The Pseudopotential As a Tool for Describing Ion Crystal Morphology

by

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Abstract

Describing the structure of ion Coulomb crystals in periodically-driven systems, such as cylindrical and linear Paul traps, is important for applications in quantum information processing, quantum simulation, spectroscopy, and frequency standard determination. The pseudopotential, a time-independent effective potential obtained by averaging the explicitly time-dependent trapping potential, is often used for this specific purpose, among other uses in the general study of trapped ion dynamics. This thesis examines the strengths and weaknesses of the pseudopotential approximation as a tool for describing few-ion crystal configurations in a Paul trap. Numerical evidence for the failure of the standard pseudopotential commonly found in the literature in predicting crystal alignment effects in such systems is provided. A method for deriving an improved pseudopotential for a general set of coupled differential equations is presented and applied to the cylindrical and linear traps, providing analytical evidence for these “exotic” crystal alignment effects. The limitations of the improved pseudopotential in explaining crystal instability in certain regions of trap parameter space and in terms of scalability to many-ion systems are discussed.
Dedication

I dedicate this thesis to anyone interested to read it.
I would like to thank my research advisor Prof. Blümel for his invaluable support and mentorship, for providing the impetus for this project and giving it direction. I would not have been able to gather or give structure to the material in this thesis without his guidance. I would like to thank my colleague Dr. Y. S. Nam for his contributions to both the content and the structure of this thesis. I thank Max Silvester for the data used in many of the figures presented. Finally, I thank my research group members Daniel Weiss and Angus Kan for making the process fun.
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Chapter 1

Introduction

1.1 Crystals in External Fields

In 1878, the American physicist Alfred Mayer published a paper on the self-assembly of floating magnets in a superposed magnetic field. In his experiment he inserted magnetized needles into pieces of cork and placed them in a container of water, with their north poles facing upward. He then placed a stronger magnet above the center of the container, with its south-pole facing the surface of the water. This external field caused the corks to preferentially assemble into a number of different spatial patterns depending on their number, depicted in Fig. 1.1 – three needles form an equilateral triangle, four a square, five a pentagon, and so on. Mayer reasoned that these patterns arose from a competition between the attractive external field, which pulled the corks towards the center of the container, and the repulsive internal forces between the needles, which limited how close they could get to each other. Then, each pattern corresponded to a local energy minimum in the configuration space of the corks in a potential created by the central magnet together with the mutual interactions of the needles. We call these minima “local” since, for certain numbers of needles (for example, 5, 6, and 8), more than one stable configuration exists. A global minimum would correspond to the
Figure 1.1: Mayer’s illustrations of the patterns formed by different number of interacting needles as shown in [1]. A single stable equilibrium pattern was observed for 2-4 needles. Beyond 5 the needles begin to show multiple stable configurations. The lettering denotes the stability of these structures. For example, 8(c) is more stable than 8(b), which is more stable than 8(a).

Mayer’s experiment gives us an intuitive picture of a phenomenon ubiquitous in atomic, molecular, and optical physics (AMO) and condensed matter systems, namely the arrangement into “crystalline” spatial patterns of charged particles in an external electric or magnetic field. Systems that show similar behaviour include plasmas [2], thin liquid films [3], and other soft matter systems [4]. While Mayer used a magnetostatic field, these phenomena appear in time-periodic, dynamic fields as well. Periodically-driven systems abound in AMO physics and the ion traps discussed in this thesis represent but one class of such systems.

1.2 Crystals in Ion Traps

An ion trap [5, 6] is a device that uses electric or magnetic fields to confine charged particles in an isolated space for extended periods of time. A prime example, for which its inventor Wolfgang Paul won a Nobel prize, is the Paul trap. The application of
alternating (ac) and direct current (dc) voltages to three hyperbolic electrodes allows this trap \[5\,7\] to create static and dynamic electric fields capable of confining ions near its center. Ions can be stably trapped for a few hours \[8\] to a few days \[9\]. Up to \(10^5\) ions have been reported to be stored simultaneously \[8\].

A crystal usually refers to a form of solid matter, involving the placement of atoms in some regular lattice structure and the resulting delocalization of atomic electrons in this lattice, as described by quantum mechanics. Similarly-charged ions in ion traps also form crystals, called Coulomb crystals, though in a fundamentally different sense \[10\, 54\]. In a Paul trap, when ions are cooled, restorative electric fields direct them towards the trap’s center. Classical electromagnetic repulsion between the like-charged ions, however, limits the interparticle spacing (typically on the order of 10\(\mu\text{m}\) \[7\,11\,12\]), causing the ions to arrange into specific spatial patterns that minimize their energy \[10\]. Like Mayer’s needles, these patterns are determined by the number of ions and the competition between the trap and Coulomb forces.

Coulomb crystals in ion traps, particularly the Paul trap, are at the heart of many current research endeavours and technological applications, including the realization of a scalable quantum computer \[13\,16\,18\], quantum simulation \[22\,26\], structural phase transitions \[27\,33\], frequency standard determination \[34\], nonlinear spectroscopy \[35\], and artificial atoms \[36\,38\]. Simple, linear ion chains, shown in the topmost image in Fig. \[1.2\] have been the most common crystal configuration investigated for these purposes \[17\,39\,60\,62\]. However, by adjusting the ratio of the confining field strength in the axial and radial directions, a variety of other crystal morphologies can be formed. In Fig. \[1.2\] as we go down, the images show crystals in potentials with higher axial confinement: first we see linear chains, then crystals with a zigzag structure, others with a flatter, pancake-like structure, and a few with a 3D helical structure. Recently, interest in such complicated 2D and 3D structures has been growing \[19\,33\].

For many of the above-mentioned applications, it is necessary to know the exact lo-
Section 1.3. Pseudopotential: An Analytical Tool for Describing Crystal Morphology

Figure 1.2: 19-ion Coulomb crystals in a linear Paul trap as shown in Fig. 2 of [40]. Each image corresponds to a different setting of trap parameters. As the axial potential is strengthened, going down the images, the crystal progresses from a linear chain to a zigzag and then a helical structure.

cations of ions within a particular crystal configuration, as well as the set of trapping conditions that are needed to induce ions into such configurations. Entangling or encoding ion qubits using lasers in a trapped-ion quantum computer, for example, requires precise knowledge of their positions [39].

1.3 Pseudopotential: An Analytical Tool for Describing Crystal Morphology

The positions of ions in Coulomb crystals are practically constant over large time scales. As such, we are usually interested in the dynamical evolution of such ion systems at time scales that are long compared to the fast radio-frequency (rf) time scale of the ac voltage applied to the trap electrodes. It is convenient, then, to average over these fast time
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scales and construct an effective, time-independent potential, called a pseudopotential, that captures much of the information in the original, explicitly time-dependent equations of motion describing ion motion. The standard pseudopotential for a 1D system is constructed as per Ref. [41] following a method suggested by Kapitza. Conveniently, the pseudopotential of an $N$-ion system is simply the sum of the pseudopotentials of each ion and the mutual Coulomb potentials between the ions. Its simplicity and analyzability make the pseudopotential a successful tool for describing ion dynamics [42]. Positions of ions in a crystal, a necessary piece of information for many applications of such structures, can be found analytically, rather than by computationally-intensive, time-consuming numerical integration of the equations of motion.

It has been shown, however, that the standard pseudopotential fails to capture some basic, qualitatively-significant features of ion crystals [45–47]. In a two-ion cylindrical Paul trap, for example, it is incapable of accounting for the alignment of a specific ion-crystal configuration, which has been observed in numerical simulations [45]. This raises questions about the limitations of the pseudopotential that are worth exploring in detail, given its ubiquitous usage.

Broadly, this thesis seeks to examine the effectiveness of the pseudopotential in describing ion-crystal configurations in a class of periodically-driven systems. The thesis discusses few-ion systems in a cylindrical Paul trap as example cases, though its central results are generalizable to many other systems, including the linear Paul trap. The basic objectives of this thesis can be stated as follows: (i) to point out the limitations of the standard pseudopotential used in the literature, specifically its inability to account for “exotic” few-ion crystal configurations in the Paul trap, (ii) to propose a general procedure for deriving an improved pseudopotential that is capable of predicting the existence of such crystals, (iii) to evaluate the limitations of this improved pseudopotential in describing ion dynamics and in relation to its standard counterpart, and (iv) to suggest possible paths for overcoming these limitations.
1.4 Outline

The thesis is structured in the following way. In Chapter 2, we introduce the basic design and theory of the cylindrical Paul trap. We formally derive the equations of motion and the standard pseudopotential for \( N \) ions in a Paul trap. In Chapter 3, we discuss the main topics of the thesis: the failure of the standard pseudopotential in accounting for certain crystal morphologies in systems of 2, 3, and 4 ions, and the derivation of an improved pseudopotential that is capable of doing so. We formally define crystal morphologies, present numerical evidence for the existence of “exotic” crystals, and propose a general framework for deriving an improved pseudopotential for a system with 3 degrees of freedom. In Chapter 4, we apply our general framework to a two-ion linear Paul trap, as an example of another periodically-driven system to which our procedure applies. Contrary to published claims in the literature about their supposed nonexistence [58], we provide numerical evidence, corroborated by analytical predictions of the improved pseudopotential, for the existence of tilted two-ion crystals in the linear Paul trap. In Chapter 5, we discuss the limitations of the improved pseudopotential in accounting for crystal instability observed in numerical simulations of a two-ion cylindrical Paul trap for certain choices of trapping parameters. We develop a discretized model of the two-ion Paul trap to provide analytical evidence for the existence of these crystal-free regions. We also report the existence of a new kind of stability in the Paul trap, where relative motion of ions is stable and center-of-mass motion is unstable. In Chapter 6, we discuss the main results of this thesis, summarizing and comparing the limitations of the standard and improved pseudopotentials, and provide an outlook on possible future research directions.
Chapter 2

Cylindrical Paul Trap

In this chapter we review the basics of Paul-trap physics. In Section 2.1, we discuss the design of the cylindrical Paul trap in relation to its function. In Section 2.2, we derive the equations of motion that describe the trajectory of a single ion and in Section 2.3, define a space of solutions, the Mathieu stability region, for which the ion is stably confined. We then do the same for a system of $N$ ions in Section 2.4. Finally, in Section 2.5, we introduce the pseudopotential approximation, deriving the single- and $N$-ion standard pseudopotential, as per the method of Kapitza, outlined in [41].

2.1 Trapping Principles

A Paul trap cannot use static electric fields alone to trap ions. The purpose of an ion trap is to establish a potential minimum that will attract and, thereby, confine ions to a delimited region in 3D space [43]. In other words, the trap must exert a restoring force $\vec{F}$ that pushes the ions back towards some equilibrium position whenever they are displaced from it. The simplest restoring force capable of doing this is linear in all three
dimensions, with components

\[ F_u = -k_u u; \quad k_u > 0, \quad u = x, y, z. \]  \hspace{1cm} (2.1)

In a Paul trap, \( \vec{F} \) is generated using electric fields \[5, 6\]. Therefore, we can write

\[ \vec{F} = -Q \nabla \Phi, \]

where \( \Phi \) is the electric potential and \( Q \) is the charge of the ion. Since \( \vec{F} \)
is linear, this potential must be quadratic and can be written as

\[ \Phi = Ax^2 + By^2 + Cz^2, \]  \hspace{1cm} (2.2)

where, in the static case if \( \Phi \) is supposed to be a trapping potential, \( A, B, \) and \( C \) are positive constants.

We now prove that the potential \( \Phi \) must be dynamic. Suppose that \( \Phi \) is static. Taking the region of confinement to be charge-free, Laplace’s equation demands

\[ \nabla^2 \Phi = 0, \]  \hspace{1cm} (2.3)

from which it follows that

\[ A + B + C = 0. \]  \hspace{1cm} (2.4)

Clearly, Eq. (2.4) cannot be satisfied if \( A, B, C > 0 \), as required by Eq. (2.2), which demands \( A, B, C > 0 \) for stable trapping. This result is an illustration of Earnshaw’s theorem \[43\], which states that point charges cannot achieve stable equilibrium in three dimensions in a static electric field alone. The implication for the design of a Paul trap is clear: to achieve its purpose, a Paul trap must use a combination of static and dynamic electric fields.

The trap consists of three cylindrically-symmetric hyperbolic electrodes: one ring electrode capped on either end by an end-cap electrode, as in Fig. 2.1 \[43\]. The dynamic and static components of the trap potential are generated by applying ac and dc voltages respectively across these electrodes. In this thesis, the symmetry axis of the two end-cap electrodes is defined as the \( z \) axis. Cylindrical symmetry demands that the strengths of the trapping force in the \( x \) and \( y \) directions be of equal strength and thus,
it is convenient to define the radial displacement from the $z$ axis by a single variable $r$.
This also implies, by Eq. (2.4), that $A = B$ and thus, $C = -A - B = -2A$. Inserting
these conditions into (2.2), it is apparent that $\Phi$ must have the form

$$\Phi = A(x^2 + y^2 - 2z^2).$$  \hspace{1cm} (2.5)  

The exact form of the trap potential is determined by the spacing between the electrodes.
Any specific trap geometry can be uniquely defined by two constants \[43\]: $r_0$, the radius
of the ring electrode at its thinnest point, and $z_0$ the axial distance between the origin
of the trap and the tips of the end-cap electrodes. Including these terms and the explicit
time-dependence of the potential, we get

$$\Phi(x, y, z, t) = \frac{\Phi_0}{r_0^2 + 2z_0^2}(x^2 + y^2 - 2z^2),$$  \hspace{1cm} (2.6)  

where $\Phi_0$ is the voltage applied between the trap electrodes. In an ideal Paul trap, this
voltage is given by

$$\Phi_0(t) = U_0 + V_0 \cos \Omega t.$$  \hspace{1cm} (2.7)
where $U_0$ is the amplitude of the dc voltage applied across the trap electrodes, $V_0$ the amplitude of the ac voltage, and $\Omega$ the frequency of the sinusoidal ac drive.

