

**Trap Loss Due to Nonadiabatic Transitions in
Electrostatically Trapped Ammonia**

by

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Trap Loss Due to Nonadiabatic Transitions in Electrostatically Trapped Ammonia

Thesis directed by Prof. Heather Lewandowski

In AMO (Atomic, Molecular, and Optical) physics, an important part of making precise measurements is obtaining and keeping high phase-space densities of the atoms or molecules being trapped. When working with atoms, nonadiabatic transitions from trapped states to non-trapped states are a key component of trap loss. The theoretical consensus has been that electrostatically trapped molecules, unlike atoms have very low loss rates due to nonadiabatic transitions. However, [1] performed the first experimental study of loss due to nonadiabatic transitions in electrostatically trapped ammonia and claim that they cause significant trap loss. This thesis presents my work on developing a computer simulation to model ammonia in an electrostatic trap and a simple model of trap loss due to nonadiabatic transitions. This thesis will provide background information on the slowing and trapping of molecules and the nonadiabatic transition that causes trap loss.

Dedication

To my mom, dad, and sister.

Acknowledgements

I would like to thank Professor Lewandowski for allowing me to work in her lab and gain invaluable experience. I would also like to thank the entire Lewandowski group for their help and especially Maya Fabrikant who made much of the work done possible and who's code was modified for the simulation.

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Chapter 1

Introduction

1.1 Motivation

The study of cold molecules is an interesting and relatively new field of study. The experimental study of ultra-cold molecules can provide precise measurements of many-body physics, quantum chemistry, fundamental forces and more [2]. Recent developments in the field of AMO physics have allowed for increased versatility in the trapping and manipulation of neutral polar molecules [1]. Once molecules are trapped, low loss rates of molecules from the trap is key for making precise measurements. So, finding the sources of trap loss and determining how to prevent that loss can be useful. Theoretical studies of trap loss due to nonadiabatic transitions had come to the conclusion that the loss rate was negligible at the temperatures of molecules in electrostatic traps [3]. However, in [1], they claim that contrary to previous theoretical expectations, nonadiabatic transitions are the dominant cause of trap loss in electrostatically trapped ammonia. A nonadiabatic transition is an abrupt change of state between adiabatic states whose variables vary relatively slowly with time.

1.2 Introduction to Project

This project involves creating a computer simulation of deuterated ammonia ND_3^{14} being loaded into, and trapped in an electrostatic trap. The simulation also has a very simplistic model for trap loss of trapped ammonia due to nonadiabatic transitions in the ammonia. The goals of the simulation are to get a realistic distribution of molecules within the trap and then to find the loss

rate due to nonadiabatic transitions in order to compare it with loss rate found by Meijer in [1].

1.3 The Stark Effect

When an atom or molecule is in the presence of an electric field, the energy levels become shifted as a result of the interaction between the dipole moment of the atom or molecule and the external electric field. The shifting effect caused by the electric field is called the Stark Effect. This effect is especially large for ND_3 because of the large dipole moment of the molecule [3]. This arises from the geometry of the molecule (Fig. 1.1).

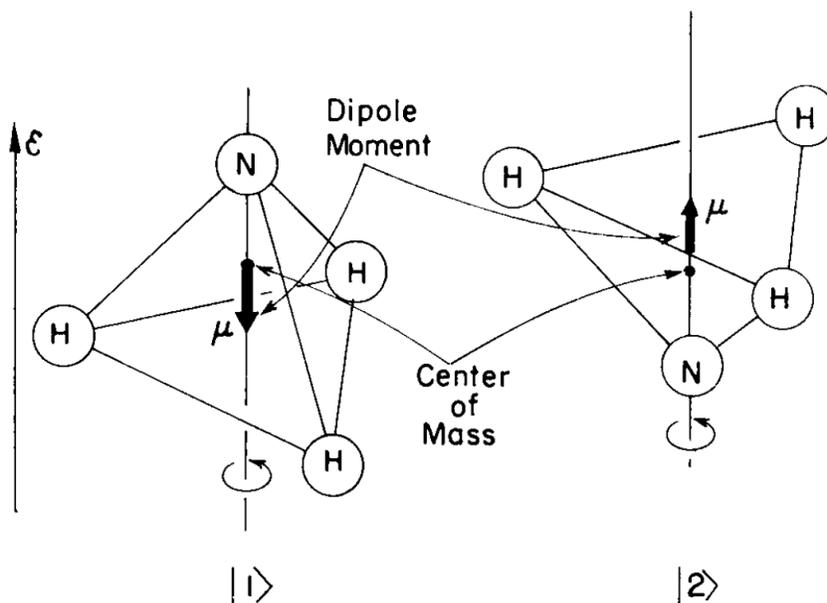


Figure 1.1: This figure shows the geometry of ammonia (although this is NH_3 , the structure is essentially the same for ND_3). The valence electrons on average are closer to the nitrogen atom creating a dipole moment. In this figure, the ammonia molecule is rotating about the axis shown passing through the nitrogen atom and perpendicular to the plane the hydrogen atoms lie in. Thus, the nitrogen atom can be either ‘above’ or ‘below’ the hydrogen atom plane. Those two states create a basis where the dipole moment is either aligned or anti-aligned with the external electric field. This is an approximation as there are rotations and vibrations within the molecule but in the lowest energy states of ammonia this is a reasonable approximation. Reference [4].

In the absence of an electric field, there will be a small probability that the nitrogen will tunnel through the potential barrier created by the hydrogen atom so the Hamiltonian of this system is:

$$H = \begin{array}{c} |1\rangle \\ |2\rangle \end{array} \begin{array}{c|c} |1\rangle & |2\rangle \\ \hline E_0 & -A \\ -A & E_0 \end{array}$$

where A is the coefficient for the tunneling interaction of the nitrogen to flip between states [5]. The perturbation to the Hamiltonian can be written as $-D \cdot \mathcal{E}$ where D is the dipole moment of ammonia and \mathcal{E} is the external electric field. Clearly, this term will be different for the two states shown in the figure.

$$-D \cdot \mathcal{E} = D\mathcal{E} \quad \text{For } |1\rangle \quad (1.1)$$

$$D \cdot \mathcal{E} = -D\mathcal{E} \quad \text{For } |2\rangle \quad (1.2)$$

The perturbed Hamiltonian shows how much this interaction shifts the energy level of the states [5]:

$$H = \begin{array}{c} |1\rangle \\ |2\rangle \end{array} \begin{array}{c|c} |1\rangle & |2\rangle \\ \hline E_0 + D\mathcal{E} & -A \\ -A & E_0 - D\mathcal{E} \end{array}$$

The $|1\rangle$ and $|2\rangle$ states are not stationary as H is not a purely diagonal matrix. Thus, to find the stationary states the Hamiltonian must be diagonalized [5]:

$$\begin{vmatrix} E_0 + D\mathcal{E} - \lambda & -A \\ -A & E_0 - D\mathcal{E} - \lambda \end{vmatrix} = 0$$

The stationary states $|a\rangle$ and $|b\rangle$, the energies of these stationary states E_a and E_b are then calculated from above as:

$$E_a = E_0 + \sqrt{A^2 + D^2\mathcal{E}^2} \quad (1.3)$$

$$E_b = E_0 - \sqrt{A^2 + D^2\mathcal{E}^2} \quad (1.4)$$

These energies have been plotted on figure 1.2. The solutions to the stationary state energies are hyperbolic. For ammonia in high electric fields ($D\mathcal{E} \gg A$), the curves approach their asymptotes:

$$E_a = E_0 + D\mathcal{E} \quad (1.5)$$

$$E_b = E_0 + D\mathcal{E} \quad (1.6)$$

Clearly, at high electric fields the splitting of energy levels no longer depends on A . This is because the high electric field inhibits tunneling [5]. The inhibiting effect can be seen by solving the transition rate between the two states. The transition rate is a function of the difference in energy between the two states and as shown, the energy difference between the two states rises as the magnitude of the electric field rises and thus lowers the rate. However, when the electric fields are lower ($D\mathcal{E} \approx A$) the tunneling interaction still has a major effect. The $|a\rangle$ and $|b\rangle$ states are known as low-field seeking and high-field seeking states. State $|a\rangle$ is the low-field seeking state as its energy is minimized by minimizing the external electric field and state $|b\rangle$ is the high-field seeking state as its energy is minimized by maximizing the electric field.

