:15]	9.57256296×10^{-3}	1.40629042×10^{-4}
[-17:17]	7.29933317×10^{-3}	1.09146903×10^{-4}
[-28:28]	3.27786227×10^{-3}	4.61124168×10^{-5}
[-30:30]	2.88307059×10^{-3}	3.43729180×10^{-5}
[-32:32]	2.74945679×10^{-3}	3.33709260×10^{-5}

TABLE 3.1: Euclidean Norm and Max Norm for error in the overlap

The results show that a very large basis set is required for getting good results. But the error remains unchanged with change in the time step.

The Timings for time propagation are recorded for different number of floquet channels at fixed time step $\Delta t = 1 a.u.$ and fixed Grid size $[-4:4]$ for nx=2001 points. It can be seen that the largest matrix is of the order of $(2001 \times 21, 2001 \times 21)$ and it took very less time to perform the calculations. The timings were recorded on *Dell, 64 bits, OptiPlex 9020 (05A4), version 00, 8 GB RAM, 4 cores*

N_F	Time(sec.)
3	105.619836
4	135.359236
5	168.107580
6	200.686867
7	236.886939
8	270.457813
9	309.217981
10	369.985104

TABLE 3.2: Timings of time propagation for different number of floquet channels at $\Delta t = 1a.u.$

3.7.2 Two-Level System

The population of the second level of a two level system changes with time as:[11]

$$pop_2^{Rabi}(t) = sin^2(\frac{\Omega_{rabi}t}{2}) \tag{3.73}$$

where $\Omega_{rabi} = \mu E$ is the rabi frequency. The same is obtained by $(t, t^{'})$ method for $\mu = 1$ and $E = 1/10000$. The parameters are again taken from Lauvergnat's paper[11].

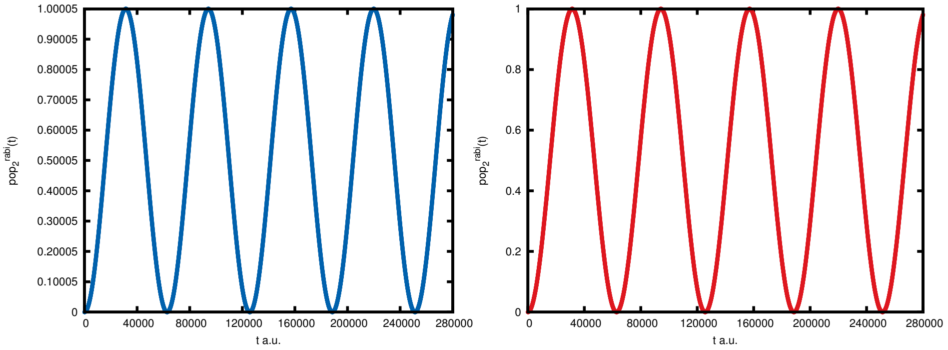

FIGURE 3.3: (a) Analytic population of second states (b) State obtained by (t,t') method

3.8 Future work

We want to extend the algorithm for:

- Inclusion of a pulse.

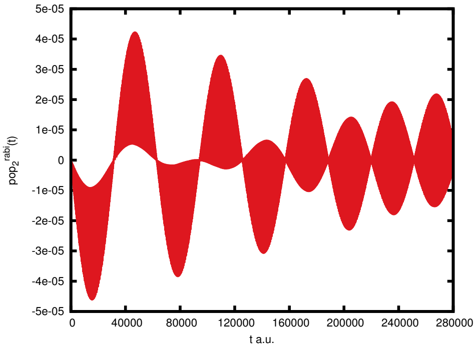

Figure 3.4: difference between the (t,t') population and the analytic population.

- Multiple Dimension.
- Multimode Floquet theory
- Linearly and Circularly polarized lights.

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Appendix A

New algorithm for the (t,t') method: Fortran90 code

The following code is written in **Fortran90** [3] to develop the method proposed in chapter 3. The test problem taken in this code is the forced harmonic oscillator.

A.1 Libraries and Packages used

FFTW[2] (Fast Fourier Transform in the West) - Used For performing Fast Fourier Transformations.

LAPACK[1] (Linear Algebra PACKage) - Used for diagonalization of the matrices.