Thus, during any one period of the ac voltage, the explicitly time-dependent component of the resulting trapping force will oscillate between being directed to and away from the origin of the trap. With suitable choices of $U_0$, $V_0$, and $\Omega$, however, it is possible for this force to be restorative when averaged over several periods of the driving field [43].

### 2.2 Equations of Motion for a Single Ion

Strictly speaking, systems of charged particles confined in a Paul trap fall under the purview of quantum mechanics. Indeed, many important applications of the Paul trap, including quantum computing, quantum simulation, frequency standard determination, and spectroscopy, exploit the quantum mechanical properties of these systems.

For our purposes, however, a classical treatment of these systems is sufficient [7, 11, 12]. The justification is twofold: (i) in a typical Paul trap, the distances between ions (on the order of $\mu$m) are large compared to the quantum length scale, and (ii) in the absence of damping, ions usually have relatively large temperatures, i.e., we are operating in the limit of large quantum numbers. Furthermore, the central result we derive using this classical view – namely, an improved pseudopotential – can easily be translated into a quantum-mechanical picture by substituting dynamical variables with corresponding operators.

With this in mind, using the trap potential described in Eqs. (2.6) and (2.7), we can immediately write down the equation of motion for a single ion in a Paul trap. The force on the ion is given by

$$\vec{F} = -Q \nabla \Phi(x, y, z, t), \quad (2.8)$$

where $Q$ is the charge of the particle and $\Phi(x, y, z, t)$ is the potential given by (2.6).
According to Newton’s equation, this leads to

\[ m \ddot{\vec{R}} + 2Q \frac{U_0 + V_0 \cos \Omega t}{r_0^2 + 2z_0^2} \begin{bmatrix} x \\ y \\ -2z \end{bmatrix} = 0, \] (2.9)

where \( m \) is the mass and \( \vec{R} \) is the vector position of the ion. To keep the results in this thesis independent of specific trap dimensions, it is convenient to re-express this equation in dimensionless units. By measuring time in units of

\[ t_0 = \frac{2}{\Omega}, \] (2.10)

and by defining dimensionless trapping parameters

\[ a = \frac{8QU_0}{m\Omega^2(r_0^2 + 2z_0^2)}, \quad q = \frac{4QV_0}{m\Omega^2(r_0^2 + 2z_0^2)}, \] (2.11)

corresponding to the amplitudes of the static dc and driving ac voltages respectively, Eq. (2.9) can be expressed as

\[ \frac{d^2}{d\tau^2} \begin{bmatrix} x \\ y \\ z \end{bmatrix} + (a + 2q \cos 2\tau) \begin{bmatrix} x \\ y \\ -2z \end{bmatrix} = 0. \] (2.12)

### 2.3 Stable Confinement Region for a Single Ion

Each of the three equations in (2.12) is an example of the second-order differential equation known as the Mathieu equation \([44]\). The solutions to this equation are well-known and admit of two general classes: (i) \textit{bounded}, where the particle’s position with respect to a defined origin (in a Paul trap, the center of the trap) is always smaller than some arbitrary distance as time goes to infinity, and (ii) \textit{unbounded}, where the particle’s position increases without bound as time goes to infinity \([44]\). Whether a solution is
Section 2.3. Stable Confinement Region for a Single Ion

Figure 2.2: Radial and axial stability regions in single-ion trap parameter space. The brown tongues delineate the region of solutions that are stable in the radial direction, while the green tongues delineate the region of solutions that are stable in the axial direction.

bounded or not depends on the values of the parameters $a$ and $q$ and thus, for a Paul trap, the values of the applied dc and ac voltages respectively [44].

For a particular choice of parameters $(q,a)$, a single ion is stably confined in all three dimensions only if all three equations of motion (2.12) simultaneously admit bounded solutions. Figure 2.2 delineates the regions in a two-dimensional space defined by parameters $a$ and $q$, where bounded solutions to the $x$, $y$, and $z$ equations exist [44]. The area within the brown tongues corresponds to the region of bounded solutions in the radial $x$ and $y$ directions, while the area within the green tongues corresponds to the region of bounded solutions in the axial $z$ direction. Thus, an ion is stably confined only for choices $(q,a)$ that lie in the intersection of these two areas. Though higher-order stability zones do exist, we confine our investigation to the lowest-order region, shown in Fig. 2.3 which we refer to henceforth as the Mathieu stable region.
Section 2.4. Equations of Motion for $N$ ions

The equations of motion describing a system of $N$ ions with the same charge and mass can easily be derived from the single-ion equation. In addition to (2.10) and (2.11), we measure distances in units of

$$l_0 = \left( \frac{Q^2}{\pi \epsilon_0 m \Omega^2} \right)^{1/3}, \quad (2.13)$$

where $Q$ is the charge of the stored particles, $m$ is their mass, and $\epsilon_0$ is the permittivity of the vacuum. Then, the $3N$ equations of motion for each of the $3$ Cartesian coordinates of each of the $N$ ions are given by

$$\ddot{\vec{R}}_i + [a + 2q \cos(2\tau)] \begin{pmatrix} \dot{X}_i \\ \dot{Y}_i \\ -2\dot{Z}_i \end{pmatrix} = \sum_{j=1}^{N} \frac{\vec{R}_i - \vec{R}_j}{|\vec{R}_i - \vec{R}_j|^3}, \quad i = 1, \ldots, N, \quad (2.14)$$

where $\vec{R}_i = (X_i, Y_i, Z_i)$ is the position vector of ion $i$ in units of $l_0$. 

Figure 2.3: Lowest-order Mathieu stability region. The intersection of the areas contained by the brown and green lines, denoted by $r$-stable, $z$-stable is the first region of stable confinement for a single ion. Outside this region, an ion may be stably confined in one direction and not the other, or unstable in both directions.

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The stability region in \((q, a)\) parameter space for \(N\) ions turns out to be the same as the single-ion stability region. This can be understood in the following way. The position of the center-of-mass \(\vec{R}_{CM}\) of \(N\) ions is given by

\[
\vec{R}_{CM} = \frac{1}{2}(\vec{R}_1 + \vec{R}_2 + \ldots + \vec{R}_N),
\]

and therefore,

\[
\ddot{\vec{R}}_{CM} = \frac{1}{2}(\ddot{\vec{R}}_1 + \ddot{\vec{R}}_2 + \ldots + \ddot{\vec{R}}_N).
\]

The equation of motion for the center-of-mass is found by summing the \(N\) vector equations, each describing an individual ion’s motion. Upon summation, we find

\[
\ddot{\vec{R}}_{CM} + \left[ a + 2q \cos(2\tau) \right] \begin{pmatrix} X_{CM} \\ Y_{CM} \\ -2Z_{CM} \end{pmatrix} = 0,
\]

where \(X_{CM}, Y_{CM},\) and \(Z_{CM}\) denote the position of the center of mass in \(x, y,\) and \(z\) directions respectively. Since the Coulomb forces between any two particles are equal and opposite and cancel each other out, the RHS of (2.17) reads 0. Thus, the center-of-mass equation (2.17) is identical to the single-ion equation of motion (2.12). For all \(N\) ions to be stably confined, their center-of-mass must also be stably confined. Thus, we can conclude that the \(N\)-ion system is stable when (2.17) admits bounded solutions and that the \(N\)-ion system shares the single-ion stability region delineated in Fig. 2.3.

### 2.5 Standard Pseudopotential

The nonlinear Coulomb term together with the time-dependence of the trap force in (2.14) make the analysis of the dynamics of multiple-ion systems difficult. It is this difficulty that prompts us to construct an approximate time-independent potential, known as a pseudopotential, that, while capturing much of the information in the original time-dependent potential, is simpler to analyze. Ion motion in this fictitious potential
mimics ion motion in the original explicitly time-dependent potential when considered in the limit of long time scales.

In constructing the pseudopotential, we make two important assumptions: (a) that ion motion can be separated into motion at long and short time scales, and (b) that the driving force in (2.14) is periodic. The motion of an ion can always be split into two components: one, a slow-varying macromotion and the other, a fast-varying micromotion, superimposed on the macromotion. Macromotion corresponds to large-scale, secular motion at a frequency much lower than the frequency Ω of the ac voltage. Micromotion is smaller in magnitude and occurs at a relatively higher frequency, on the order of Ω. This notion is visualized in Fig. 2.4. The purple line depicts the actual trajectory of an ion. The green line, representing the macromotion, smoothly interpolates this trajectory at regular intervals. The micromotion is responsible for the deviation of the purple and green lines within any such interval. When we are interested in the long-term dynamics of an ion, the micromotion within each interval provides too much information – the overall motion is well-approximated by the secular part.

We now proceed to derive the pseudopotential as prescribed by Kapitza in [41], first for a particle in a general 1D periodic potential and then, a particle in a Paul trap.

### 2.5.1 General 1D Periodic Potential

We are interested in systems described by equations of motion of the form

\[
m\ddot{x} = -\frac{dU}{dx} + f(x)\cos\omega t,
\]

(2.18)

where \( m \) is the mass of the particle, \( U \) is some static potential, \( f(x) \) is some spatially-dependent function, and \( \omega \) is the frequency of the periodic force. The derivation begins by splitting ion motion \( x \) into a macromotion \( X \) and micromotion \( \xi \), i.e.,

\[
x(\tau) = X(\tau) + \xi(\tau).
\]

(2.19)
Substituting (2.19) into (2.18) and expanding the two terms on the RHS of (2.18) up to first order in $\xi$ around the secular coordinate $X$, we get

$$m\ddot{X} + m\ddot{\xi} = -\frac{dU}{dX} - \xi\frac{d^2U}{dX^2} + F(X,t) + \xi\frac{\partial F}{\partial X},$$

(2.20)

where $F(X,t) = f(X)\cos\omega t$. Now, assuming that $X$ changes negligibly over the course of one period of the driving force, we have that

$$m\ddot{X} + \frac{dU}{dX} = -m\ddot{\xi} - \xi\frac{d^2U}{dX^2} + F(X,t) + \xi\frac{\partial F}{\partial X} \approx 0,$$

(2.21)

where the last approximation is based on the assumption that $X$ is a good approximate solution of the unperturbed equation. Assuming that the micromotion amplitude $\xi$ is small, the term in the middle becomes

$$-m\ddot{\xi} + F(X,t) = 0,$$

(2.22)

which can be solved to yield

$$\xi = -\frac{F(X,t)}{m\omega^2}.$$  

(2.23)
We now resubstitute the micromotion amplitude $\xi$ into (2.20) to produce

$$m\ddot{X} = -m\ddot{\xi} + F(X, t) - \frac{dU}{dX} + \frac{F(X, t)}{m\omega^2} \frac{d^2U}{dX^2} - \frac{F(X, t)}{m\omega^2} \frac{\partial F}{\partial X}. \quad (2.24)$$

Finally, we take the time-average of (2.24) over one period of the driving term. We define an averaging operation given by

$$\overline{G(t)} = \frac{1}{T} \int_{t}^{t+T} G(t) dt. \quad (2.25)$$

We assume that the average over one driving period of the force $F(X, t)$ and the micromotion $\xi(t)$ is 0, i.e. $\overline{F(X, t)} = 0$ and $\overline{\xi(t)} = 0$. This implies that $\overline{\ddot{\xi}} = 0$ as well. Thus, the first two terms on the RHS of (2.24) vanish. $X$ is assumed to vary slowly and hence, can be assumed constant over one trapping cycle. Applying the averaging operation to the remaining terms in (2.24), we get

$$\overline{\frac{dU}{dX}} = \frac{dU}{dX}, \quad (2.26)$$

$$\overline{\frac{F(X, t)}{m\omega^2} \frac{d^2U}{dX^2}} = 0, \quad (2.27)$$

and

$$\overline{\frac{F(X, t)}{m\omega^2} \frac{\partial F}{\partial X}} = \frac{\partial}{\partial X} \frac{F^2}{2m\omega^2} = \frac{d}{dX} \frac{f(x)^2}{4m\omega^2}. \quad (2.28)$$

The equation (2.24) can now be re-expressed in the form

$$m\ddot{X} = -\frac{dU_{\text{eff}}}{dX}, \quad (2.29)$$

where

$$U_{\text{eff}} = U + \frac{f(x)^2}{4m\omega^2} \quad (2.30)$$

is the time-independent pseudopotential.