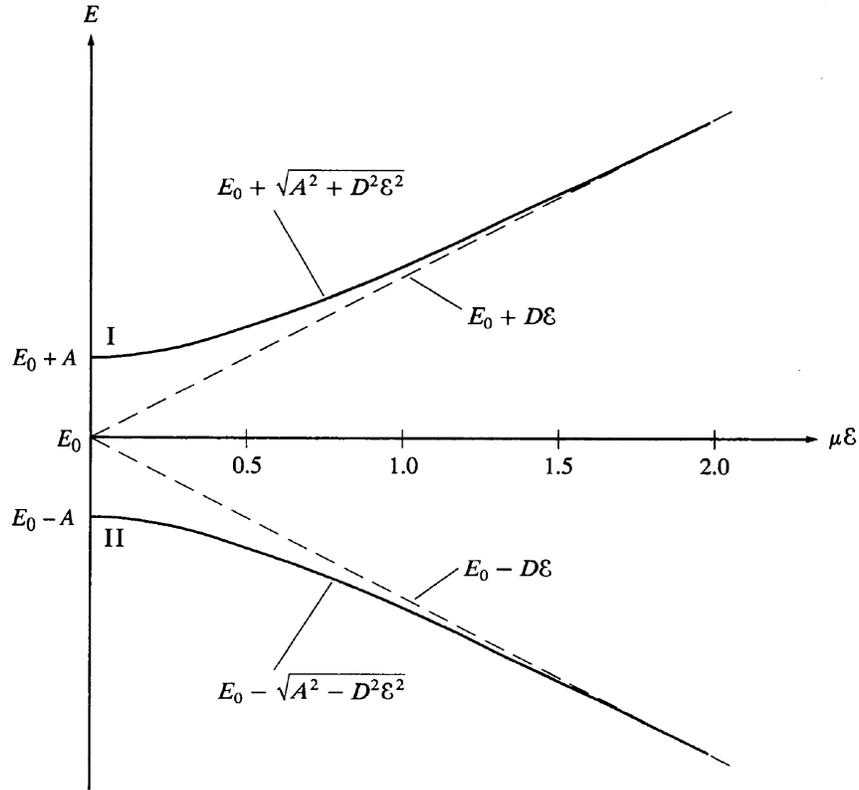


Figure 1.2: This is a graph of the energies of states a and b versus the external electric field. It shows the dominants of the tunneling factor at low external electric field and the asymptotic behavior of the energy as the electric field increases. Reference [5]

1.4 Cooling Ammonia

Lasers are commonly employed to cool atoms to cold and ultracold temperatures. An atom that is moving towards the laser will see the photons at a slightly higher frequency because of the doppler shift and an atom moving away from the laser will see the photons at a lower frequency. Because of the simple electron ground states of atoms, it will only absorb photons of specific wavelengths. So, if the lasers are at a frequency slightly lower than the frequency the atom will absorb, then the atom will only absorb (and thus have momentum transferred from) photons moving towards it that are doppler shifted to the absorption frequency. However, lasers cannot easily cool molecules as the inter-atom interactions cause complications in electron ground states making transferring momentum from the photon and molecule significantly more difficult [6]. Kajita describes three methods for cooling polar molecules: (1) creating molecules out of their constituent atoms that have been pre-cooled; (2) cooling molecules in a cold buffer gas; and (3) creating a beam of cold molecules from using Stark deceleration [3]. The Lewandowski group utilizes stark deceleration to cool ammonia. This is convenient with ammonia as it is strongly affected by the stark effect due to its relatively large dipole moment as mentioned earlier.

1.4.1 Stark Deceleration

Stark deceleration uses the stark effect and the shift in energy of ammonia (and polar neutral molecules in general) to slow it down. The shift in energy from the stark effect is called the stark energy. As shown previously, ammonia in a low-field seeking state will gain stark energy (ie. potential energy) when it enters a region of high electric field. To gain this energy and to conserve energy, the ammonia molecule must lose kinetic energy thus slowing the molecule down.

1.4.2 The Stark Decelerator

Molecules being stark decelerated are first cooled by supersonic expansion by being shot out of a pulsed valve in a supersonic molecular beam. Supersonic molecular beam can be used to create

pulses of molecules that have a mean velocity around a few hundred meters per second but have a small velocity spread around the mean velocity. In the comoving frame of the molecules, this velocity spread corresponds to a temperature distribution around 1K. The stark decelerator lowers this mean velocity to rest so that you are left with a group of cold molecules in the laboratory frame [11]. After the beam has been shot out of the pulsed valve, it first travels through a skimmer. It then passes through a hexapole which focuses the beam effectively ‘transversely cooling’ it [12]. The beam then enters the stark decelerator. The decelerator itself consists of electrode pairs that are oriented orthogonally to its neighboring electrode pairs as seen in figure 1.3. Positive high voltage is applied to one electrode and negative high voltage is applied to the other electrode in the pair creating a high electric field between the electrodes (see figure 1.4). When ammonia in the low-field seeking state enters the high electric field it gains potential energy and loses kinetic energy. If the electrodes were kept at constant potential, then the ammonia molecules would simply exit the region of high electric field losing the potential energy regaining the lost kinetic energy. The key to stark deceleration is discharging the electrodes rapidly eliminating the electric field when a majority of the ammonia molecules are at the maximum electric field of that stage. Thus the molecules lose their stark energy without regaining it as kinetic energy. In the decelerator in the Lewandowski lab, there are around 150 of these stages [7]. After the beam goes through the stark decelerator, the molecules are then loaded into an electrostatic trap.

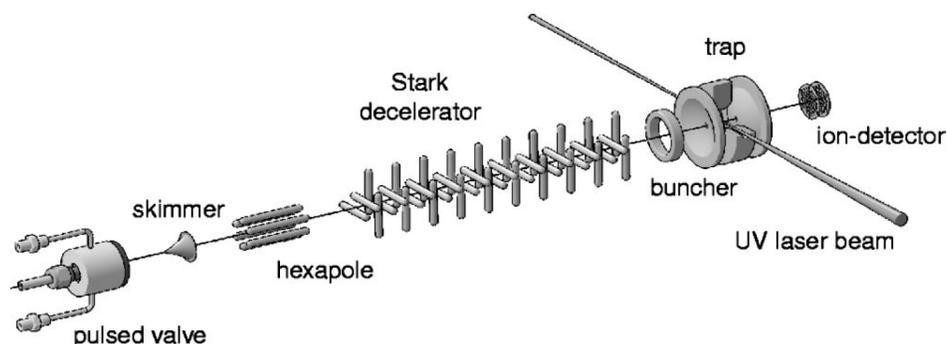


Figure 1.3: A schematic showing a typical experimental setup of a Stark decelerator and electrostatic trap. Reference [8]

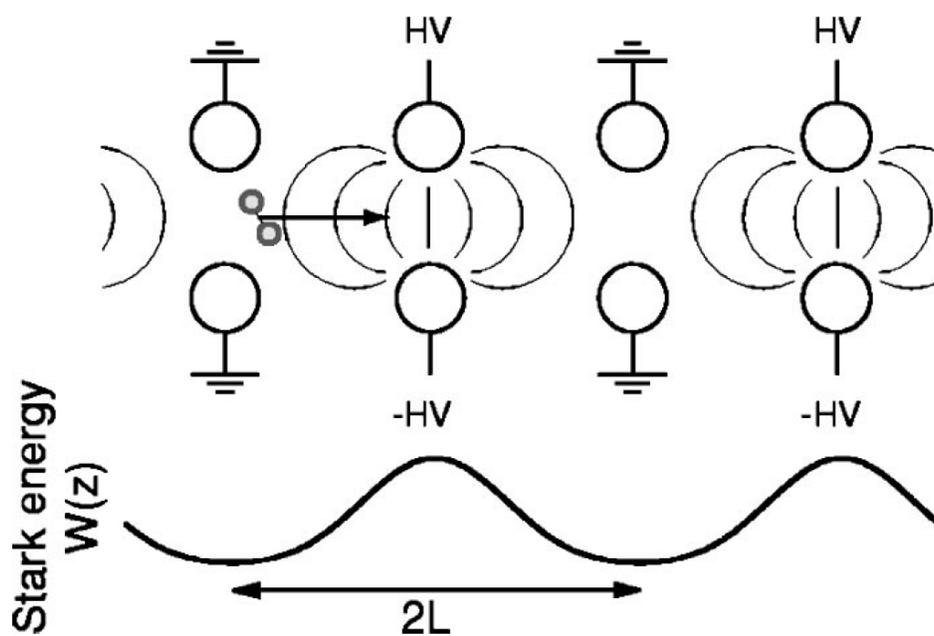


Figure 1.4: A simple diagram of a Stark decelerator with stark energy of the molecule as a function of the position longitudinally down the decelerator at some time [8]. The stark energy is proportional to the electric field and is thus highest between the +HV electrode and the -HV electrode. When the molecules are near the maximum stark energy, the electrodes become grounded to take away the stark energy without the molecule re-gaining kinetic energy. Reference [8]

1.5 Phase Stability Principle

Consider the system in figure 1.4. The energy lost by a molecule being stark decelerated per stage depends on its position in the stark energy $W(z)$ when the electric fields are switched on and off as described in the previous section. The position of a molecule can be expressed in terms of a phase angle φ which has a periodicity of $2L$. A molecule that is at the maximum stark energy when the field is switched off is assigned $\varphi = 90^\circ$ and a molecule that would be unaffected by the switching electric field (ie. is at a position with zero stark energy) is assigned $\varphi = 0^\circ$. The molecule at $\varphi = 0^\circ$ and has a velocity such that it travels a distance L in the time interval between switching it is referred to as the ‘synchronous molecule’. Molecules that have a slightly different phase and velocity will be automatically corrected towards the synchronous molecule. This is because if it has a larger phase, it will be slowed and the phase will become closer to φ_0 the phase of the synchronous molecule. A molecule that has smaller phase will be accelerated to have a larger phase thus closer to φ_0 [6]. So, as a packet of molecules travels down the decelerator molecules will oscillate around the synchronous molecule. When the synchronous molecule is being decelerated, the molecules will oscillate around this deceleration and decelerate with it while staying in a group.