A.2 Compilers used

ifort[4] (Intel ® Fortran Compiler)

A.3 Compilation Instruction

Use the following command on terminal: ifort -o outfile codefile.f90 $-L/usr/local/lib$ -lfftw3 -llapack

A.4 Code

```
!program to implement the new ttprime algorithm
!Author  ::  Alkit Gugalia
!Date    ::  12/1/2018
       program ttprime_method
       implicit none
       include 'fftw3.f' !FFTW LIBRARY
       !********** variable declaration **********!
       integer (kind = 4), parameter :: nx=2001,ny=1
       integer (kind = 4) :: i, j, k, ii, jj, kk, ix, iy, l, ll, jx, jy, n
       integer (kind = 4) :: m, mm,it,pt
       integer (kind = 8) :: plan
       real (kind = 8) :: dx, dy, x1, x2, y1, y2, x, y , lenx, leny
       real (kind = 8) :: pi,twpi,twpinx,twpiny,twpinlx,twpinly,E,w
       real (kind = 8) :: mass,fconst,findx,tab,start,finish
       real (kind = 8), allocatable :: evecs(:,:),evals(:),psi1(:,:)
       complex (kind = 8), parameter :: io=(0.00000d0,1.00000d0)
       complex (kind = 8) :: const1,const2,iodt
       complex (kind = 8) :: val,mtval,val1,val2,val3,cof(nx,40000)
       complex (kind = 8) :: anal(nx,40001),nom(40000)
       complex (kind = 8), allocatable :: psqx(:),psqy(:),psqz(:),V(:)
       !********** Floquet variables **********!
       integer (kind = 4), parameter :: Nf = 3
       integer (kind = 4) :: flag,totfq
       real (kind = 8) :: tau,omega,ucons
```

```
complex (kind = 8) :: sum1,alpha
complex (kind = 8), allocatable :: coeff(:),nomega(:),coeff1(:)
complex (kind = 8), allocatable :: umat(:,:),floq(:,:)
!********** time variables **********!
integer (kind = 4), parameter :: nt=40000
real (kind = 8) :: t1,t2,dt,t
real (kind = 8) :: ton,pls
complex (kind = 8) :: pconst,pkconst
!********** constants **********!
flag=(2*Nf+1) ; totfq=nx*(2*Nf+1) ; omega=11.0d0/1000.0d0
E=0.010d0 ; w=0.010d0 ; mass=10000.0d0 ; fconst=1.00000d0
pi=3.14159265358979323846200d0 ; twpi=2.00000d0*pi
twpinx=twpi/nx ; n=nx ; tau= pi/(flag+1.00000d0)
ucons=dsqrt(2.0000d0/(flag+1.0000d0))
x1=-4.00d0 ; x2=4.00d0 ; t1=0.000d0 ; t2=40000.00d0
pconst=(0.500d0*pi)/(ton) ; lenx=x2-x1 ; leny=y2-y1
dx=lenx/real(nx-1, kind = 8) ; dy=leny/real(ny-1, kind = 8)
dt=(t2-t1)/real(nt-1, kind = 8) ; twpinlx=twpi/lenx
twpinlx=twpinlx*twpinlx ; iodt=io*dt
!********** fourier grid hamiltonian **********!
allocate(psqx(nx),psqy(nx))
psqx(1)=(1.00d0,0.00d0); alpha=-(iodt*0.5d0*twpinlx)/mass
do i=1,(nx+1)/2
  psqx(i+1)=cdexp(alpha*dfloat(i)*dfloat(i))
  psqx(nx-i+1)=psqx(i+1)
enddo
alpha=(0.00000d0,0.00000d0)
allocate(psi1(nx,nx))
!********** DVR Hamiltonian **********!
call raddvr(x1,nx,dx,psi1)
allocate(V(nx))
```

```
alpha=0.50000d0*fconst
!********** Potential Energy + Kinetic Energy **********!
do i=1,nx
    x = x1 + (i-1)*dx
    V(i)=alpha*(x*x)
    psi1(i,i) = psi1(i,i) + V(i)
enddo
alpha=(0.00000d0,0.00000d0)
allocate(evals(nx),evecs(nx,nx))
!********** Diagonalization for Eigenfunctions **********!
CALL CALL_DSYEV_WT_EIGVECS(psi1,n,evals,evecs)
deallocate(evals,psi1)
!********** Full Floquet wavefunction **********!
allocate(coeff(totfq))
ii=0
kk=0
jj=totfq
do k=1,Nf+1
  ii= Nf + 2 - k
  jj=jj-(nx)
  do i=1,nx
      kk=kk+1
      x=x1 + (i-1)*dx
      coeff(kk)=(evecs(i,1))
      coeff(jj+i)=coeff(kk)
  enddo
enddo
deallocate(evecs)
!********** Bigger Unitary matrix*Number Matrix **********!
ii=0
alpha=iodt*omega
```

```
allocate(umat(flag,flag))
do i=1,flag
  findx=-(flag+1)/2 + i
  nomega(i)=cdexp(-alpha*0.5000d0*findx)
  do j=1,flag
    umat(i,j)=ucons*dsin(i*j*tau)*nomega(i)