### 2.5.2 Single-Ion Pseudopotential

We now apply this general framework to the specific case of a single ion in a Paul trap. This is done by direct comparison of the general equation of motion (2.18) to each of
the equations of motion for a single ion (2.12). Since the motion of the ion in the three spatial dimensions is decoupled, we separately compare each component of the vector equation (2.12) to the general equation (2.18). With $m = 1$, $\omega = 2$, $f_x = 2qx$, $f_y = 2qy$, $f_z = -4qz$, and $U(x, y, z) = \frac{1}{2}a(x^2 + y^2) - az^2$, it can immediately be found that

$$U_{\text{eff}} = \frac{1}{2}[a + \frac{1}{2}q^2](x^2 + y^2) + [q^2 - a]z^2. \quad (2.31)$$

Comparing (2.31) to the functional form of a harmonic oscillator, we find

$$U_{\text{eff}} = \frac{1}{2}\omega_x^2 x^2 + \frac{1}{2}\omega_y^2 y^2 + \frac{1}{2}\omega_z^2 z^2, \quad (2.32)$$

where $\omega_x = \omega_y = \sqrt{a + \frac{1}{2}q^2}$ and $\omega_z = \sqrt{2(-a + q^2)}$. Physically, these three terms give the frequency of the ion’s secular motion in each of the three spatial directions.

### 2.5.3 $N$-Ion Pseudopotential

The procedure for deriving a single-ion pseudopotential can easily be extended to a system of $N$ ions, described by $3N$ coupled equations of motion, each of the form

$$m_i \ddot{x}_i = -\frac{\partial U(x_1, x_2, ..., x_N)}{\partial x_i} + f_i(x_i) \cos \omega t, \quad (2.33)$$

where $m_i$ is the effective mass of each particle or coordinate, $U$ is a static potential whose derivatives appear in all $N$ equations, and $f_i$ is the spatially-dependent amplitude of the periodic force in the $i$th equation of motion. Unlike the single-ion case, the potential $U$ is now a combination of the dc component of the trapping potential and the mutual Coulomb potential between ions, i.e. $U = U_{\text{dc \, trap}} + U_{\text{Coulomb}}$. The pseudopotential for $N$ ions is then given by the sum of the pseudopotentials for each of the $N$ ions and their mutual Coulomb interactions

$$U_{\text{eff}} = U_{\text{dc \, trap}} + \sum_{i=1}^{N} \frac{f_i^2}{4m_i\omega^2} + U_{\text{Coulomb}}. \quad (2.34)$$
Chapter 3

Exotic Ion Crystals

We now proceed to the central topics of this thesis: the failure of the standard pseudopotential in accounting for the existence of “exotic” crystal morphologies in the Paul trap and the derivation of an improved pseudopotential. Much of the literature on ion crystals in the Paul trap has focussed on three kinds of crystal configurations [19–33]: (a) linear chains, where the particles are aligned along the $z$-axis of the trap, (b) zigzagged crystals, where consecutive ions in the linear chain pop out in opposite directions in the $rz$-plane, and (c) planar crystals, where the ions lie in the $xy$-plane. However, the wealth of crystal configurations that trapped ions are capable of forming is not limited to these three types. In systems of 2-6 ions, we already find evidence for the existence of these nonintuitive “exotic” crystal configurations. A natural question then is: can the existence of these configurations be accounted for analytically? This chapter attempts to give an answer.

In this chapter, we (i) formally define the concept of a crystal in a Paul trap, (ii) outline a computational procedure for finding crystals using the $N$-ion equations of motion (2.14), (iii) provide numerical evidence for the existence of “exotic” crystal morphologies in systems of 2, 3, and 4 ions, (iv) show how the pseudopotential is used to delineate crystal morphological boundaries in trap parameter space, (v) demonstrate
the failure of the standard pseudopotential in accounting for the existence of exotic crystal morphologies, (vi) derive an improved version of the pseudopotential for a 3-ion system, and (vii) provide predictions for “exotic” morphologies using this improved pseudopotential.

3.1 Definitions

The modified Mathieu differential equations (2.14) that govern the dynamics of systems of ions in the Paul trap admit a variety of solutions, each corresponding to a distinct physical structure [51]. These structures may be differentiated in terms of their: (i) periodicity in time, (ii) geometry, and (iii) stability [45, 46, 51].

The most common $N$-ion structure is a cloud, where the motion of individual ions is chaotic [51]. These structures are disordered and aperiodic, as is revealed by the Fourier amplitudes in the frequency spectrum of an individual ion’s motion. The stability of these clouds is determined by the competition between the damping effect of laser cooling and the effect of rf heating, wherein energy from the rf field is pumped into interparticle interaction, causing the cloud to become more energetic and expand [51]. If the cooling and heating effects are balanced, the cloud stabilizes, in the sense that it can be held in its state for hours. Alternatively, if the cooling effect exceeds the rf heating rate, the cloud may cool and transition into a lower energy configuration, corresponding to an attractor [51].

A period-$n$ attractor is a structure that returns to the same point in phase space after every $n$ trap cycles, i.e. the $N$ ions will have the same positions and velocities as they did $n$ cycles earlier. An ion crystal is defined simply as a period-1 attractor. Though ions in a crystal are constantly in motion, when viewed stroboscopically (for example, at the end of every trap cycle), their positions and velocities are constant. Henceforth, when we refer to the positions of ions in the crystal state, we mean the equilibrium
positions of the ions at the end of every trap cycle.

These definitions lead to two theoretical implications: if left unperturbed, (a) crystals do not experience rf heating, since their energy is stroboscopically constant \[7, 50\], and (b) crystals will remain in their respective state forever. Experimentally, however, crystals are constantly perturbed by background noise, which, if not cooled, limits their lifetimes.

Ion crystals themselves may be differentiated according to the spatial configuration of their constituting ions, i.e. by their morphology. The lowest-energy equilibrium configuration of a system of ions is determined by the ratios of the axial and radial confining forces. A weaker axial confinement will likely produce linear ion chains, while weak radial confinement will likely produce planar crystals. Thus, as we shall see, different morphologies occupy different regions in the Mathieu stability region.

3.2 Numerical Simulations of 2, 3, and 4 Ions

3.2.1 Procedure

Finding \(N\)-ion crystals in the Mathieu stability region shown in Fig. 2.2 is equivalent to finding stable fixed points for a mapping defined by integrating the set of \(3N\) equations (2.14) over one period of the driving trap force. Even for the simplest case of two ions stored simultaneously in the trap, this is extremely difficult, if not impossible, to do analytically \[10\].

Experimentally, crystals are usually created by laser-cooling systems of ions in a cloud state, thereby inducing a cloud \(\rightarrow\) crystal transition \[7\]. This procedure can be simulated computationally by using a 5th-order Runge-Kutta integrator \[52\] to propagate a damped version of (2.14) forward in time. By introducing a “friction” term \(\gamma \dot{\vec{R}}_i\), where \(\gamma\) is the strength of the (laser-) cooling, we can write a modified set of equations
describing the laser-cooled system

\[
\ddot{\vec{R}}_i + \gamma \dot{\vec{R}}_i + [a + 2q \cos(2\tau)] \begin{pmatrix} X_i \\ Y_i \\ -2Z_i \end{pmatrix} = \sum_{j=1 \atop j \neq i}^{N} \frac{\vec{R}_i - \vec{R}_j}{|\vec{R}_i - \vec{R}_j|^3}, \quad i = 1, \ldots, N. \tag{3.1}
\]

The undamped version of the \( N \)-ion system is Hamiltonian. Thus, unless initialized in such a state, the undamped system will not spontaneously transition into a crystal state. In the literature, the damping parameter \( \gamma \) is often used to simulate the effects of laser-cooling on ion dynamics as seen in experiments. Here, we should be clear that it is used purely as a tool to find crystals by inducing a cloud \( \rightarrow \) crystal transition \([7, 49]\). Once this transition has been induced, \( \gamma \) is adiabatically switched off (i.e. finally, \( \gamma = 0 \)) over a suitable number of trap cycles, so that the particles remain crystallized. If the particles remain at the fixed point under small perturbations, the crystal is said to be stable.

A few further notes on the above procedure are warranted, specifically with regard to:

(i) the initialization of the ion system, (ii) the time-length of a typical simulation, and (iii) the criterion used to determine the stability of a crystal.

(i) Ion dynamics are sensitive to initial conditions. Depending on the choice of initial ion positions, velocities, and the damping parameter \( \gamma \), ions may form a crystal, attractor, or remain in a cloud state. To minimize the dependency of results on initial conditions, for every choice of \( a \) and \( q \), multiple simulations were performed using different initial conditions. These were chosen using a standard random-number generator. Seeds were recorded for replicability of results.

(ii) Typically, the simulations are allowed to run until the ions crystallize, subject to some predetermined upper bound to the total time. \( \gamma \) is increased adiabatically over the course of the simulation to increase the chances of a cloud \( \rightarrow \) crystal transition. If the ions do not crystallize even with this increased damping, it is concluded that a crystal
does not form at that point in parameter space.

(iii) The stability of a crystal is determined by slightly perturbing the ions once they have crystallized. This is done by introducing noise into the system. If the crystal persists, it is stable.

### 3.2.2 Results

Performing the procedure outlined above, we identify several distinct crystal morphologies in the stability regions of 2, 3, and 4 ions. Here we discuss each one in detail.

For two ions, we find three possible morphologies: (i) rod crystals, where both ions are aligned along the $z$-axis with coordinates $(0, 0, z)$ and $(0, 0, -z)$, (ii) tilted crystals, where the ions have coordinates $(x, y, z)$ and $(-x, -y, -z)$, (iii) planar crystals, where both ions lie in the $xy$-plane, with coordinates $(x, y, 0)$ and $(-x, -y, 0)$. These three morphologies are shown in Figs. 3.1 (a), (b), and (c) respectively. Rod crystals are found when the axial confining force is relatively weaker than the radial confining force. When the axial confinement is increased, the two-ion rod is forced to tilt with respect to the $z$-axis. The two ions tilt further with increasing axial confinement until they come to lie in the $xy$-plane as a planar crystal.

For three ions, we find four morphologies: (i) rod crystals, where the three ions are aligned along the $z$-axis, with coordinates $(0, 0, z)$, $(0, 0, 0)$, and $(0, 0, -z)$, (ii) popout crystals, where the central ion in the rod configuration pops out into the $rz$-plane and the ions above and below pop out in the opposite direction, with coordinates $(x, 0, z)$, $(2x, 0, 0)$, and $(x, 0, -z)$ (iii) tilted crystals, where the popout crystal begins to tilt away with respect to the $z$-axis into the $xy$-plane, with coordinates $(x, y, z)$, $(2x, 0, 0)$, and $(x, -y, -z)$ and (iv) planar crystals, where the three ions form an equilateral triangle in the $xy$-plane. These four morphologies are show in Figs. 3.2 (a), (b), (c), and (d) respectively. When axial confinement is increased for a crystal in the rod configuration, the central ion is forced to pop out into the $rz$-plane. Increasing axial confinement
Figure 3.1: Two-ion crystals. Images (a), (b), and (c) depict two-ion rod, tilted, and planar crystals respectively. In the rod morphology, the coordinates of the two ions are \((0,0,z)\) and \((0,0,-z)\). In the tilted morphology, \((x,y,z)\) and \((-x,-y,-z)\). In the planar morphology, \((x,y,0)\) and \((-x,-y,0)\).
Figure 3.2: Three-ion crystals. Images (a), (b), (c), and (d) portray three-ion rod, popout, tilted, and planar crystals respectively. The center of mass of all configurations is always at the origin of the trap.

Further, the ions at the two ends begin to move towards the xy-plane, making it appear as if the popout structure is tilting. Finally, the tilted structure tilts into the xy-plane and forms a planar structure.

For four ions, we find seven morphologies: (i) rod crystals, where the four ions are aligned along the z-axis, with coordinates (0, 0, z), (0, 0, w), (0, 0, −z), and (0, 0, −w), (ii) zigzag crystals, where consecutive ions pop out in opposite directions into the rz-plane, with coordinates (x, 0, z), (v, 0, w), (−x, 0, −z), and (−v, 0, −w), (iii) diamond-shaped crystals, where two of the ions lie on the same axis in the xy-plane and the other two are aligned with the z-axis, with coordinates (0, 0, z), (x, 0, 0), (0, 0, −z), and (−x, 0, 0), (iv) popped-out diamond crystals, where the two sets of ions identified in (iii) are displaced in opposite directions in the radial direction, with coordinates (0, y, z), (x, −y, 0), (0, y, −z), and (−x, −y, 0), (v) tilted crystals, where (iv) begins to tilt with respect to the z-axis, with coordinates (x, y, z), (−x, y, −z), (v, −y, w), and (−v, −y, −w), (vi) double-rod crystals, where the ions form two pairs, whose projections onto the xy-plane are perpendicular to each other, and with the first set lying above the xy-plane, while the second lying below it, with coordinates (x, y, z), (−x, y, z), (−x, −y, z), and (x, −y, −z), (vii) square crystals, where the particles form a square in the xy-plane, with coordinates (x, y, 0), (x, −y, 0), (−x, y, 0), and (−x, −y, 0). These seven morphologies are show in
Figs. 3.3 (a), (b), (c), (d), (e), (f), and (g) respectively.