1.6 Electrostatic Trapping

Using electrostatic traps to trap ammonia uses the same stark energy principle as the stark decelerator. A majority of electrostatic traps are configured such that the absolute value of the electric field is zero [1] [8]. As it is the low-field seeking state of ammonia that we are slowing and trapping, it gains stark energy as the electric field increases. Thus, the further from the trap center the ammonia molecule moves, the potential energy increases while the kinetic energy decreases. Thus, similar to a marble in a bowl, molecules with a low enough total energy will remain trapped within the electrostatic trap. This kind of trap works because the stark energy only depends on the magnitude of the electric field as shown in equation 2.5

1.7 Fine Structure

For molecules, the principles of the fine structure are similar to the fine structure of Hydrogen atoms. The fine structure is a corrective perturbation of the Hamiltonian and thus energy levels of atoms and molecules. It is made up of two separate effects, a relativistic correction stemming from the motion of electrons bound to atoms and a correction for the effect of spin-orbit coupling. Spin-orbit coupling arises because electrons have charge $-e$ and spin, they have a magnetic moment μ_e . In the electron's frame, the positively charged nucleus is orbiting around the electron. This moving charge creates a magnetic field which then interacts with the dipole moment of the electron μ_e . This exerts a torque on the electron which causes the energy levels to split based on the spin and orbital quantum numbers.

1.8 Hyperfine Structure

The hyperfine structure is another corrective measure that is typically orders of magnitude smaller than the fine structure. The source of this is similar to the fine structure in that it arises from an interaction between the magnetic moment and a field. With hyperfine splitting, it is the magnetic moment of the proton as it also has a charge e and spin. This interacts with the dipole moments of the electrons in what is called spin-spin coupling. The nucleus with angular momentum I couples with the total angular momentum of the electron J and gives the total angular momentum of the system [9] [10].

$$F = I + J \quad (1.7)$$

For Deuterated ammonia specifically, with I_N as the nuclear spin of a Nitrogen atom and I_D as the nuclear spin of the deuterium [10]:

$$F_1 = J + I_N \quad F = F_1 + I_D \quad (1.8)$$

K is the projection of the total angular momentum J onto the central axis passing through the nitrogen atom and the plane of hydrogen atoms (figure 1.5). A useful way to label three hyperfine

states for ammonia is by combining the MF and K states into an MK label such that the hyperfine levels can be separated into $MK = +1$, $MK = -1$, and $MK = 0$ levels. The $MK = \pm 1$ hyperfine levels correspond to high and low field seeking states while the $MK = 0$ hyperfine levels are almost unshifted in an external electric field as shown in figure 1.6 [1].

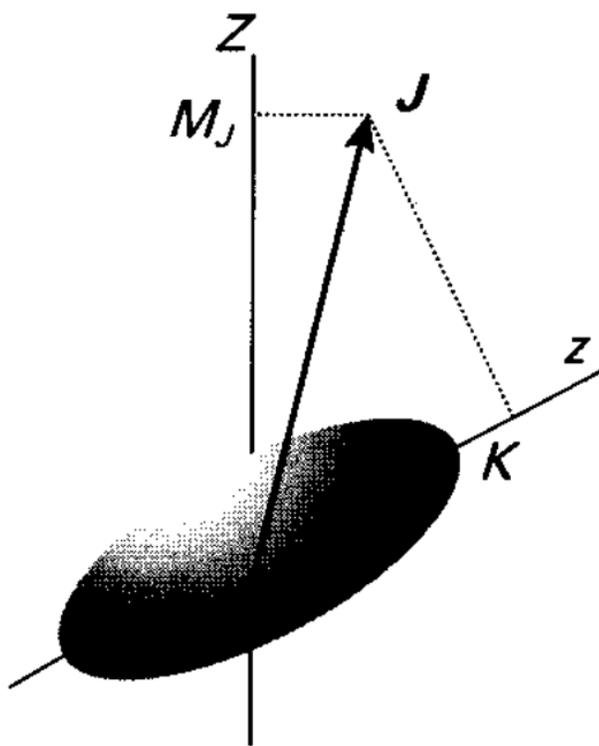


Figure 1.5: This figure shows an example of angular momenta of a molecule and K , the projection of the total angular momentum onto the central axis. Reference [14]

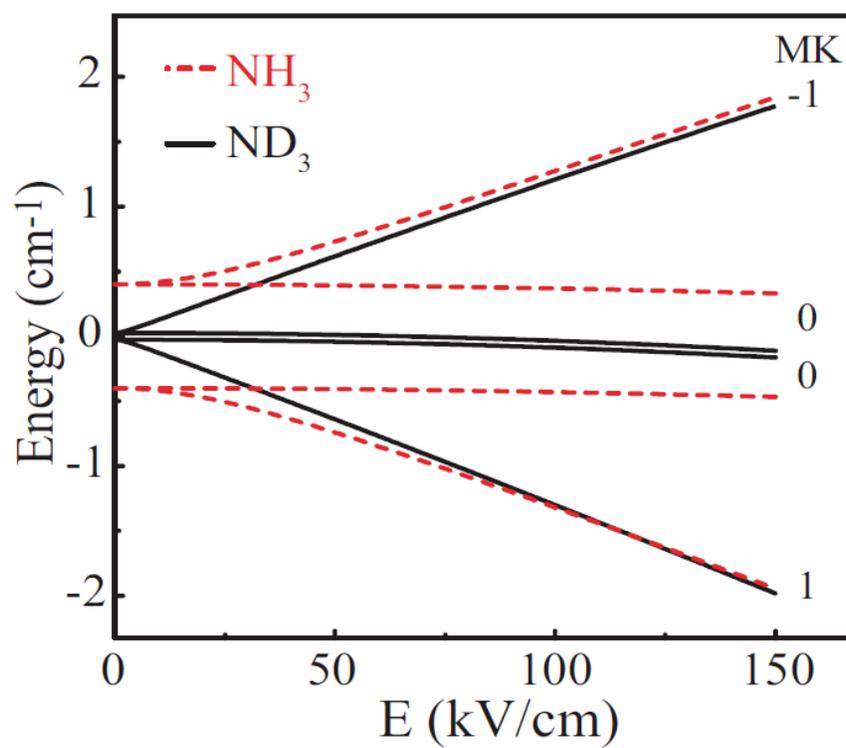


Figure 1.6: This figure shows the energy versus the external electric field of the three MK hyperfine levels of ammonia and deuterated ammonia. Reference [1]

1.9 Loss Causing Transitions

1.9.1 Majorana Transitions

The Majorana transition is an M quantum number changing transition which is caused by a change in the direction of the electric field. This transition can change the state of an ammonia molecule to a high-field seeking state [10]. If a trapped ammonia molecule undergoes this transition, it will no longer be attracted towards the trap center and is thus lost from the trap. In his paper [3] Kajita calculates the Majorana transition rate for ND_3 as a function of electric field and gives a plot (figure 1.7). Because the electric field in electrostatic traps are so large (maximum on the order of 10 kV/cm) and rise so quickly it is clear that for much of the trap, the rate at which molecules are lost due to the Majorana transition is very small and negligible. Not only that, but his calculated rate does not apply for electric fields lower than 100V/cm because the energy state is described mostly by the hyperfine structure of the molecules state [3]. Thus, the Majorana transition will not be the cause of most of the trap loss due to nonadiabatic transitions.

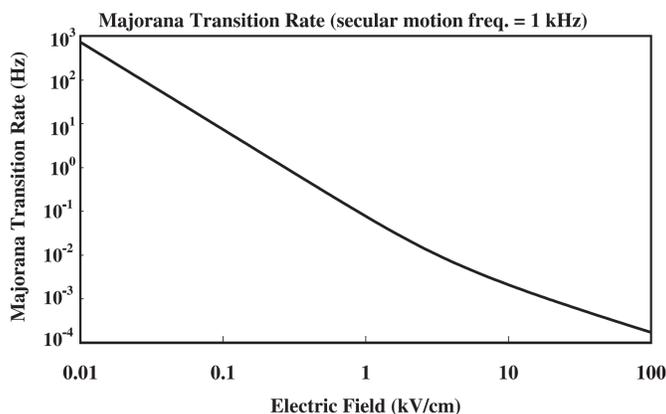


Figure 1.7: This figure shows the rate at which Majorana Transitions occur in ammonia in an external electric field. Reference [3]

1.9.2 Transitions Between Hyperfine Levels

Ammonia in the three sets of hyperfine levels $M_K = +1$, $M_K = -1$, and $M_K = 0$ interact differently with external electric fields. The $M_K = -1$ levels correspond to the low-field seeking

states, the $MK = +1$ levels the high-field seeking states, and the $MK = 0$ levels are essentially unshifted in electric fields and gain little Stark energy. In a similar manner to the Majorana transitions, nonadiabatic transitions of $\Delta MF = 0$ and $\Delta MF = \pm 1$ can occur if the change in electric field direction is too sudden or rapid and the molecule cannot follow the field. If a molecule with a velocity v is at a small distance from the trap center (the spot where there is zero electric field) x then a transition between hyperfine levels can occur. Particularly, if the ratio v/x is greater than the energy difference between hyperfine levels (in the proper units) then the transition has a chance of occurring [1]. For ammonia in an electrostatic trap, the transition from the $MK = -1$ levels to the $MK = 0$ levels in particular will cause trap loss as it will transition from a low-field seeking state to a state basically unaffected by the electric field of the trap. This transition is the transition which the Meijer group claims is responsible for major trap loss from nonadiabatic transitions.