  enddo
enddo
allocate(coeff1(totfq))
mm=0;ll=0
!********** Time-Propagation **********!
do it=1,nt
  t= t1 + (it-1)*dt !time increment
  ii=0
  !********** Wavefunction*U(omega) **********!
  do k=1,flag
    do i=1,nx
      ii=ii+1
      val1=(0.0000d0,0.000000d0)
      do j=1,flag
        pt= Nf-flag+j
        kk=(j-1)*nx+i
        val1=val1+(umat(j,k)*coeff(kk))
      enddo
       coeff1(ii)=val1
    enddo
  enddo
  ii=0 ; kk=0
  !********** Exp of KE, PE and Dipole Multiplication ********!
  do k=1,flag
    pkconst=-E*dcos(k*tau)!*cdexp(0.50000*io*(pi))
```

```
    psqy=(0.00000d0,0.000000d0)
    alpha=-iodt*0.500000d0
    do i=1,nx
      x = x1 + (i-1)*dx
      ii=ii+1
      psqy(i)=cdexp(alpha*(V(i)+x*pkconst))*coeff1(ii)
    ! Multiplying by (potential+dipole)/2 term
    enddo
   !********* FAST FOURIER TRANSFORM using FFTW **********!
    !Inverse FFT
  call dfftw_plan_dft_1d(plan,nx,psqy,psqy,FFTW_BACKWARD,FFTW_ESTIMATE)
  call dfftw_execute_dft(plan,psqy,psqy)
  call dfftw_destroy_plan(plan)
  do i=1,nx
    psqy(i)=psqx(i)*psqy(i) !Fourier Grid KE multiplication
  enddo
  call dfftw_plan_dft_1d(plan,nx,psqy,psqy,FFTW_FORWARD,FFTW_ESTIMATE)
  call dfftw_execute_dft(plan,psqy,psqy)
  call dfftw_destroy_plan(plan)
  !Forward FFT
  do i=1,nx
    x=x1 + (i-1)*dx
    kk=kk+1
    coeff(kk)=cdexp(alpha*(V(i)+x*pkconst))*(psqy(i)/nx)
    ! Multiplying by (potential+dipole)/2 term
  enddo
  enddo
  coeff1=(0.0000d0,0.00000d0)
  kk=0 ; ii=0
 !********** Wavefunction*U(omega) **********!
  do k=1,flag
```

```
    do i=1,nx
      ii=ii+1
      val2=(0.0000d0,0.0000d0)
      do j=1,flag
        kk=(j-1)*nx + i
        val2 = val2 +(umat(k,j)*coeff(kk))
      enddo
        coeff1(ii)=val2
    enddo
  enddo
  coeff=(0.0000d0,0.00000d0); coeff=coeff1; ll=ll+1
  do i=1,nx
    cof(i,ll)=coeff((Nf*nx-1)+i)
  enddo
 enddo
 deallocate(psqx,psqy,umat,nomega,V,coeff,coeff1)
end
!DVR grid K.E. MATRIX
subroutine raddvr(a,n,dx,T)
integer (kind = 4) :: n,i,j,s,u
real (kind = 8)  :: a,dx,x,T(n,n)
real (kind = 8)  :: const,pi = 3.14159265358979323846Od0
const = (pi*pi)/(6.00000d0*dx*dx)
do i=1,n
 x = a + (i-1)*dx
 T(i,i) = const
 do j=1,i-1
  s=i-j
  T(i,j)=(((-1.0d0)**s)/((dx*dx)*(s*s)))
  T(j,i)=T(i,j)
 enddo
```

```
       enddo
       T=T/10000.00000d0
       return
       end
       !********** SUBROUTINE for diagonalization using LAPACK *******!
       !-----------------------------------------------------------------------------
!-----------------------------------------------------------------------------!
           SUBROUTINE CALL_DSYEV_WT_EIGVECS(H,ndim,eigvals,eigvecs)
!-----------------------------------------------------------------------------!
            implicit none

        integer      (kind = 4)            :: ndim

        real         (kind = 8)            :: H(ndim,ndim)
        real         (kind = 8)            :: eigvals(ndim)
        real         (kind = 8)            :: eigvecs(ndim,ndim)

        character    (len  = 1),parameter  :: JOBZ="V"
        character    (len  = 1),parameter  :: UPLO="L"
        integer      (kind = 4)            :: LDA
        integer      (kind = 4)            :: LDWORK
        integer      (kind = 4)            :: INFO
        real         (kind = 8),allocatable:: WORK(:)

      LDA = ndim
      LDWORK=3*ndim-1
      allocate(WORK(LDWORK))
      CALL DSYEV(JOBZ,UPLO,ndim,H,LDA,EIGVALS,WORK,LDWORK,info)
      deallocate(WORK)
      eigvecs=H
                    END
```

References

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