3.3 Failure of the Standard Pseudopotential

Having identified the possible morphologies that 2, 3, and 4 ions can form, we will now show how the pseudopotential, as constructed in Chapter 2, can be used to analytically predict where these morphologies can be found in the stability region of the trap. The key insight here is that the stroboscopic positions of the ions in each of these crystal configurations correspond to minima in the pseudopotential. To find these configurations, then, we simply need to solve the equations \( \partial U_{\text{eff}}/\partial x_i = 0 \), where \( U_{\text{eff}} \) is the pseudopotential, for the \( i \) coordinates that describe our system.

3.3.1 Standard Pseudopotential for Three Ions

A configuration of \( N \) ions is described by the \( 3N \) Cartesian coordinates denoting the positions of individual ions in the trap. The dynamics of this system are thus described by \( 3N \) coupled equations of motion. While the standard pseudopotential can be computed using all \( 3N \) coordinates, it is convenient to choose a simpler set of equations that describe the system in question. Since crystals are highly ordered, we can exploit the geometric relations between different ion coordinates to reduce the number of equations needed to describe the system.

In a three-ion crystal, which will serve as an example system in this section, knowing the coordinates of one ion, say, the topmost ion in Fig. 3.3 (c), allows us to know the positions of the other two ions. Let us denote the coordinates of the topmost ion by \( (X, Y, Z) \). Since the center-of-mass of the three ions must be located at the trap origin, we can infer that the bottommost ion has coordinates \( (X, -Y, -Z) \) while the center ion has coordinates \( (-2X, 0, 0) \).

To derive the 3-ion pseudopotential, we first need the equations of motion that govern
Figure 3.3: Four-ion crystals. Images (a), (b), (c), (d), (e), (f), and (g) show rod, zigzag, diamond-shaped, popped-out diamond, tilted, double rod, and square crystals respectively.
this system, in terms of our three generalized coordinates. The Lagrangian of the system is

$$L = \left[ T^{(1)} + T^{(2)} + T^{(3)} \right] - \left[ U_{\text{Trap}}^{(1)}(1) + U_{\text{Trap}}^{(2)}(2) + U_{\text{Trap}}^{(3)}(3) \right] - \left[ U_{\text{Coul}}^{(1)-(2)} + U_{\text{Coul}}^{(2)-(3)} + U_{\text{Coul}}^{(3)-(1)} \right].$$

(3.2)

For the positions \((X_i, Y_i, Z_i)\) and velocities \((\dot{X}_i, \dot{Y}_i, \dot{Z}_i)\) of the \(i\)th particle, in suitable dimensionless units [55], the kinetic energy is given by

$$T^{(i)} = \frac{1}{2}(\dot{X}_i^2 + \dot{Y}_i^2 + \dot{Z}_i^2),$$

(3.3)

the trap potential by

$$U_{\text{Trap}}^{(i)}(X_i, Y_i, Z_i) = \frac{1}{2}[a + 2q \cos(2\tau)](X_i^2 + Y_i^2 - 2Z_i^2),$$

(3.4)

and the Coulomb potential between particles \(i\) and \(j\) by

$$U_{\text{Coul}}^{(i)-(j)} = \frac{1}{\sqrt{(X_i - X_j)^2 + (Y_i - Y_j)^2 + (Z_i - Z_j)^2}}.$$  

(3.5)

We can now insert (3.3), (3.4), and (3.5) in (3.2) and use the Euler-Lagrange equations [56] to obtain 3\(N\) equations of motion. By writing \((X_i, Y_i, Z_i)\) in terms of \((X, Y, Z)\) as defined earlier, these 3\(N\) equations can be reduced to a set of three equations of motion given by

$$6\ddot{X} = -6[a + 2q \cos(2\tau)] + \frac{18X}{(9X^2 + Y^2 + Z^2)^{3/2}},$$

$$2\ddot{Y} = -2Y[a + 2q \cos(2\tau)] + \frac{Y}{2(Y^2 + Z^2)^{3/2}} + \frac{2Y}{(9X^2 + Y^2 + Z^2)^{3/2}},$$

$$2\ddot{Z} = +4Z[a + 2q \cos(2\tau)] + \frac{Z}{2(Y^2 + Z^2)^{3/2}} + \frac{2Z}{(9X^2 + Y^2 + Z^2)^{3/2}}.$$  

(3.6)

We can now immediately find the standard pseudopotential by comparing these equations with (2.33). Here \(m_x = 6, m_y = 2, m_z = 2, \omega = 2, f_x = -12qx, f_y = -4qy,\) and \(f_z = 8qz\) and

$$U(X, Y, Z) = \frac{2}{(9X^2 + Y^2 + Z^2)^{1/2}} + \frac{1}{2(Y^2 + Z^2)^{1/2}} + a\left[3X^2 + Y^2 - 2Z^2\right].$$

(3.7)
Inserting these into (2.34) and substituting in macromotion coordinates, we obtain the three-ion pseudopotential

\[ U^{(s)}_{\text{eff}}(x, y, z) = \frac{2}{\rho} + \frac{1}{2} \frac{\mu_x^2 x^2}{\rho^3} + \frac{1}{2} \frac{\mu_y^2 y^2}{r^3} + \frac{1}{2} \frac{\mu_z^2 z^2}{r^3}, \]  

(3.8)

where \( \rho = (9x^2 + y^2 + z^2)^{1/2} \), \( r = (y^2 + z^2)^{1/2} \), \( \mu_x = [6(a + q^2/2)]^{1/2} \), \( \mu_y = [2(a + q^2/2)]^{1/2} \), and \( \mu_z = [4(q^2 - a)]^{1/2} \).

### 3.3.2 Analytical Predictions of the Standard Pseudopotential

Since crystal configurations correspond to minima in the pseudopotential, by calculating when different configurations become unstable as a function of \( a \) and \( q \), we can identify where different morphologies can be found in the trap parameter space. A certain crystal configuration will become unstable when the first and second derivatives of the pseudopotential are zero. Thus, to predict the boundaries between different crystal regions, we first need to compute the first and second derivatives of the pseudopotential.

The first derivatives are:

\[ \frac{\partial U^{(s)}_{\text{eff}}}{\partial x} = \left( -\frac{18}{\rho^3} + \mu_x^2 \right) x = 0, \]
\[ \frac{\partial U^{(s)}_{\text{eff}}}{\partial y} = \left( -\frac{2}{\rho^3} - \frac{1}{2r^3} + \mu_y^2 \right) y = 0, \]
\[ \frac{\partial U^{(s)}_{\text{eff}}}{\partial z} = \left( -\frac{2}{\rho^3} - \frac{1}{2r^3} + \mu_z^2 \right) z = 0. \]  

(3.9)
The second derivatives are:

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial x^2} = \frac{486x^2}{\rho^5} - \frac{18}{\rho^3} + \mu_x^2,
\]

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial y^2} = \frac{6y^2}{\rho^5} - \frac{2}{\rho^3} + \frac{3y^2}{2r^5} - \frac{1}{2r^3} + \mu_y^2,
\]

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial z^2} = \frac{6z^2}{\rho^5} - \frac{2}{\rho^3} + \frac{3z^2}{2r^5} - \frac{1}{2r^3} + \mu_z^2,
\]

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial x \partial y} = \frac{54xy}{\rho^5},
\]

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial y \partial z} = \frac{6yz}{\rho^5} + \frac{3yz}{2r^5},
\]

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial z \partial x} = \frac{54zx}{\rho^5}.
\]

(3.10)

The solutions of this system of 9 nonlinear equations (3.9) and (3.10) can be classified into eight distinct cases, each corresponding to a particular crystal morphology:

<table>
<thead>
<tr>
<th>Case</th>
<th>x</th>
<th>y</th>
<th>z</th>
<th>Observed?</th>
<th>Reason not observed</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>Coulomb repulsion</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>Yes</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>Violation of cylindrical symmetry</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>Violation of cylindrical symmetry</td>
</tr>
<tr>
<td>6</td>
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<td>0</td>
<td>0</td>
<td>Yes</td>
<td></td>
</tr>
<tr>
<td>7</td>
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<td>0</td>
<td>0</td>
<td>Yes</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>Yes</td>
<td></td>
</tr>
</tbody>
</table>

We note that case 4, corresponding to a tilted rod crystal, is not observed, though an immediate explanation for its absence is lacking. Cases 2, 6, 7, and 8 in the above table are all observed in our numerical simulations and correspond to the rod, popout, tilted, and planar crystal phases respectively. We now discuss the observed Rod ↔ Popout, Popout ↔ Tilted, and Tilted ↔ Planar transitions in detail.
1. Rod ↔ Popout

Solving (3.9) for the rod-phase case \((x = 0, y = 0, z)\) yields \(z^3 = \frac{5}{8(q^2 - a)}\) for the coordinates of the potential minima in terms of the parameter \(a\). Inserting this expression for \(z(a)\) into (3.10) gives zero for all mixed derivatives and a positive value for the second derivative in \(z\). The rod crystal transitions to the popout configuration when stability is lost in \(x\) direction, and would transition to the tilted rod configuration (case 4) when stability is lost in the \(y\) direction, i.e., when the second derivatives of \(x\) and \(y\) are zero, respectively. These conditions can be expressed mathematically as

\[
\frac{\partial^2 U^{(s)}_{\text{eff}}}{\partial x^2} = 0 \rightarrow a = \frac{43}{58} q^2, \\
\frac{\partial^2 U^{(s)}_{\text{eff}}}{\partial y^2} = 0 \rightarrow a = \frac{1}{2} q^2.
\]  

(3.11)

Thus, it is clear that the tilted rod phase does not occur, since stability is lost in \(x\) before it is lost in \(y\). The loss in stability in \(x\) at \(a = \frac{43}{58} q^2\) marks the boundary with the popout morphology region and is depicted in Fig. 3.5.

2. Popout ↔ Tilt

Beginning in the popout phase \((x, y = 0, z)\), (3.9) yields \(\rho(y = 0) = \sqrt{9x^2 + z^2} = \left[\frac{3}{(a + q^2/2)}\right]^{1/3}\) and \(z = \left[\frac{3}{(22q^2 - 28a)}\right]^{1/3}\). The \(x\), \(y\) and \(z\) mixed derivatives in (3.10) are zero, the \(z, x\) mixed derivative is positive, and the second derivatives in \(x\) and \(z\) are also positive. The popout crystal begins to tilt when the second derivative in \(y\) is zero, and therefore, popout phase stability is lost when

\[
\frac{\partial^2 U^{(s)}_{\text{eff}}}{\partial y^2} = 0 \rightarrow a = \frac{1}{2} q^2.
\]  

(3.12)

This expression marks the boundary between the popout and tilted phases and is depicted in Fig. 3.5.
Section 3.3. Failure of the Standard Pseudopotential

3. Planar ↔ Tilt

Solving (3.9), beginning in the planar case \((x, y, z = 0)\), yields \(y = \sqrt{3}x = \left[3/(a + q^2/2)\right]^{1/3}/2\). The mixed derivatives in \(y, z\) and \(z, x\) are zero, in \(x, y\) are positive, and second derivatives in \(x\) and \(y\) are zero. Planar crystals transition to the tilted configuration when stability is lost in \(z\) and thus, when

\[
\frac{\partial^2 U_{\text{eff}}^{(s)}}{\partial z^2} = 0 \rightarrow a = \frac{1}{2} q^2,
\]

(3.13)
giving the boundary between the planar to the tilted phases.

Figure 3.5 depicts the regions in the stability zone where the four morphologies are found, together with the analytical boundary lines between these regions as predicted by the standard pseudopotential. The most salient feature of this figure is that the analytical boundary lines for the Popout ↔ Tilted and Planar ↔ Tilted transitions occur along the same curve in the stability region. This implies that the tilted phase exists only on the line \(a = q^2/2\), which is plainly incorrect, as can be discerned from the numerical data in Fig. 3.5. While the analytical boundary for the Rod ↔ Popout is quite accurate, even for large \(q\), the standard pseudopotential is completely incapable of predicting the existence of the more complicated tilted morphology observed for three ions. Repeating the above procedure for 2 and 4 ions, we observe a similar phenomenon.

In Fig. 3.4, for example, we see that the two-ion standard pseudopotential is unable to predict the tilted two-ion crystal, which occupies the blue region in the figure and is illustrated in Fig. 3.1(b). In Fig. 3.6 we see that the four-ion standard pseudopotential is unable to predict many four-ion crystals, including the diamond, popped-out diamond, tilted, and double-rod phases as shown in Fig. 3.3.