1.10 Previous Experimental Work

In their paper [1], the Meijer group present their findings from their experimental study of trap loss of electrostatically trapped ammonia due to nonadiabatic transitions. To find the loss rate, they compared the trap lifetimes of two different trapping configurations. As shown in figure 1.8 a), their trap consists of six cylindrical electrodes. The four outer electrodes with a diameter of 3mm, had a high positive and negative voltages ($\pm V_Q$) applied to them to create a quadrupole field. The two inner electrodes are centered the symmetry axis of the trap and they can either be charged to be the same polarity, or opposite polarity at lower voltages ($\pm V_D$). If they are charged to the same polarity, then the trap simply generates a standard quadrupole with zero electric field at the center. But, if they are charged to opposite polarity, then they generate a nonzero offset to the center of the quadrupole field as seen in figure 1.8 a) [1]. Figure 1.8 b) shows the Stark energy of ammonia and deuterated ammonia along the longitudinal axis of the trap. For values of $V_Q = 10$ kV and $V_D = 3$ kV, the trap depth (for ND_3) of the offset trap is 290 mK and the depth of the zero-field trap is 1.2 K [1]. The offset at the center of the trap in the opposite polarity case is 16

kV/cm. Because the electric field is nonzero, molecules in the offset field trap configuration will be unable to undergo the nonadiabatic transition from the $MK = -1$ to $MK = 0$ hyperfine levels. Therefore, by comparing the lifetimes of molecules in the two different trapping configurations the Meijer group was able to find the rate of trap loss due to nonadiabatic transitions.

1.11 Zone of Death Approximation

In order to predict the loss rate due to nonadiabatic transitions, the Meijer group analyzed the trap system by approximating the loss mechanism as a ‘zone of death’. The zone of death refers to a small volume at the center of the trap. Because the probability of a molecule undergoing a trap-loss causing nonadiabatic transition is high within the small volume around the trap center, for the approximation, it is assumed that the molecule is lost. Thus, for a volume with a cross sectional area of b^2 near the center of the trap, the loss rate was approximated by $f_{osc}(\frac{b}{b_o})^2$ with f_{osc} as the frequency of oscillation of trapped molecules and b_o being the mean value of the impact parameter of the trajectories of the trapped molecules. The Meijer group used values of f_{osc} around 1-2 kHz, b_o about 0.2mm and b around 5 μ m. This led to an approximate loss rate on the order of 1 Hz [1].

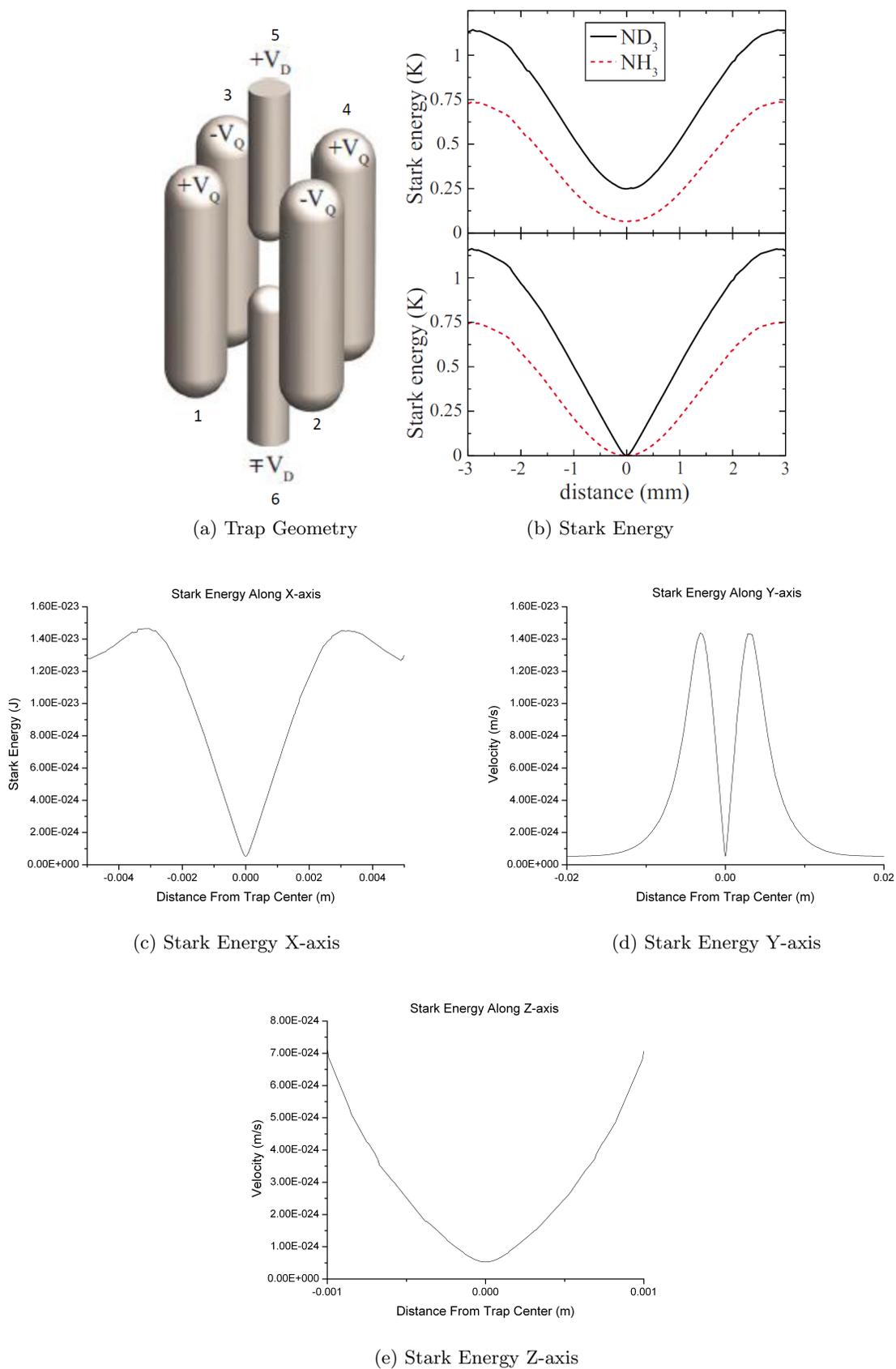


Figure 1.8: a) Shows the geometry of the trap used in [1] and b) shows the Stark energy along the longitudinal axis of that trap. c), d), and e) show the Stark energy of the trap from the model of the trap shown in a created in matlab. a) and b) Reference [1]

Chapter 2

Simulation and Work

This chapter outlines the simulation, the work done with it, and an analysis of the results.

2.1 Overview of Simulation

A matlab program was created to simulate loading cooled ND₃ into an electrostatic trap and the motion of ND₃ after it had been loaded into the trap. Specifically, the simulation models ND₃ trapped in the trap used in the experiments detailed in [1] shown in figure 1.7. This was done with the goal of implementing a zone of death within the simulated trap in order to measure the rate of molecules lost from the zone of death to compare that rate with the rate predicted and measured in [1].

2.2 Simulating The Motion of Molecules

In order to solve the differential equation of motion, the simulation used the Euler method. To find the electric field (and thus the Stark potential for ND₃ in a low-field seeking state) at various electrode voltage configurations, a model of the trap used in [1] was created in COMSOL multiphysics. COMSOL then differentiated the Stark potential in x, y, and z directions and divided by the mass of ND₃ to get the acceleration of ND₃ in the x, y, and z directions. This created a grid of accelerations in the x, y, and z directions at given coordinates. The grid consists of points spaced 0.25mm from each other. For the trap loading sequence, the grid was a 10mm x 30mm x 10mm which amounts to 192000 points total which were interpolated using the TriScatteredInterp

function in matlab to get a smooth function of acceleration as a function of position. This function of acceleration is what was used in my Euler approximation to solve the new position of my molecules after each timestep. The timestep dt was $1\mu s$ long. A smaller timestep could have been used for better accuracy in the Euler approximation, the time the simulation took to run was directly proportional to the size of the timestep. With $dt = 1\mu s$ running the simulation out to 100ms took roughly 8 hours so making dt smaller would have made running the simulation take an unfeasible amount of time. For $dt > 1\mu s$, the resolution of the simulation would have been too poor to effectively model the zone of death. For a molecule with velocity 1m/s and $dt > 1\mu s$, the spatial steps the molecule would take would be greater than $1\mu m$ and with a zone of death radius of $5\mu m$ the chances that a molecule would pass through the zone of death but not be registered as passing through are high.

2.3 Loading the Trap

The loading of molecules into electrostatic traps causes great loss from the initial decelerated packet of molecules. Unlike the deceleration process where low losses are guaranteed by phase stability, the loading process has very large loss associated with it as phase stability no longer holds true for the low velocities of post-decelerated molecules [13]. To accurately test the zone of death model, it is key to have a realistic distribution of molecules in the trap. To get this distribution, the program was used to simulate the loading of the trap. When molecules exit the Stark decelerator, they typically come out with a mean velocity of around 20m/s. For optimal loading of the trap, one slows the ‘synchronous’ molecule to zero velocity at the center of the trap and then turns the trap on. The synchronous molecule is the molecule that has the mean velocity and position of the initial distribution. To slow the molecules, one uses a sequence of different voltage configurations on the trap to create different Stark potential hills to slow the molecules to a near stop near the trap center. My sequence involved three different stages. The ‘off’ stage had no charge on any electrode, giving me zero field everywhere. For the ‘on-back’ stage, (as numbered in figure 1.4) electrode 1 was at -6 kV, electrode 2 at +6 kV, electrode 3 at +15 kV, electrode 4 at -15 kV, electrode 5 at -3

kV, and electrode 6 at +3 kV. That created a Stark potential along the longitudinal axis shown in figure 2.1. The ‘trap-on’ stage was charged as shown in figure 1.4 with $V_Q = 10$ kV and $V_D = 3$ kV with both center electrodes having the same polarity to create zero field at the trap center.