Thus, despite the accuracy of the standard pseudopotential in predicting the existence of certain crystal morphologies and boundary lines – particularly the boundary of the rod region – it seems to be incapable of predicting qualitatively significant features of other crystals.
Figure 3.4: Analytical standard pseudopotential predictions for two-ion crystal morphology boundaries. The yellow, blue, and purple regions depict where rod, tilted, and planar crystals form, respectively. The red region depicts a crystal-free region, i.e. crystals do not form there. The solid black curve is the analytical boundary between both the rod and tilted and the tilted and planar phases, as predicted by the standard pseudopotential.
Figure 3.5: Analytical standard pseudopotential predictions for three-ion crystal morphology boundaries. The green, yellow, blue, and magenta regions depict where rod, popout, tilted, and planar crystals form respectively. The red region depicts a crystal-free region. The upper dashed black curve is the analytical boundary between the rod and popout regions, as well as the popout to tilted regions. The lower dashed black curve is the analytical boundary between the tilted and planar regions.
Figure 3.6: Analytical standard pseudopotential predictions for four-ion crystal morphology boundaries. The red, orange, black, green, magenta, blue, and sea green regions depict where rod, zigzag, diamond, popped-out diamond, tilted, double-rod, and planar crystals form respectively. The grey region depicts a crystal-free region. The solid black, white, and red lines are the analytical boundaries between the rod and zigzag, zigzag and diamond, and planar and double rod regions respectively, as predicted by the four-ion standard pseudopotential.
3.4 An Improved Pseudopotential

3.4.1 Three General Coupled Equations of Motion

Given the failure of the standard pseudopotential to account for these “exotic” crystal configurations, i.e., crystal configurations beyond the linear chain, zigzag crystal, and the planar crystal, we develop here an improved pseudopotential that is capable of doing so. The derivation is based on a method for computing an improved two-ion pseudopotential, developed in [45], but generalizes the method to a system of $N$ ions. A full treatment of a general system of $N$ coupled equations is given in Ref. [64]. In this section, we focus on the three-ion case, to capture the basic idea behind the method.

The main difference between the standard and improved pseudopotentials is that in the latter the expansion of the trapping and Coulomb forces in terms of the micromotion amplitudes, as was performed in (2.20), is carried out up to second order rather than the first. Before looking specifically at the case of three ions in a Paul trap, we will first derive an improved pseudopotential for a general set of three coupled differential equations of the form

\begin{align*}
    m_X \ddot{X} &= -U_X(X, Y, Z) - k_Y X \cos(\omega t), \\
    m_Y \ddot{Y} &= -U_Y(X, Y, Z) - k_Y Y \cos(\omega t), \\
    m_Z \ddot{Z} &= -U_Z(X, Y, Z) - k_Z Z \cos(\omega t),
\end{align*}

(3.14)

where $m_I, I \in \{X, Y, Z\}$ are effective masses, $U_I = \partial U / \partial I$, and $k_I$ are constants. These equations apply to numerous periodically-driven systems, not only the 3-ion cylindrical Paul trap. In Chapter 4, for example, we will see how these can be applied to the two-ion linear Paul trap.

As before, we assume that the actual motion of an ion can be split into a slow macro-
motion and a fast micromotion, according to

\[ X(t) = x(t) + \xi \cos(\omega t), \]
\[ Y(t) = y(t) + \eta \cos(\omega t), \]
\[ Z(t) = z(t) + \zeta \cos(\omega t), \]  
(3.15)

where \( x, y, z \) are secular, macromotion coordinates and \( \xi, \eta, \zeta \) are the amplitudes of the fast-varying, sinusoidal micromotion in \( x, y, z \) respectively. We assume that the micromotion amplitudes vary slowly on the macromotion timescale and hence, can be taken to be constant over a cycle of the trapping potential. Assuming they are small, we now expand \( U(X, Y, Z) \) in the micromotion amplitudes around the secular coordinates up to second order

\[ U(X, Y, Z) \approx U(x, y, z) + U_x \xi \cos(\omega t) + U_y \eta \cos(\omega t) + U_z \zeta \cos(\omega t) \]
\[ + \frac{1}{2} U_{xx} \xi^2 \cos^2(\omega t) + \frac{1}{2} U_{yy} \eta^2 \cos^2(\omega t) + \frac{1}{2} U_{zz} \zeta^2 \cos^2(\omega t) \]
\[ + U_{xy} \xi \eta \cos^2(\omega t) + U_{yz} \eta \zeta \cos^2(\omega t) + U_{zx} \zeta \xi \cos^2(\omega t), \]  
(3.16)

where, for \( i, j \in \{ x, y, z \} \) \( (I, J \in \{ X, Y, Z \}) \), \( U_i \) or \( U_{ij} \) denote, in their respective order, \( \partial U/\partial I \) or \( \partial^2 U/\partial I \partial J \), evaluated at \( X = x, Y = y, \) and \( Z = z \).

Now we need to calculate the first derivatives of (3.16) in \( X, Y, Z \). For \( X \), this gives

\[ U_X \approx U_x + U_{xx} \xi \cos(\omega t) + U_{yx} \eta \cos(\omega t) + U_{zx} \zeta \cos(\omega t) \]
\[ + \frac{1}{2} U_{xxx} \xi^2 \cos^2(\omega t) + \frac{1}{2} U_{yxx} \eta^2 \cos^2(\omega t) + \frac{1}{2} U_{zxx} \zeta^2 \cos^2(\omega t) \]
\[ + U_{xyx} \xi \eta \cos^2(\omega t) + U_{yxx} \eta \zeta \cos^2(\omega t) + U_{zxx} \zeta \xi \cos^2(\omega t). \]  
(3.17)

The derivatives in \( Y \) and \( Z \) have the exact same form and can be calculated straightforwardly. We now insert (3.15), as well as (3.17) and its counterparts in \( Y \) and \( Z \), into our original equations of motion (3.14). Equating first-order \( \cos(\omega t) \) terms on the LHS and RHS of this equation, we find
\[ m_X \omega^2 \xi = U_{xx} \xi + U_{yx} \eta + U_{zx} \zeta + k_X x, \]
\[ m_Y \omega^2 \eta = U_{yy} \eta + U_{zy} \zeta + U_{xy} \xi + k_Y y, \]
\[ m_Z \omega^2 \zeta = U_{zz} \zeta + U_{xz} \xi + U_{yz} \eta + k_Z z. \] (3.18)

These three algebraic equations can now be solved for the micromotion amplitudes \( \xi, \eta, \zeta \), giving
\[ \xi = \{ k_X x[(m_Y \omega^2 - U_{yy})(m_Z \omega^2 - U_{zz}) - U_{yz}^2] \]
\[ + k_Y y[U_{yz}(m_Z \omega^2 - U_{zz}) + U_{xz} U_{yz}] \]
\[ + k_Z z[U_{xz}(m_Y \omega^2 - U_{yy}) + U_{xy} U_{yz}] \} / \Delta, \]
\[ \eta = \{ k_Y y[(m_Z \omega^2 - U_{zz})(m_X \omega^2 - U_{xx}) - U_{xx}^2] \]
\[ + k_Z z[U_{xy}(m_X \omega^2 - U_{xx}) + U_{yx} U_{xx}] \]
\[ + k_X x[U_{xx}(m_Y \omega^2 - U_{yy}) + U_{zy} U_{xy}] \} / \Delta, \]
\[ \zeta = \{ k_Z z[(m_X \omega^2 - U_{xx})(m_Y \omega^2 - U_{yy}) - U_{xy}^2] \]
\[ + k_X x[U_{xx}(m_Y \omega^2 - U_{yy}) + U_{zy} U_{xy}] \]
\[ + k_Y y[U_{yz}(m_X \omega^2 - U_{xx}) + U_{zx} U_{yz}] \} / \Delta, \] (3.19)

where
\[ \Delta = (m_X \omega^2 - U_{xx})(m_Y \omega^2 - U_{yy})(m_Z \omega^2 - U_{zz}) \]
\[ - (m_X \omega^2 - U_{xx})U_{yz}^2 - (m_Y \omega^2 - U_{yy})U_{zx}^2 \]
\[ - (m_Z \omega^2 - U_{zz})U_{xy}^2 - 2U_{xy}U_{yz}U_{zx}. \] (3.20)

Once again inserting (3.15), as well as (3.17) and its counterparts in \( Y \) and \( Z \), into (3.14), but now averaging over one cycle of the driving field, we obtain
Section 3.4. An Improved Pseudopotential

\[ m_X \ddot{x} = -U_x - \frac{1}{4} U_{xxx} \xi^2 - \frac{1}{4} U_{yyx} \eta^2 - \frac{1}{4} U_{zzx} \zeta^2 \]
\[ - \frac{1}{2} U_{xyx} \xi \eta - \frac{1}{2} U_{xyz} \xi \zeta - \frac{1}{2} U_{zzx} \zeta \xi - \frac{1}{2} k_X \xi, \]
\[ m_Y \ddot{y} = -U_y - \frac{1}{4} U_{xx} \xi^2 - \frac{1}{4} U_{yy} \eta^2 - \frac{1}{4} U_{zz} \zeta^2 \]
\[ - \frac{1}{2} U_{xy} \xi \eta - \frac{1}{2} U_{yy} \eta \xi - \frac{1}{2} U_{zz} \zeta \eta - \frac{1}{2} k_Y \eta, \]
\[ m_Z \ddot{z} = -U_z - \frac{1}{4} U_{xx} \xi^2 - \frac{1}{4} U_{yy} \eta^2 - \frac{1}{4} U_{zz} \zeta^2 \]
\[ - \frac{1}{2} U_{xyz} \xi \eta - \frac{1}{2} U_{yyz} \eta \zeta - \frac{1}{2} U_{zzx} \zeta \xi - \frac{1}{2} k_Z \zeta. \] (3.21)

These equations can be derived from an effective potential given by
\[ U_{\text{eff}}(x, y, z) = U(x, y, z) + \frac{1}{4} k_X x \xi + \frac{1}{4} k_Y y \eta + \frac{1}{4} k_Z z \zeta, \] (3.22)
which is the pseudopotential for this three-coordinate system.

3.4.2 Three Ions in a Paul trap

Now that we have an expression for the improved pseudopotential for three general equations of the form given by (3.14), we now look specifically at the 3-ion Paul trap. By directly comparing (3.14), the three general equations, with (3.6), the three equations describing the three-ion system, we see that
\[ m_X = 6, \quad m_Y = 2, \quad m_Z = 2, \quad \omega = 2, \quad k_X = 12q, \]
\[ k_Y = 4q, \quad k_Z = -8q, \] and
\[ U(X, Y, Z) = \frac{2}{(9X^2 + Y^2 + Z^2)^{1/2}} + \frac{1}{2(Y^2 + Z^2)^{1/2}} + a \left[ 3X^2 + Y^2 - 2Z^2 \right]. \] (3.23)

The actual coordinates \((X, Y, Z)\) in (3.23) can be replaced with the secular motion coordinates \((x, y, z)\) by taking a cycle average. To finally obtain the three-ion improved pseudopotential (3.22), we need to compute the partial derivatives of \(U\) in (3.23). These
where \( \rho = (9x^2 + y^2 + z^2)^{1/2} \) and \( r = (y^2 + z^2)^{1/2} \). Applying these in our general framework, we arrive at the three-ion improved pseudopotential

\[
U_{\text{eff}}(x,y,z) = \frac{2}{\rho} + \frac{1}{2r} + 3ax^2 + ay^2 - 2az^2 + \frac{3qx\xi\Delta + qy\eta\Delta - 2qz\zeta\Delta}{\Delta},
\]

where the last term on the right-hand side may be evaluated explicitly using (3.19) and (3.20) with the partial derivatives (3.24).

### 3.5 Analytical Predictions of the Improved Pseudopotential

Armed with the three-ion improved pseudopotential, we now proceed to analytically predict the boundaries between the four three-ion crystal morphologies. The procedure is exactly the same as that outlined in Section 3.3.2, with the standard pseudopotential replaced by the improved version in (3.25). Thus, for each transition we need to solve a fixed set of equations, as described below.

(i) Rod \( \leftrightarrow \) Popout

\[
\frac{\partial U_{\text{eff}}(x = 0, y = 0, z)}{\partial z} = 0, \quad \frac{\partial^2 U_{\text{eff}}(x, y = 0, z)}{\partial x^2} \bigg|_{x=0} = 0.
\]
(ii) Popout $\leftrightarrow$ Tilt
\[ \frac{\partial U_{\text{eff}}(x, y = 0, z)}{\partial z} = 0, \quad \frac{\partial U_{\text{eff}}(x, y = 0, z)}{\partial x} = 0, \quad \frac{\partial^2 U_{\text{eff}}(x, y, z)}{\partial y^2} \bigg|_{y=0} = 0, \quad (3.27) \]

(iii) Tilted $\leftrightarrow$ Planar
\[ \frac{\partial U_{\text{eff}}(x, y = \sqrt{3}x, z = 0)}{\partial x} = 0, \quad \frac{\partial^2 U_{\text{eff}}(x, y = \sqrt{3}x, z)}{\partial z^2} \bigg|_{z=0} = 0. \quad (3.28) \]

Given the complicated form of (3.25), solving the above equations analytically is not trivial. Instead, we find these solutions numerically. The solutions are illustrated by the three solid lines depicted in Fig. 3.7, and each one represents a boundary between two different three-ion morphologies. These lines approximate the boundaries observed in our numerical simulations extremely well. Furthermore, the improved pseudopotential, unlike its standard counterpart, is capable of predicting both the existence and location of the tilted crystal morphology in parameter space.

As in the previous section, we derive an improved pseudopotential for two and four ions, and carry out a similar procedure for finding the boundary lines. We obtain the boundary lines depicted in Figs. 3.8, 3.9 and 3.10. The two-ion improved pseudopotential predicts the existence of the tilted crystal region and the boundaries of the other two regions with better accuracy than the standard pseudopotential, whose predictions are given by the dashed black curve. The four-ion pseudopotential predicts all but one of the four-ion crystal morphology boundaries.
Figure 3.7: Semi-analytical improved pseudopotential predictions for three-ion crystal morphology boundaries. The green, yellow, blue, and magenta regions depict where rod, popout, tilted, and planar crystals form respectively. The red region depicts a crystal-free region. The upper, central, and lower black curved represent the semi-analytical boundary between the rod and popout, popout and tilted, and tilted and planar regions respectively. The semi-analytical boundaries match the numerical boundaries very well.
Figure 3.8: Semi-analytical improved pseudopotential predictions for two-ion crystal morphology boundaries. The green, yellow, and pink regions depict where rod, tilted, and planar crystals form respectively. The red region depicts a crystal-free region, i.e., crystals do not form there. The upper and lower solid black curves are the semi-analytical boundaries between (i) the rod and tilted, and (ii) the tilted and planar phases respectively. The dashed black line is the standard pseudopotential prediction for these boundaries.
Figure 3.9: Semi-analytical improved pseudopotential predictions for four-ion crystal morphology boundaries. The red, orange, black, green, magenta, blue, and sea green regions depict where rod, zigzag, diamond, popped-out diamond, tilted, double-rod, and planar crystals form respectively. The grey region depicts a crystal-free region. The black, blue, white, and red points delineate the semi-analytical boundaries between the rod and zigzag, zigzag and tilted, tilted and double rod, and double rod and planar crystal regions, respectively.
Figure 3.10: Semi-analytical improved pseudopotential predictions for four-ion crystal morphology boundaries (zoomed). The red, orange, black, green, magenta, blue, and sea green regions depict where rod, zigzag, diamond, popped-out diamond, tilted, double-rod, and planar crystals form, respectively. The black triangles and black squares delineate the semi-analytical boundaries between the zigzag and diamond, and diamond and popped diamond crystal regions respectively.
Chapter 4

Linear Paul Trap

Armed with a general procedure for deriving the improved pseudopotential for any system of dynamical equations of the form (3.14), we now apply this method to the case of two ions confined in a linear Paul trap, as an example of a periodically-driven AMO system other than the cylindrical Paul trap, for which (a) the improved pseudopotential can be derived, and (b) the improved pseudopotential predicts physics that goes beyond that of its standard counterpart. Additionally, our analysis lead us, in passing, to correct an erroneous result published in the peer-reviewed literature [58].

The linear Paul trap is a particularly significant system, because of its ubiquitous usage in the trapped-ion quantum computing community [13–16]. Two conditions must be satisfied in order to achieve a working quantum computer: (a) qubits (here, the trapped ions) must be prepared and preserved in an entangled state, and (b) the spatial location of the qubits must be known so that they may be accessed by electromagnetic pulses [13–16]. The fast-varying, spatially dependent micromotion exhibited by particles in Paul traps has usually been an impediment to achieving both of these and thus, minimizing micromotion has been a goal of experimental quantum computing research. In the cylindrical trap, micromotion vanishes at only a single spatial coordinate, i.e., the central origin of the trap. In the linear Paul trap, however, micromotion is zero along an entire
axis of the trap, as will be explained in the next section. This advantage provides the basis for the preference of these traps in the quantum computing literature.

In Section 4.1, we introduce the architecture of the linear Paul trap and provide a mathematical description of the trapping potential. Then, in Section 4.2, we proceed to derive the equations of motion for one and two ions in the trap. In Section 4.3, we detail our numerical results for the two-ion system, comparing them to results from the literature. In Section 4.4 we derive both the standard and improved pseudopotentials for the two-ion system, providing proof of the existence of “exotic” crystals in the two-ion system. We use these numerical and analytical results to correct an error published in the literature [58].

4.1 Trap Architecture

The fundamental trapping principles, as outlined in Section 2.1, that underlie the functionality of the cylindrical Paul trap – including the use of dynamic rather than solely static fields and the requirement that the trapping potential must satisfy Laplace’s equation – apply to the linear Paul trap as well [43, 44]. The main difference between the two traps is in their design. The linear Paul trap generally consists of four segmented metallic rods symmetrically placed as per Fig. 4.1 [58, 59]. Here, we investigate the trap architecture as given in Ref. [58]. The radial directions here are x and y, and the axial direction, i.e., the direction along the length of the rods, is z.

Each rod in Fig. 4.1 has three segments: four shorter central segments and eight longer end-cap segments [58]. An ac voltage is applied to all the electrodes, with amplitude $U_{RF}$ and frequency $\Omega$. A dc voltage $U_{end}$ is applied to the eight end-cap electrodes to generate a confining potential in the axial direction. An additional voltage $U_{dc}$ can be applied to diagonal electrodes, though we take $U_{dc} = 0$ here.
Section 4.2. Equations of Motion for 1 and 2 Ions

Figure 4.1: Linear Paul trap architecture as shown in Fig. 1 of [58]. Each rod is sectioned into three components. A dc voltage $U_{\text{end}}$ is applied to all eight end-electrode pieces. The voltages of each of the segments are: $U_1(t) = -(U_{RF}/2) \cos \Omega t$, $U_2(t) = (U_{RF}/2) \cos \Omega t + U_{dc}$, $U_3(t) = -(U_{RF}/2) \cos \Omega t + U_{\text{end}}$, and $U_4(t) = (U_{RF}/2) \cos \Omega t + U_{dc} + U_{\text{end}}$. $U_{RF}$ is the amplitude of the ac voltage applied to the electrodes. $\Omega$ is the frequency of the ac voltage. $U_{dc}$ is an additional dc voltage applied to diagonal electrodes. Here we take $U_{dc} = 0$.

The trap potential resulting from these ac and dc voltages has the form

$$\Phi(x, y, z, t) = \frac{U_{RF}}{2r_0^2} (x^2 - y^2) \cos \Omega t + \frac{U_{\text{ax}}}{2r_0^2} [2z^2 - (x^2 + y^2)], \tag{4.1}$$

where $r_0$ is the radial distance between the axial axis of the trap and the rod electrodes and $U_{\text{ax}}$ is proportional to $U_{\text{end}}$ (see Fig. 4.1).

4.2 Equations of Motion for 1 and 2 Ions

By using the following dimensionless parameters,

$$\tau \equiv \frac{\Omega t}{2}, \quad a \equiv \frac{4QU_{\text{ax}}}{M\Omega^2 r_0^2}, \quad q \equiv \frac{2QU_{RF}}{M\Omega^2 r_0^2},$$
Figure 4.2: Single-ion stability zone. By integrating the single-ion equations of motion (4.2), we can determine the $a$ and $q$ values for which ion motion is bounded. The black region gives the region in $(q,a)$ parameter space where the ion is confined.

where $M$ is the mass of the ion, and using (4.1), we can immediately write down the following set of equations for the motion of a single ion in the linear Paul trap

$$\ddot{x} - (a - 2q \cos 2\tau) x = 0,$$
$$\ddot{y} - (a + 2q \cos 2\tau) y = 0,$$
$$\ddot{z} + 2az = 0.$$  \hspace{1cm} (4.2)$$

Numerically solving these equations using a 5th-order Runge-Kutta integrator \cite{52}, we can identify a region of stable confinement of the single ion in the parameter space defined by the trapping parameters $a$ and $q$. This region is depicted in Fig. 4.2. Moreover, as in the cylindrical Paul trap, since the single-ion equation of motion is equivalent to the center-of-mass equation of motion for an $N$-ion system, the black region delineated in Fig. 4.2 is also the region in parameter space where we will later search for two-ion crystals.

We now turn to the main system of interest in this chapter: the two-ion linear Paul trap. For two ions, we can separate the motion of the two particles into a center-of-mass motion and a relative motion. The center-of-mass equation is equivalent to the
Section 4.3. Numerical Results and Mismatch with the Literature

single-ion equation of motion. The three relative-motion equations are [58]

\[\ddot{x} - (a - 2q \cos 2\tau)x = \frac{x}{\rho^3},\]
\[\ddot{y} - (a + 2q \cos 2\tau)y = \frac{y}{\rho^3},\]
\[\ddot{z} + 2az = \frac{z}{\rho^3},\] (4.3)

where \(x\), \(y\), and \(z\) are now the relative coordinates of the two ions, and \(\rho = (x^2 + y^2 + z^2)^{\frac{1}{2}}\) is the distance between the two ions.

4.3 Numerical Results and Mismatch with the Literature

By following the procedure outlined in Section 3.2.1, the two-ion crystal configurations can be found using (4.3). According to Ref. [58], there are only two such configurations. These correspond to (a) a rod crystal, where the two ions are aligned along the \(z\)-axis, i.e., their coordinates are \((0, 0, z)\) and \((0, 0, -z)\), and (b) a planar crystal, where the two ions are positioned symmetrically in the \(xy\)-plane, with coordinates \((x, y, 0)\) and \((-x, -y, 0)\). This would suggest that for any \(a\), there exists some critical \(q\) at which the two ions would switch discontinuously from a rod to a planar configuration, unlike the two-ion cylindrical Paul trap, where there is an intermediate tilted configuration.

Our numerical results indicate the existence of a third tilted morphology in addition to the two described above. In this tilted morphology, the coordinates of the two ions are \((x, y, z)\) and \((-x, -y, -z)\). The three morphologies are depicted in Fig. 4.3. A possible explanation for this mismatch in numerical results may be the precision of our simulations compared to those used in Ref. [58]. Though tilted crystals exist, our simulations indicate that they are stable only for very few choices of trap parameters. For \(q = 0.6\), for example, the range of \(a\) values for which tilted crystals are stable is on the order of \(10^{-4}\). This begs the question: are these crystals real or are they a numerical anomaly?
Numerically, a simple dragging experiment can be done to check this. Beginning in the rod configuration, if we slowly increase $a$, the two ions switch into the tilted configuration. Increasing $a$ further, the two ions tilt further and further towards the $xy$-plane, until they come to lie in the $xy$-plane as a planar configuration. Thus, the tilt of the axis between the two ions can be controlled continuously as a function of $a$. The shift from the rod to the planar phase is smooth, rather than a discontinuous jump, resembling the behaviour of the two-ion cylindrical Paul trap.

This numerical experiment is compelling evidence for the existence of tilted crystals, but not sufficient. The authors of Ref. [58] provide, in addition, an analytical prediction of their numerical results. Using the machinery in Ref. [45], they derive an improved pseudopotential for the two-ion linear Paul trap. They use this pseudopotential to compute the boundaries between the rod and planar crystal, as was performed in section 3.3.2 and find that it does not predict the existence of a third, tilted morphology.

This is a surprising result since, in the cylindrical Paul trap the power of the improved pseudopotential is its ability to predict the existence of tilted, “exotic” configurations that the standard pseudopotential does not. To resolve this apparent conflict, we re-derive the improved pseudopotential for the two-ion system in the next section.

Figure 4.3: Two-ion Crystals in a linear Paul trap. The grey points give the positions of the two ions in each of the three crystal morphologies.
4.4 Pseudopotentials for the 2-Ion Linear Paul Trap

Since we already possess expressions for the standard and improved pseudopotentials for a system of three general equations, to derive these for the two-ion linear Paul trap, we need only compare \(4.3\) with \((2.33)\) and \((3.14)\) respectively. The static component \(U\) of the potential whose derivatives appear in the two-ion equations of motion is given by

\[
U(x, y, z) = \frac{1}{2}a[2z^2 - x^2 - y^2] + \frac{1}{\rho}.
\]  

(4.4)

For the standard pseudopotential, \(m_i = 1\) for all \(i = x, y, z\), \(\omega = 2\), \(f_x = -2qx\), \(f_y = 2qy\), and \(f_z = 0\). As per \((2.34)\), the standard pseudopotential is given by

\[
U_{\text{standard}} = \frac{1}{2}a[2z^2 - x^2 - y^2] + \frac{1}{\rho} + \frac{1}{4}q^2[x^2 + y^2],
\]  

(4.5)

where \(\rho = (x^2 + y^2 + z^2)^{1/2}\). For the improved pseudopotential, \(m_X = m_Y = m_Z = 1\), \(k_X = 2q\), \(k_Y = -2q\), and \(k_Z = 0\). As per \((3.22)\), the improved pseudopotential is given by

\[
U_{\text{eff}} = \frac{1}{2}a[2z^2 - x^2 - y^2] + \frac{1}{\rho} + \frac{1}{2}q[x\xi - y\eta],
\]  

(4.6)

where the last term may be explicitly evaluated using \((3.19)\), \((3.20)\), and the second derivatives

\[
\begin{align*}
\frac{\partial^2 U}{\partial x^2} &= -a + \frac{3x^2}{\rho^5} - \frac{1}{\rho^3}, \\
\frac{\partial^2 U}{\partial y^2} &= -a + \frac{3y^2}{\rho^5} - \frac{1}{\rho^3}, \\
\frac{\partial^2 U}{\partial z^2} &= 2a + \frac{3z^2}{\rho^5} - \frac{1}{\rho^3}, \\
\frac{\partial^2 U}{\partial x \partial y} &= \frac{3xy}{\rho^5}, \\
\frac{\partial^2 U}{\partial y \partial z} &= \frac{3yz}{\rho^5}, \\
\frac{\partial^2 U}{\partial z \partial x} &= \frac{3zx}{\rho^5}.
\end{align*}
\]

(4.7)
We may now use (4.6) to obtain the boundaries between the three crystal morphologies in Fig. 4.3. We solve the following set of simultaneous equations for the two expected transitions:

(i) Rod ↔ Tilt

\[
\frac{\partial U_{\text{eff}}(x = 0, y = 0, z)}{\partial z} = 0, \quad \frac{\partial^2 U_{\text{eff}}(x, y = 0, z)}{\partial x^2} \bigg|_{x=0} = 0, \tag{4.8}
\]

The first equation yields \( z^3 = 1/2a \), which can be inserted into the second equation to yield

\[
a = \frac{1}{3}(-2 + \sqrt{2}/2 + q^2), \tag{4.9}
\]

which gives the boundary line between the rod and tilted crystal regions in the two-ion stability region.