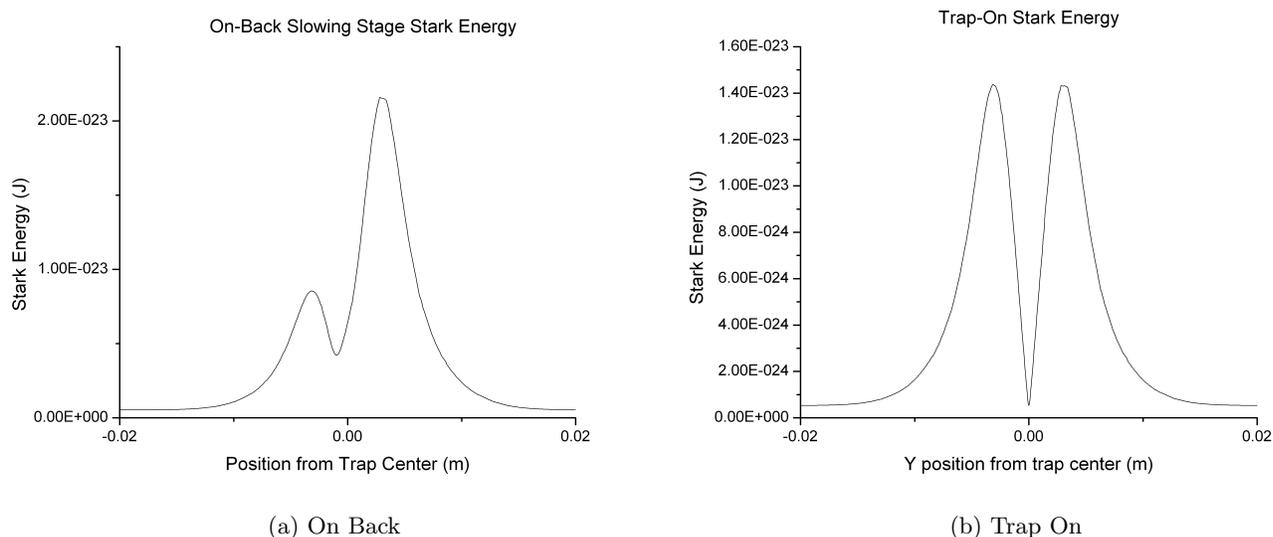


Figure 2.1: These graphs shows the Stark Energy of ND_3 as a function of the longitudinal position from the trap center for the two stages of the slowing sequence.

The sequence used to load the molecules in my simulation was Trap Off from 0 - 0.096ms, Trap On from 0.096ms - 0.25ms, Trap Off from 0.25ms - 0.448ms, On Back from 0.448 - 0.588ms, and finally Trap On at 0.588ms. While there are many different sequences that will trap some molecules, many of those sequences are sub-optimal for trap loading as they lose many of the molecules in the pack during the sequence. For instance, if the first Trap Off stage is too long, the phase space distribution of the molecular packet become stretched into a cigar shape as the molecules with higher velocity rush ahead and the molecules with lower velocity are left behind. This can be seen in figure 2.4 d). Another difficulty, is that if the slope of the Stark energy is too steep, molecules with lower velocity can simply be reflected off of the slowing potential and lost. Because of the velocity spread of the initial incoming molecules, there was a sizable fraction of the packet that was at untrappable velocities. This was not because they could not be slowed to be trapped. Because they were significantly higher velocity than the synchronous molecule, they were

not slowed very much and thus exited the trapping region of the trap during the trap off periods of the sequence. This can be seen in figure 2.4 c) and d) as the molecules with a velocity greater than 20 m/s are quickly moving away from the trap center and are beyond the slowing potential of the on-back stage. The loss of these molecules is also shown in figure 2.2. The majority of molecules that are being lost must be the high-velocity molecules moving in the positive longitudinal direction because figures 2.4 a-f show that fewer molecules had negative velocity and were lost due to being reflected off of the potential wall. To find trapping sequences which worked, initially only the motion of the synchronous molecule was modeled with the goal of finding a sequence which slowed the synchronous molecule to a stop at the trap center. The reason for this was two-fold. First, because the synchronous molecule describes a molecule at the center of the initial distribution, by bringing the synchronous molecule to a stop at the trap center, you can maximize the number of molecules that are trapped. This is because most of the molecules had a velocity and position near the synchronous molecule (figure 2.3). Second, because the zone of death is a very small volume around the trap center, it is important that molecules are able to get near the trap center. If the synchronous molecule still has a fairly high velocity and is not near the trap center, then many of the molecules will simply rotate around the trap center without getting close enough to fall into the zone of death. Thus, having a realistic and well-trapped distribution was important and a key to that was slowing the synchronous molecule well. A major difficulty with getting the synchronous molecule to stop at the trap center was motion in the transverse direction. Because of the trap geometry, creating a voltage configuration which confined molecules in the transverse z-direction while the molecules were being slowed and not near the center of the trap was impossible (Figure 2.3). Thus, it was not possible to get the synchronous molecule to a complete stop at the trap center. The constraints placed by the trap geometry also created another problem which was that the phase space of packet slowly became more dispersed in the transverse directions. The fact that the transverse phase space was un-constrained is seen in figure 2.6 as the transverse phase space distribution becomes less dense as the distribution is loaded. This decrease in phase space density leads to a decrease in trapped molecules. This distribution of molecules loaded into the

trap was saved and then as the initial distribution in the simulation of the evolution of molecules in an electrostatic trap.

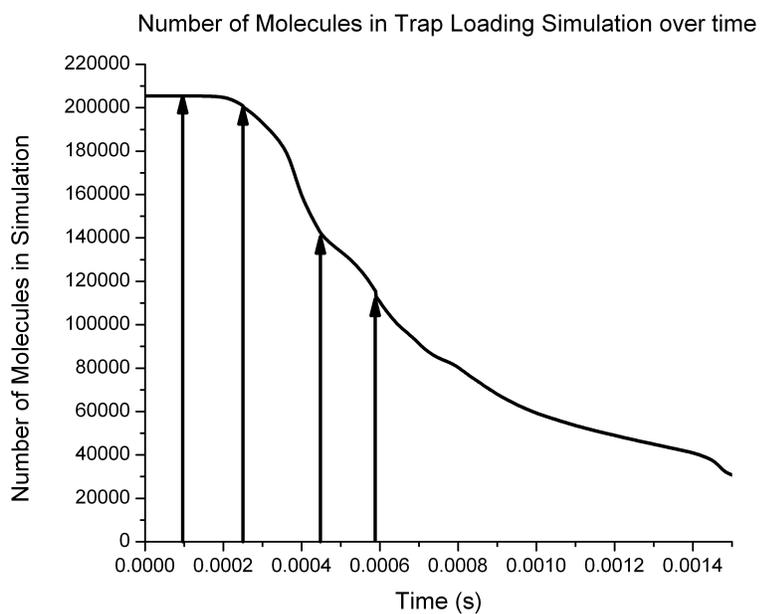


Figure 2.2: This figure shows the number of molecules in the trap loading simulation over time. The arrows point to the switching points in the simulation. It shows a large amount of loss during the third interval. This is because this interval is during the trap off stage so molecules with velocities significantly higher than the synchronous molecule use this trap off stage to quickly leave the simulation. After the last switch point, molecules that were not actually within the trap are slowly lost as the simulation runs.

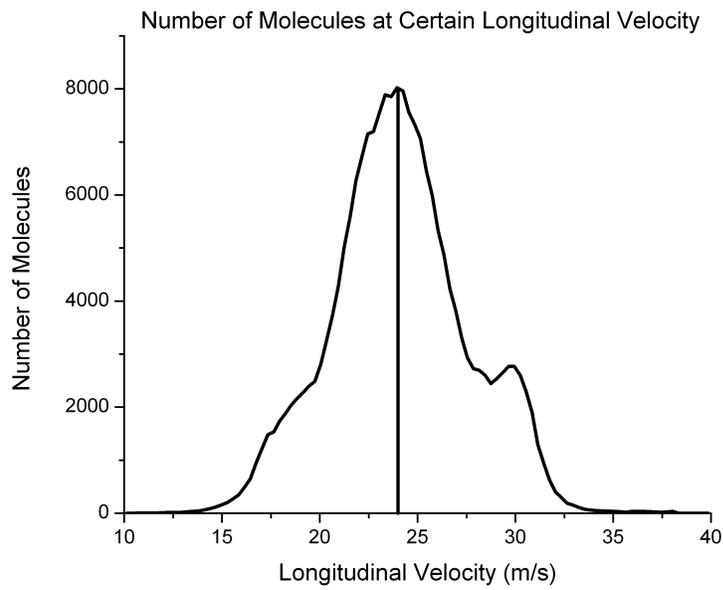


Figure 2.3: This graph shows the number of molecules that are at various longitudinal velocities. The line shows the velocity of the synchronous molecule. This clearly shows that most of the molecules have longitudinal velocity near the synchronous molecule. This is also true for the longitudinal position and transverse phase spaces.