(ii) Tilt ↔ Planar

\[
\frac{\partial U_{\text{eff}}(x, y, z = 0)}{\partial x} = 0, \quad \frac{\partial^2 U_{\text{eff}}(x, y, z)}{\partial z^2} \bigg|_{z=0} = 0. \tag{4.10}
\]

These equations are difficult to solve analytically, so we use a numerical simultaneous equation solver instead. The resulting curve, delineating the Tilt ↔ Planar boundary, is shown in Figs. 4.4 and 4.5 alongside (4.9). The predicted boundaries match the observed numerical boundaries reasonably well, though they begin to deviate for larger \( q \) values. The spacing between the two curves is relatively small, indicating that though two-ion tilted crystals do exist, the region where they do is quite small.
Figure 4.4: Analytical and semi-analytical boundaries for two-ion crystals. The green, yellow, and blue regions indicate where planar, tilted, and rod crystals form, respectively. Due to the resolution of the plot, the yellow region is difficult to discern, though we can make out some yellow points. The solid black line is the boundary between the rod and tilted configurations. The solid yellow line is the boundary between the tilted and planar configurations.

Figure 4.5: Analytical and semi-analytical boundaries for two-ion crystals (zoomed). The green, yellow, and blue regions indicate where planar, tilted, and rod crystals form, respectively. Here, we look specifically at the region between $q = 0.6$ and $q = 0.61$. The solid black line is the boundary between the rod and tilted configurations. The solid yellow line is the boundary between the tilted and planar configurations.
Chapter 5

Delineating Crystal-Free Regions

In this chapter, we discuss a limitation of the improved pseudopotential. The stability diagrams for 2, 3, and 4 ions, shown in Chapter 3, all indicate that for certain choices of $a$ and $q$ crystals do not form. Numerical evidence for the existence of these crystal-free regions exists in the literature as well [57], though, an analytical explanation is lacking [51 57]. The boundaries between different crystal regions were found by using the pseudopotential to identify when certain configurations became unstable. Intuitively, then, the pseudopotential’s ability to accurately determine when a crystal loses stability would make it a good tool for analytically delineating the crystal-free region described above. This turns out not to be the case.

This can be shown in the following way. Consider the case of two ions in a cylindrical Paul trap. For any point $(q, a)$ in the trap parameter space, we can find a critical point in the pseudopotential $U_{\text{eff}}(x, z)$, as given in Ref. [45], by solving the equations

$$\frac{\partial U_{\text{eff}}}{\partial x} = 0, \quad \frac{\partial U_{\text{eff}}}{\partial z} = 0.$$ 

Whether this critical point $(x_0(q, a), z_0(q, a))$ corresponds to a potential minimum can
then be determined using the Hessian matrix \[52, 53\], defined as 
\[ H_{ij} = \frac{\partial^2 U_{\text{eff}}}{\partial i \partial j}, \]
where \(i, j \in \{x, z\}\). For any point \((q, a)\) in the red, crystal-free region shown in Fig. 3.4, both the standard and improved pseudopotentials predict that the solution is a minimum and thus, stable. Indeed, the pseudopotential cannot be used to explain the loss of crystal stability in these regions, a qualitatively significant phenomenon in the Paul trap.

Analytically delineating this region, even for the two-ion case, is quite challenging \[51\]. Our goal here is more modest: to gain some analytical insight into the crystal-free region, we investigate an analytically-tractable, discrete model of the two-ion Paul trap. In this model, the continuous sine drive of the trap is replaced by alternating periodic kicks, and the continuous Coulomb force between the two particles is replaced with a discrete Coulomb “flash”. In Section 5.1, we mathematically describe this model and show that the single-ion stability region derived from the model’s governing differential equations is remarkably similar to that of the single-ion cylindrical Paul trap. We then show numerically, semi-analytically, and analytically the existence of a crystal-free region in the stable trapping space of the two-ion model. In Section 5.2, we briefly propose the existence of a region in the two-ion Paul trap parameter space, where relative motion is stable and center-of-mass motion is unstable.

5.1 Discretized Paul Trap

Let \(\vec{R}_1\) and \(\vec{R}_2\) be the positions of two ions in a cylindrical Paul trap. We can define a center-of-mass coordinate \(\vec{R}_{CM} = (\vec{R}_1 + \vec{R}_2)/2\) and a relative-motion coordinate \(\vec{\rho} = \vec{R}_1 - \vec{R}_2\), where \(\vec{\rho} = (X, Y, Z)\). The center-of-mass equation is identical to that of the single-ion equation, as was proved in Chapter 2. Due to axial symmetry of the trap, we can simplify the relative motion equations by setting \(Y = 0\). The dynamics of the two ions are then fully described by two coupled, dimensionless equations in the coordinates \(X\) and \(Z\):
Section 5.1. Discretized Paul Trap

\[ \ddot{X} + [a - 2q \cos(2t)]X = \frac{X}{(X^2 + Z^2)^{3/2}}, \quad (5.1) \]
\[ \ddot{Z} - 2[a - 2q \cos(2t)]Z = \frac{Z}{(X^2 + Z^2)^{3/2}}, \quad (5.2) \]

where \( a \) and \( q \) are the same as defined in previous chapters.

To create a discrete model of the two-ion system, we replace the cosine drive in (5.1) and (5.2) with a periodic delta kick and the Coulomb force with two Coulomb “kicks” that act in equal intervals every trap cycle. The periodic delta kicks are given by

\[ f(t) = A\delta_\pi(t) - B\delta_\pi \left( t - \frac{\pi}{2} \right), \quad (5.3) \]

where \( A \) and \( B \) are tunable parameters corresponding to the strengths of the kicks, and \( \delta_\pi \) is the \( \pi \)-periodic delta function. One trap cycle is defined to be the time between two successive kicks of strength \( A \). The periodic Coulomb “kicks” \( h_x(t) \) and \( h_z(t) \) in the \( x \) and \( z \) directions, respectively, are given by

\[ h_x(t) = \frac{\pi}{2} \frac{x}{(x^2 + z^2)^{3/2}} \delta_{\frac{\pi}{2}} \left( t - \frac{\pi}{4} \right), \quad h_z(t) = \frac{\pi}{2} \frac{z}{(x^2 + z^2)^{3/2}} \delta_{\frac{\pi}{2}} \left( t - \frac{\pi}{4} \right), \quad (5.4) \]

where \( \delta_{\frac{\pi}{2}} \) is the \( \frac{\pi}{2} \)-periodic delta function. Given the discrete nature of the model, the motion of the two ions during any one trap cycle can then be broken down into eight well-defined steps: (i) a kick of strength \( A \), (ii) free motion, (iii) a Coulomb kick, (iv) free motion, (v) a kick of strength \( B \), (vi) free motion, (vii) a second Coulomb kick, (viii) free motion. We now choose \( A \) and \( B \) such that \( f(t) \) approximates

\[ g(t) = a - 2q \cos(2t), \quad (5.5) \]

up to first order. Writing \( f(t) \) as a Fourier series,

\[ f(t) = \frac{A}{\pi} \sum_{j=-\infty}^{N=\infty} e^{2\pi ij} - \frac{B}{\pi} \sum_{j=-\infty}^{N=\infty} e^{2\pi ij \left( t - \frac{\pi}{2} \right)}, \quad (5.6) \]

and expanding as a cosine series, we have

\[ f(t) = \frac{A}{\pi} - \frac{B}{\pi} + 2 \left[ \frac{A}{\pi} + \frac{B}{\pi} \right] \cos(2t) + ... \quad (5.7) \]
Equating the zeroth- and first-order cosine terms in (5.7) and (5.5), we have

\[ A = \frac{\pi}{2} (a - q), \quad B = -\frac{\pi}{2} (a + q). \]  

(5.8)

Choosing appropriate length units, the equations of motion for the discrete model then read

\[ \ddot{x} + \left[ \frac{\pi}{2} (a - q) \delta_x(t) + \frac{\pi}{2} (a + q) \delta_x \left( t - \frac{\pi}{2} \right) \right] x = \frac{\pi}{2} \frac{x}{(x^2 + z^2)^{3/2}} \delta_x \left( t - \frac{\pi}{4} \right), \quad (5.9) \]

\[ \ddot{z} - 2 \left[ \frac{\pi}{2} (a - q) \delta_x(t) + \frac{\pi}{2} (a + q) \delta_x \left( t - \frac{\pi}{2} \right) \right] z = \frac{\pi}{2} \frac{z}{(x^2 + z^2)^{3/2}} \delta_x \left( t - \frac{\pi}{4} \right). \]  

(5.10)

Let \((x_n, z_n, \dot{x}_n, \dot{z}_n)\) be the initial conditions at the beginning of a trap cycle, i.e., immediately before step (i), and \((x_{n+1}, z_{n+1}, \dot{x}_{n+1}, \dot{z}_{n+1})\) the conditions at the end of a trap cycle, i.e., immediately after step (viii). Since the equations are discretized, they are easily integrated to produce a fully analytical, though complicated, mapping \((x_n, z_n, \dot{x}_n, \dot{z}_n) \rightarrow (x_{n+1}, z_{n+1}, \dot{x}_{n+1}, \dot{z}_{n+1})\). We perform, as an example, the integration for steps (i), (ii), and (iii). Here \(i_j\) denotes the \(i\)-th coordinate immediately after step \(j\).

Integrating (5.9) and (5.10) from

(i) \(t = -\epsilon\) to \(t = \epsilon\) for \(\epsilon \to 0\), we obtain:

\[
\begin{align*}
x_1 &= x_0, \\
z_1 &= z_0, \\
\dot{x}_1 &= \dot{x}_0 - Ax_0, \\
\dot{z}_1 &= \dot{z}_0 + 2Az_0,
\end{align*}
\]  

(5.11)

(ii) \(t = 0\) to \(t = \frac{\pi}{4}\), we obtain:

\[
\begin{align*}
x_2 &= x_1 + \frac{\pi}{4} \dot{x}_1, \\
z_2 &= z_1 + \frac{\pi}{4} \dot{z}_1, \\
\dot{x}_2 &= \dot{x}_1, \\
\dot{z}_2 &= \dot{z}_1,
\end{align*}
\]  

(5.12)
(iii) \( t = -\epsilon \) to \( t = \epsilon \) for \( \epsilon \to \pi \frac{1}{4} \), we obtain:

\[
x_3 = x_2, \\
z_3 = z_2, \\
\dot{x}_3 = \dot{x}_2 + \frac{\pi}{2} \frac{x_2}{(x_2^2 + z_2^2)^{\frac{1}{2}}}, \\
\dot{z}_3 = \dot{z}_2 + \frac{\pi}{2} \frac{z_2}{(x_2^2 + z_2^2)^{\frac{1}{2}}},
\]

\( (5.13) \)

As in the regular two-ion Paul trap, we can simulate (laser) cooling in this discrete model by adding a \( \gamma \dot{x} \) and a \( \gamma \dot{z} \) term to \((5.9)\) and \((5.10)\), respectively. This only alters the free-motion steps, where, for example, \((5.14)\) is replaced by

\[
x_2 = x_1 + \frac{\dot{x}_1}{\gamma} (1 - e^{-\gamma \frac{\pi}{4}}), \\
z_2 = z_1 + \frac{\dot{z}_1}{\gamma} (1 - e^{-\gamma \frac{\pi}{4}}), \\
\dot{x}_2 = \dot{x}_1 e^{-\gamma \frac{\pi}{4}}, \\
\dot{z}_2 = \dot{z}_1 e^{-\gamma \frac{\pi}{4}}.
\]

\( (5.14) \)

The inclusion of a damping parameter allows us to find the fixed points of this mapping using the procedure in Section 3.2.1.