2.4 Applying the Zone of Death

To test the results of the zone of death approximation made in [1], A zone of death was created in the electrostatic trap of the simulation. To do this, the simulation ran a check after every timestep dt to see if the molecules were positioned closer to the trap center than the radius of the zone of death. If a molecule is within the zone of death, then the program removed it from the simulation to simulate the assumption that a molecule entering the zone of death is lost from the trap as it transitions into an $MK = 0$ hyperfine state. It then increased a counter for every molecule lost this way to find the rate at which molecules were lost because of the zone of death.

2.5 Problems With the Simulation

While the trap loading sequence of the simulation appeared to work as shown in figure 2.4, the simulation did not simulate reality when it tried to simulate the evolution of trapped molecules over longer periods of time (100ms). In this section, the problems found in the simulation will be presented along with an analysis that attempts to find the cause of the problems.

2.5.1 Extreme Trap Loss

The problem with the simulation is that it does not appear to actually trap my molecules. The clearest evidence for this is shown in figure 2.7. Because the Zone of Death was disabled in the simulation, the only loss mechanism allowed in the simulation was by exiting the simulated space by leaving the volume enclosed by the trap. This could only occur if the lost molecule had energy greater than the height of the potential well of the trap. So, if the simulation was working properly, no molecules that were within the phase space separatrix of the trapping potential would be lost. Thus, it would appear that most of the ‘trapped’ molecules in the simulation were in fact not trapped at all.

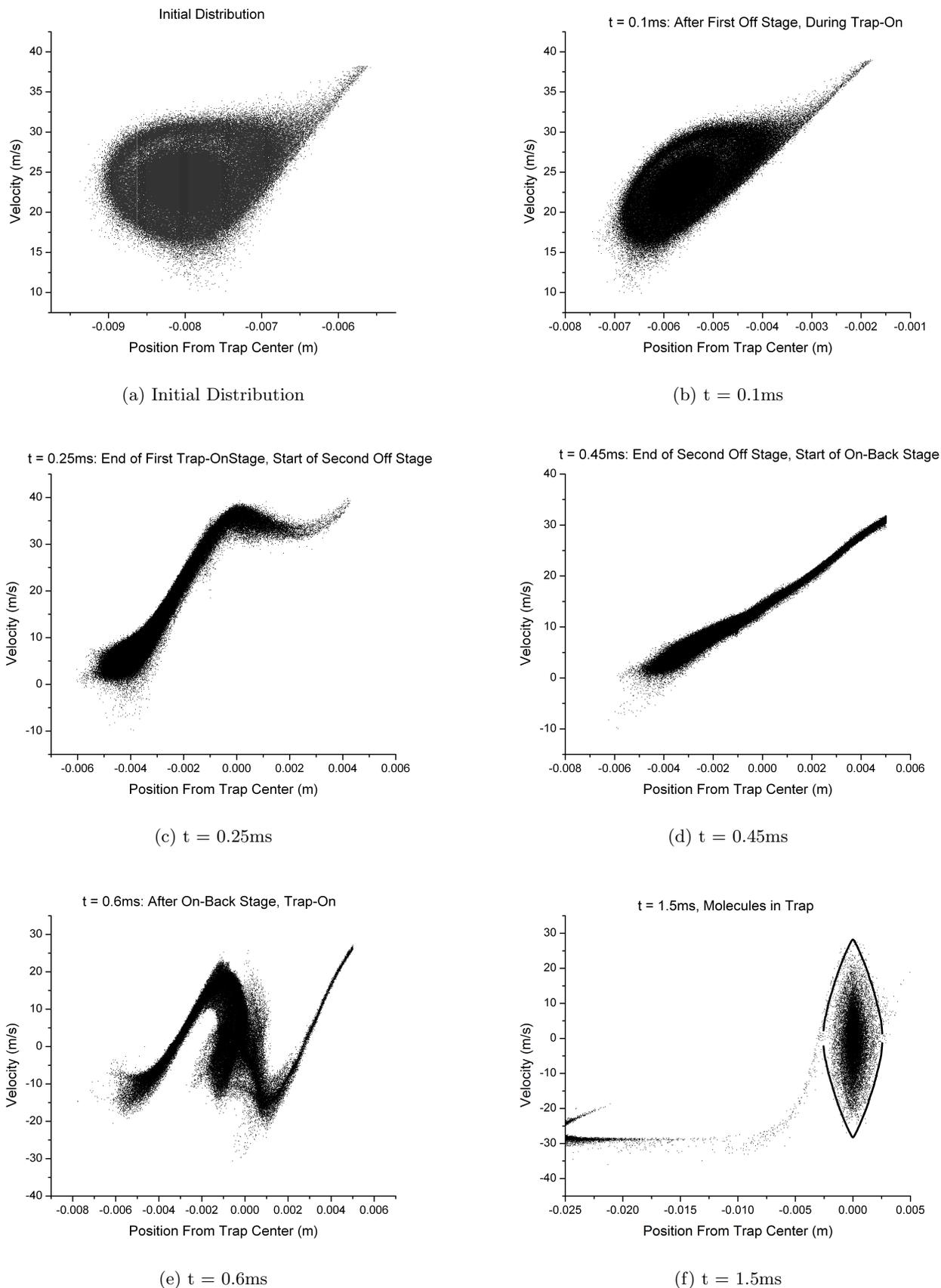


Figure 2.4: This figure shows the longitudinal phase space distribution of the molecules in my trap loading simulation at various times and finally the loaded trap distribution. The times are near or at times when the voltage configurations on the trap were switched. It shows the initial distribution become stretched and eventually some of the molecules become trapped.

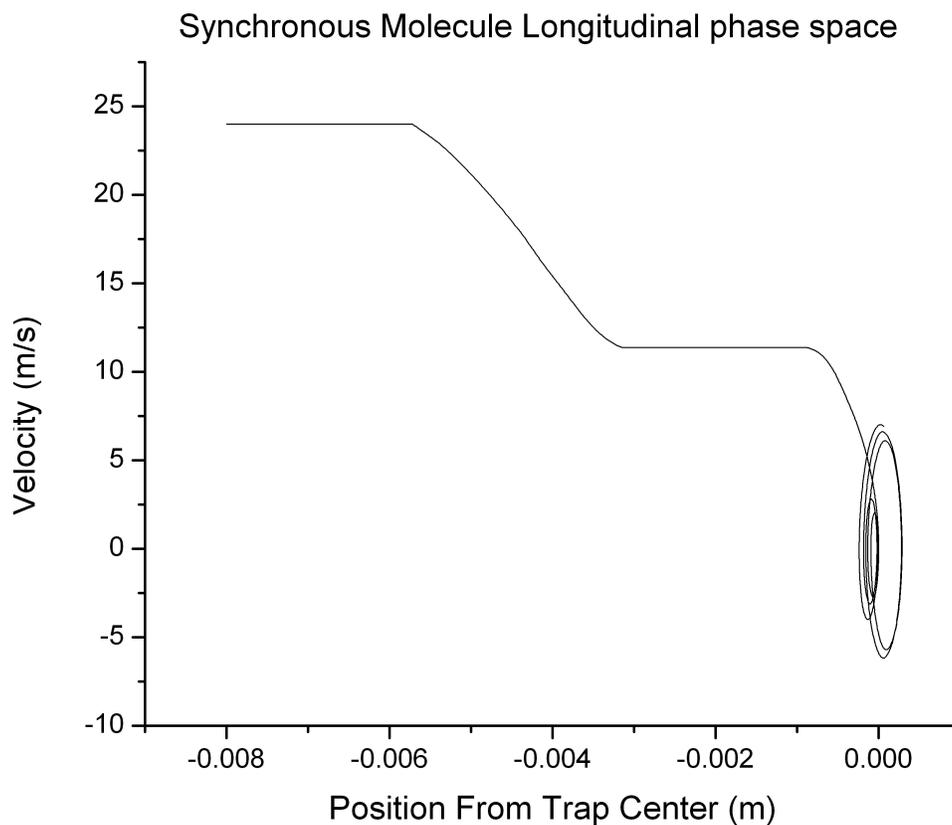
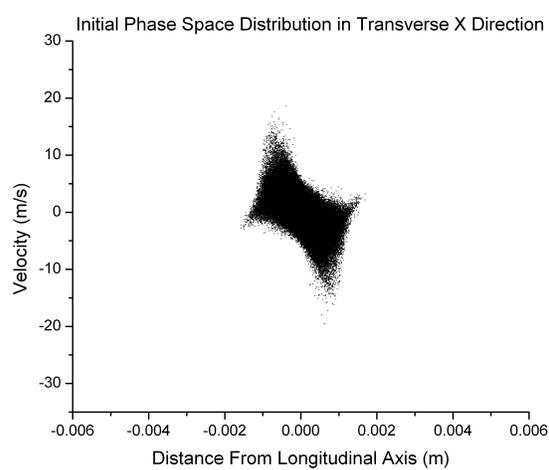


Figure 2.5: This graph shows the phase space trajectory of the synchronous molecule in the longitudinal direction. The sudden changes in slope occur when the slowing configuration switch. When the synchronous molecule is within a few micrometers and has a velocity within a tenth of a meter per second the trap is turned on. If the molecule had no transverse velocity and were perfectly centered on the longitudinal axis when the trap was turned on, then the trajectory of the molecule would circle around a very small area. However, the large circular trajectory of the molecule shows clearly that this is not the case.



(a) Initial X Phase Space

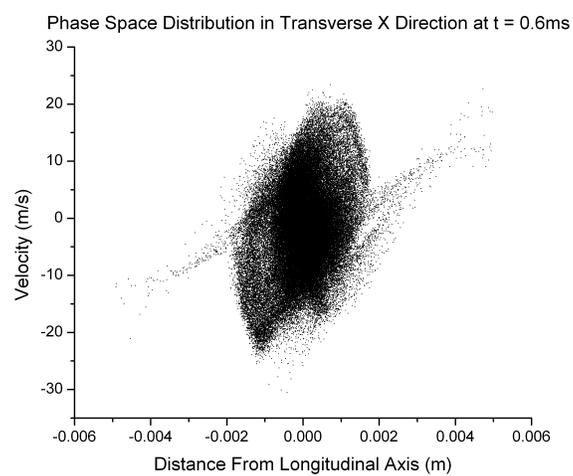
(b) X Phase Space $t=0.6\text{ms}$

Figure 2.6: This figure shows the reduction in phase space density over time as the molecular packet is loaded into the trap. The packet remains centered on the longitudinal axis but because the trap geometry does not allow confinement in the transverse directions, the molecules slowly drift apart.

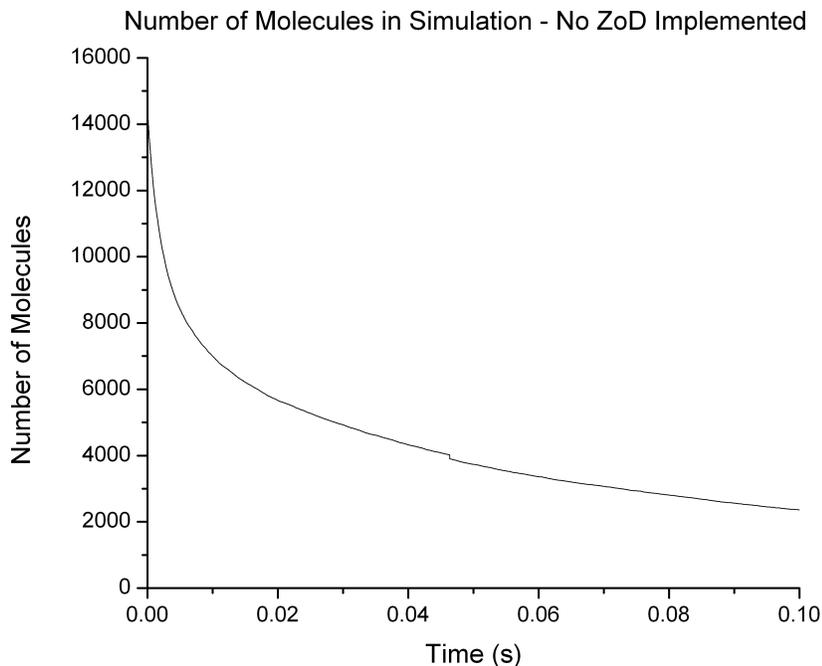


Figure 2.7: This graph shows the number of molecules inside the simulation versus time. This data was created by a simulation that had no Zone of Death implemented. The discontinuous drop in number of molecules at around 0.047s happened because of a different error in my code. 121 of the molecules were set to have the same values as the synchronous molecule so when that left the simulation, 121 other molecules left the simulation at the same time causing a discontinuous drop.

2.5.2 Investigating Loss

To further investigate this, the separatrix of the trap was calculated in the longitudinal phase space and two transverse phase spaces. The separatrix is the line which describes the phase space trajectory of a molecule that has the maximum energy that can still be trapped. Because the system is conservative, it can be said that:

$$H_{max} = T(v) + U(x) \quad (2.1)$$

where H_{max} = the maximum potential energy of the trapping potential in that direction. The velocity as a function of position was then solved:

$$H_{max} = \frac{1}{2}mv^2 + U(x) \quad (2.2)$$

$$v = \pm \sqrt{(H_{max} - U(x))\left(\frac{2}{m}\right)} \quad (2.3)$$

The values for $U(x)$ were taken from the Comsol model of the trap. The separatrix was then plotted onto the initial ‘trapped’ distribution to see if that distribution was in fact trapped. Figure 2.8 shows that a majority of the molecules were within the separatrix and thus, the large loss that the simulation produces should not be present.

An initial distribution in the trap which was well within the calculated separatrix was created to see if molecules which started well within the trap would remain trapped. This was done to see if the the loss could have been caused by anomalous behavior near the peaks of the potential well or if the calculated separatrix was significantly wrong. A distribution which was normal in the 6 phase space dimensions was created to do this. First, the maximum position and maximum velocity of the separatrix were found. Those values were halved and were used as 3σ for the normal distribution to make sure that the molecules were well within the trap. Figure 2.9 shows that this normal distribution is initially well within the boundaries of the trap.

That distribution was then used as the initial distribution in the simulation without the Zone of Death to see if there was a significant difference in the trap lifetime. In figure 2.10, the evolution of the trap is shown for both the normal distribution and the distribution taken from the trap loading simulation. The percentage of molecules lost from the simulation was very similar for both distributions (around 80%). It is thus clear that the trap is not trapping molecules because the number of molecules lost did not change despite the distribution being well within the trapping phase space. This is despite the fact that the trap potential is configured such that molecules should be trapped (figure 1.4 c,d,e). Therefore, there is some error in the simulation.

2.5.3 Causes of Error

The cause of this error is still unknown and currently being investigating. The initial thought had been that it could be from energy drift. Because the simulation was using the Euler method to solve my positions and velocities, there can be a gradual upward drift in the energy of the molecules. Figure 2.11 shows that this could be the cause of trap loss. There is a clear trend of the molecule

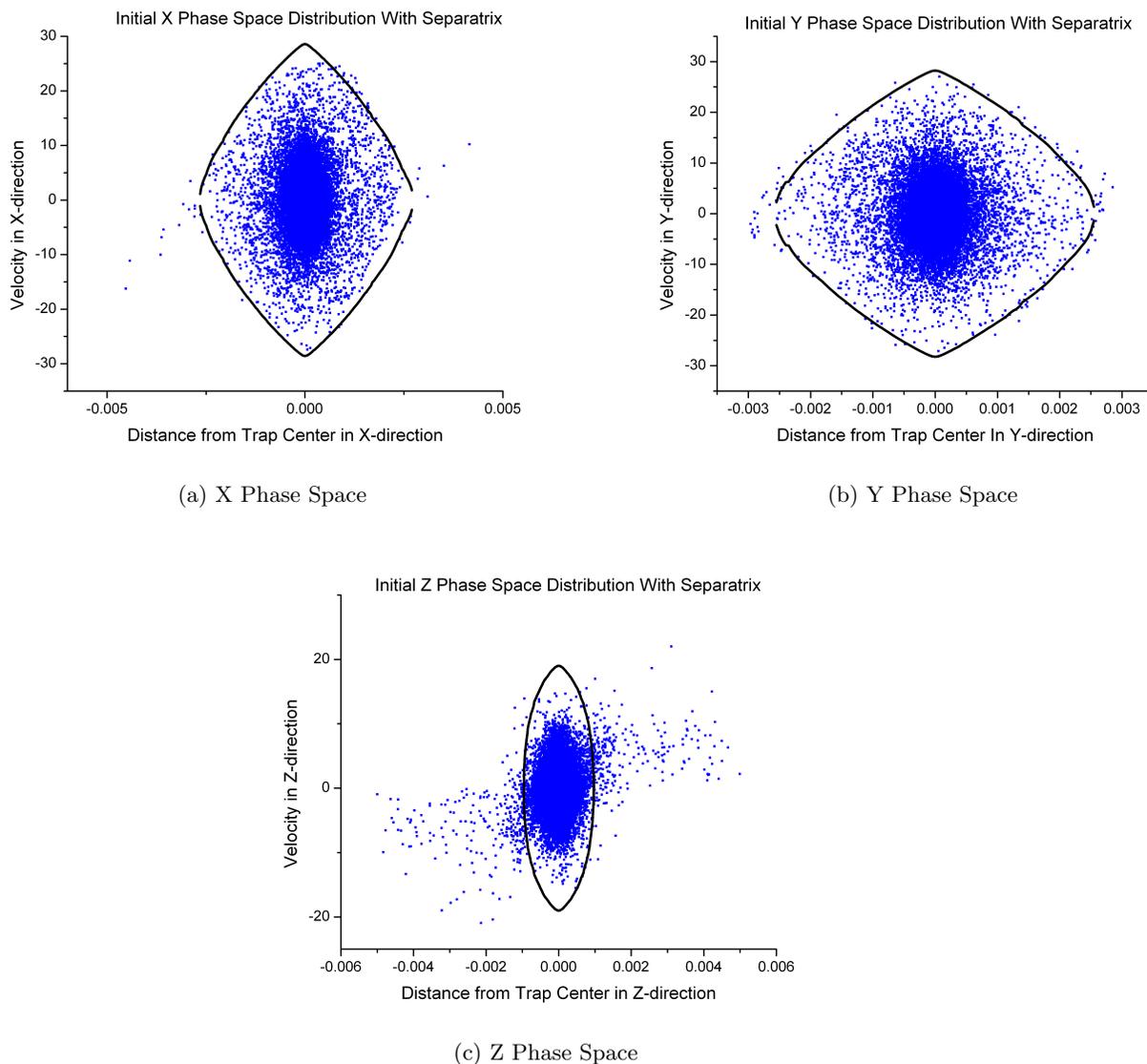


Figure 2.8: This figure shows the initial distribution of the molecules in the trap evolution simulation plotted with the separatrix in that dimension. Figure a and b show that while a few of the molecules were not trapped in the x and y directions, most of the molecules are well within the trap. 2.6c shows that more molecules are un-trapped in z but a majority were still within the separatrix. Because this is a conservative system, if a molecule begins in the separatrix, its phase space trajectory should stay within the separatrix.

gaining energy as shown by the blue line. However, when the synchronous molecule was lost, its velocity was still within the separatrix which would point to the possibility of another cause for the error.

Another idea is that because of the resolution of the simulation, the interpolation method

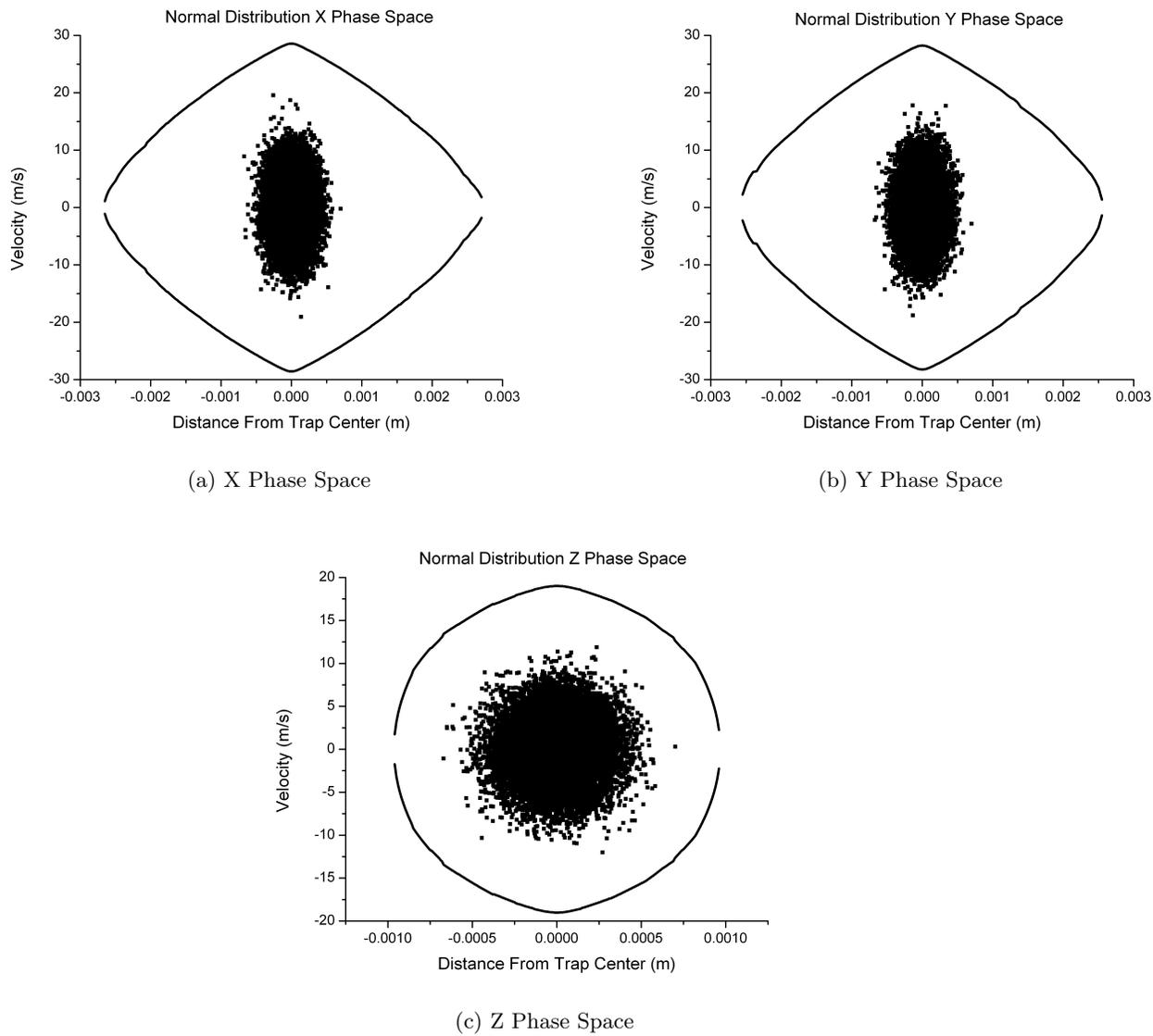


Figure 2.9: These graphs show the normal distribution of molecules within the trap. Clearly, this distribution was well within the separatrix and thus, should be well within the trap even if the separatrix was incorrect by a significant amount.

could be causing nonphysical accelerations to arise. While data was gathered for the rate of loss of molecules due to the Zone of Death, because the simulation is not trapping my molecules properly, that data is most likely tainted by the errors in the simulation and would provide an incorrect analysis of the system.

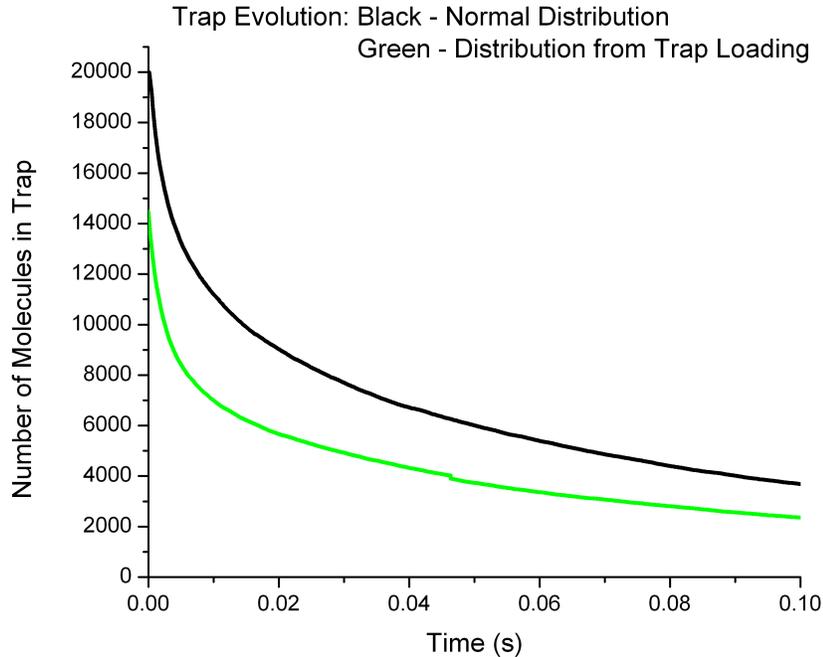


Figure 2.10: This shows the number of molecules remaining in the simulation over time for two different initial distributions. The black line shows the normal distribution and the green line shows the distribution saved from the trap loading simulation. The normal distribution lost 81.58% of the molecules while the loading sequence distribution lost 83.70% of the molecules.

2.6 Conclusions

The simulation was very successful in modeling the loading of Stark decelerated molecules into an electrostatic trap. It was able to trap around 10% of the cold ammonia which is on par with the efficiency of trap loading in the laboratory. It was also able to create a realistic phase space distribution of well-loaded molecules in an electrostatic trap.

Further work would first involve fixing the simulation so that it accurately models ammonia trapped in an electrostatic trap in addition to its accurate modeling of trap loading. At the moment, the simulation does not properly trap the ammonia, the most likely cause being the energy drift in the differential solver. Once the simulation correctly models the trap, it would be very interesting to apply a zone of death approximation to the simulation. Taking that data and comparing it to the analysis done in [1] showing that nonadiabatic transitions in electrostatically trapped ammonia

are a dominant cause of trap loss is the final goal of future work.

Magnitude of Velocity of Synchronous Molecule in Trap Loaded Distribution

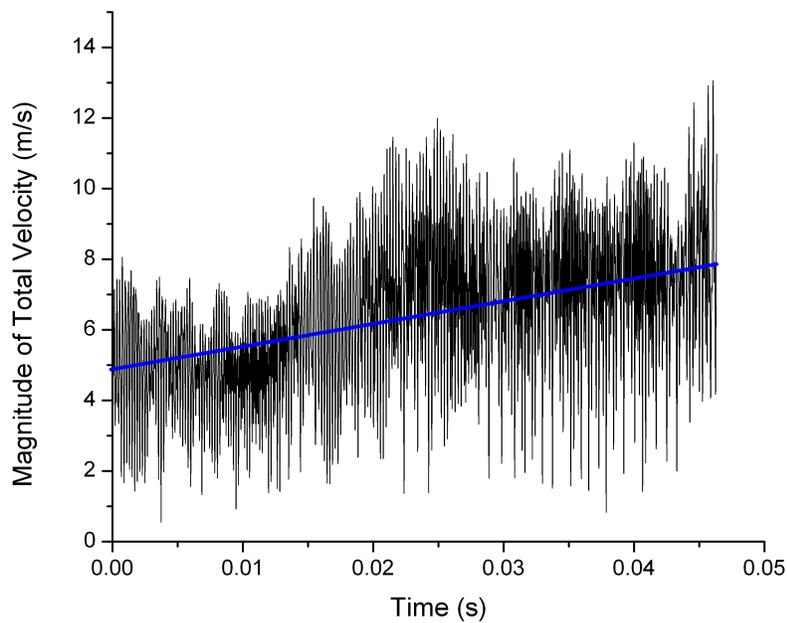


Figure 2.11: This figure shows the magnitude of the total velocity the synchronous molecule over time in the trap evolution simulation using the initial distribution taken from the trap loading simulation.

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