### 5.1.1 Single-ion Stability Region

Before discussing our two-ion model in depth, it is worth investigating the case of a single ion subjected to periodic delta kicks. The equation of motion for such a system is given by

\[
\ddot{\mathbf{r}} + [A\delta_\pi(t) - B\delta_\pi \left( t - \frac{\pi}{2} \right)] \begin{pmatrix} x \\ y \\ -2z \end{pmatrix} = 0,
\]

\( (5.15) \)

with \( A \) and \( B \) as defined in \( (5.8) \).
Choosing \( Y = 0 \) and integrating this equation as in the two-ion case, we derive a mapping \( \vec{\xi}_i(x_i, z_i, \dot{x}_i, \dot{z}_i) \rightarrow \vec{\xi}_{i+1}(x_{i+1}, z_{i+1}, \dot{x}_{i+1}, \dot{z}_{i+1}) \) from one cycle to the next, given by

\[
\begin{align*}
    x_{i+1} &= \dot{x}_i \left( 1 + \frac{\pi B}{2} \right) + x_i \left( -A + B - \frac{\pi AB}{2} \right), \\
    z_{i+1} &= \dot{z}_i \left( 1 - \pi B \right) + z_i \left( 2A - 2B - 2\pi AB \right), \\
    \dot{x}_{i+1} &= \dot{x}_i \left( \pi + \frac{\pi^2 B}{4} \right) + x_i \left( 1 - \pi A + \frac{B\pi}{2} - \frac{\pi^2 AB}{4} \right), \\
    \dot{z}_{i+1} &= \dot{z}_i \left( \pi - \frac{\pi^2 B}{2} \right) + z_i \left( 1 + 2\pi A - B\pi - \pi^2 AB \right).
\end{align*}
\] (5.16)

Solving the vector equation \( \xi_{i+1} = \xi_i \), we find that the only fixed point, irrespective of the values of \( A \) and \( B \), is \( \vec{\xi} = \vec{0} \). This means that, in the absence of noise, once the ion comes to rest at the center of the trap, it will remain there forever. This is analogous to the stroboscopic behaviour of a single ion in the regular Paul trap.

To find the values of \( A \) and \( B \) for which this fixed point is stable, we consider the Jacobian matrix of the mapping (5.16). \( x \) and \( z \) are decoupled. Therefore, we may analyze them separately. Solving the eigenvalue equations

\[
\begin{vmatrix}
    1 + \frac{\pi B}{2} - \lambda_1 & -A + B - \frac{\pi AB}{2} \\
    \pi + \frac{\pi^2 B}{4} & 1 - \pi A + \frac{B\pi}{2} - \frac{\pi^2 AB}{4} - \lambda_1
\end{vmatrix} = 0,
\] (5.17)

and

\[
\begin{vmatrix}
    1 - \pi B - \lambda_2 & 2A - 2B - 2\pi AB \\
    \pi - \frac{\pi^2 B}{2} & 1 + 2\pi A - B\pi - \frac{\pi^2 AB}{2} - \lambda_2
\end{vmatrix} = 0,
\] (5.18)

requiring that the eigenvalues \( \lambda_1, \lambda_2 \) be complex, we find that the ion is stable in the \( x \) direction, for all \( a \) and \( q \) values bounded by the lines

\[
a = \frac{8 \pm \pi^2 q}{\pi^2}, \quad a = \frac{8 \pm \sqrt{64 + \pi^4 q^2}}{\pi^2},
\] (5.19)

and stable in the \( z \) direction, for all \( a \) and \( q \) values bounded by the lines

\[
a = \frac{-4 \pm \pi^2 q}{\pi^2}, \quad a = \frac{4 \pm \sqrt{16 + \pi^4 q^2}}{\pi^2}.
\] (5.20)
Figure 5.1: Single-ion model stability zone. The four brown lines indicate the boundaries at which the fixed point at the origin of the trap in the discrete model becomes unstable. For comparison, the green lines outline the Mathieu stability region, i.e., the stable trapping region of a single ion in a regular, cylindrical Paul trap.

The four lines relevant to our analyses are depicted in Fig. 5.1. The region within the lines defines a \((q, a)\) parameter space that is remarkably similar to the Mathieu stability region shown in Fig. 2.3.

5.1.2 Numerical Results

We now return to the two-ion model. As in the regular Paul trap, a crystal in our model system corresponds to a fixed point of the cycle-to-cycle mapping \((x_n, z_n, \dot{x}_n, \dot{z}_n) \rightarrow (x_{n+1}, z_{n+1}, \dot{x}_{n+1}, \dot{z}_{n+1})\). We can identify these fixed points numerically, semi-analytically, or analytically.

The numerical protocol for finding crystals is similar to the protocol used in Section 3.2.1. We randomly initialize a vector \((x, z, \dot{x}, \dot{z})\) and then iteratively propagate it according to the damped mapping derived above. Once a crystal has been found, the damping is adiabatically switched off. When \(\gamma = 0\), the crystal is perturbed and if it persists, it is deemed stable, and if it does not, then unstable.
Figure 5.2: Numerical results for the two-ion discrete-model stability region. The black and red regions depict where two-ion crystals are stable and unstable, respectively.

Figure 5.2 depicts the results of this protocol for all choices of \((q,a)\) within the single-ion stability region, defined in Fig. 5.1. The black region shows where crystals are stable, while the red region denotes the region where crystals are unstable. The location of the crystal-free region, in the upper right corner of the stability diagram, is reminiscent of the red region seen in the regular two-ion Paul trap (see Fig. 3.4). Though the exact shapes of the red regions in the model and regular trap differ, our crude discretized picture already provides numerical evidence for the existence of a crystal-free region.

5.1.3 Semi-analytical Results

Crystal stability can be determined semi-analytically. First, we numerically solve for the fixed points \((x,z,\dot{x},\dot{z})\) for all points in the parameter space \((q,a)\), using a bisection or Newton-Raphson algorithm to solve the vector equation \((x_n,z_n,\dot{x}_n,\dot{z}_n) =\)
Section 5.2. Relative Stability, Center-of-mass Instability

\[(x_{n+1}, z_{n+1}, \dot{x}_{n+1}, \dot{z}_{n+1})\]. The Jacobian matrix for a mapping is defined as \(J_{ik} = \partial f_i / \partial k\), where \(f_i\) is the function mapping variable \(i\) and \(i, k \in \{x, z, \dot{x}, \dot{z}\}\). For each \((q, a)\) the Jacobian can be computed numerically by finding the derivatives of the functions \(f_i\) and inserting the corresponding fixed point found in the first step. Finally, by calculating the eigenvalue of the Jacobian and identifying whether it is complex or real, we can determine whether the fixed point is stable or unstable respectively.

Two kinds of fixed points were found: (i) rod and (ii) planar crystals, shown in Fig. 3.1 (a) and (c) respectively. Unlike the regular two-ion Paul trap, tilted crystals (see Fig. 3.1 (b)) were not found. The solid lines in Fig. 5.3 delineate the single-ion or two-ion center-of-mass stability. Blue and green regions within the single-ion stability region depict where rod and planar configurations are stable, respectively. Thus, the white region within the single-ion stability zone corresponds to a crystal-free zone. This matches the numerical result in Fig. 5.2. Thus, the semi-analytical results too indicate the existence of a region of no crystals in the discrete two-ion system.

5.1.4 Analytical Results

The regions of relative stability of rod and planar crystals can be found analytically. Fixed points are found as a function of parameters \(a\) and \(q\). These are then inserted into an analytical Jacobian matrix. The values of \(a\) and \(q\) at which the eigenvalues of this matrix are complex can then be found by solving an eigenvalue equation. Preliminary results are shown Fig. 5.4. Not all the stability lines have been found as yet.

5.2 Relative Stability, Center-of-mass Instability

For our two-ion model system, we note that in Fig. 5.3, the blue and green regions extend beyond the single-ion stability region delineated by the solid lines. This implies that, for certain choices of \(q\) and \(a\), the fixed points of the two-ion mapping are stable, while
Figure 5.3: Semi-analytical results for two-ion discrete-model stability region. The region bounded by the solid lines is the single-ion or center-of-mass stability region. Blue and green regions within the single-ion stability region show where stable rod and planar crystals are found, respectively. The white region in the single-ion stability zone is a crystal-free region.
Section 5.2. Relative Stability, Center-of-mass Instability

(a) Rod crystal stability lines  
(b) Planar crystal stability lines

Figure 5.4: Analytical results for two-ion discrete-model stability region. The solid lines in (a) and (b) bound the regions where rod and planar crystals are stable respectively. Certain lines are missing, for example, in (b) the boundary line between the rod and planar crystal regions.

the fixed points for the single-ion mapping are unstable, a counterintuitive proposition at first. However, we recall that our two-ion mapping describes the relative motion of the two ions. A fixed point of this mapping corresponds to a structure wherein the relative positions and velocities of the two ions are constant when viewed at the end of every trap cycle. The center-of-mass motion of the two ions, on the other hand, may be stable or unstable. If both relative and center-of-mass motion are stable, the two ions are stably confined in a crystalline state. If the relative motion is unstable and the center-of-mass motion is stable, the two ions are stably confined, but are in an attractor or chaotic cloud state. If the relative motion is stable and the center-of-mass motion unstable, we encounter a counterintuitive phenomenon, where relative displacement and velocity between the two ions is stroboscopically constant, but the absolute coordinates of the ions are increasing unboundedly. This might correspond to a situation where a two ion crystal preserves its relative spatial structure while moving away from the trap origin towards its periphery.
Since our model is meant to crudely capture the behaviour of ions in a regular cylindrical Paul trap, a natural question is: is this stable relative, unstable center-of-mass motion observed in the regular two-ion Paul trap as well? Indeed, we find for certain choices of trapping parameters \((q, a)\) in the original two-ion parameter space, relative motion is stable, while centre-of-mass motion is unstable. For \(a = 0\), for example, this can be checked numerically by initializing the system in a stable crystalline state for some \(q\) within the Mathieu stability region and then adibatically increasing \(q\). Once \(q\) exceeds the limit determined by the Mathieu stability boundary, the center-of-mass motion becomes unstable, while the relative separation between the two ions remains constant. This is qualitatively new behaviour and has not been observed in the literature before, to the best of our knowledge.
Chapter 6

Discussion and Outlook

6.1 Discussion

In this chapter we summarize and discuss the main results of this thesis. Specifically, we compare the standard and improved pseudopotentials and discuss their strengths and weaknesses in describing ion crystal morphologies across systems. Through numerical simulations of systems of 2, 3, and 4 ions in a cylindrical Paul trap, we have provided evidence for the existence of crystal configurations beyond linear ion chains, zigzag and planar crystals. The existence of these structures are, to the best of our knowledge, new, neither having been predicted nor observed experimentally before. Moreover, we have shown that the standard pseudopotential for each of these systems, as derived in Ref. [41] and commonly used in the literature, fails to both capture the existence of these “exotic” crystal morphologies and correctly describe ion positions in these “exotic” configurations. We modified the pseudopotential method in Ref. [41], deriving an improved pseudopotential for a general system of three coupled equations with periodic driving terms. A derivation for a system of $N$ such equations has been supplied in Ref. [64]. This was done by carrying out the expansion of the time-dependent potential in these equations up to second order in the micromotion coordinates, rather than first.
We used this improved pseudopotential framework to analytically predict the existence of “exotic” crystal configurations in 2, 3, and 4-ion systems in the cylindrical Paul trap, previously uncaptured by the standard pseudopotential, thus demonstrating the power of this improved method. To demonstrate the generalizability of this improved method, we applied this framework to a system of two ions in a linear Paul trap. We predicted analytically the existence of two-ion tilted crystals, that have been predicted not to exist in the literature [58]. We also showed the limitations of the pseudopotential, standard and improved, in capturing the existence of crystal-free regions in the cylindrical Paul trap. Numerical evidence for these regions exists already, but we provided some analytical insight by showing the existence of such regions in an analytically-tractable, discretized model of a two-ion Paul trap. Using results from the model system, we also reported the existence of a new kind of stability in the cylindrical Paul trap: stable relative motion and unstable center-of-mass motion.

In principle, the improved pseudopotential provides significant advantages over the standard pseudopotential in analytically confirming the existence of and describing “exotic” crystal morphologies in the Paul trap. Ion positions are more accurately captured by the improved pseudopotential – for “exotic” morphologies, which are not even captured by the standard pseudopotential, but also for traditional morphologies, such as linear ion chains. Analytical boundaries between crystal regions as predicted by the improved pseudopotential are remarkably closer to boundaries determined by numerical simulations than the standard pseudopotential.

However, this accuracy comes at the price of added complexity. Deriving the improved pseudopotential is significantly more complicated than deriving the standard pseudopotential. A system of three ions in the Paul trap is described by the 9 Cartesian coordinates giving the positions of the ions. Finding an analytical expression for the improved pseudopotential for a 9-dimensional system is too complicated. However, by exploiting symmetries found through numerical simulations and choosing an appropriate set of generalized coordinates, we can reduce this 9-dimensional problem to a 3-dimensional
one, thereby, making it analytically tractable. Thus, even for very few ion systems, deriving the improved pseudopotential effectively requires previous knowledge of the crystal configurations, which requires numerical simulations. This takes away from the analytical nature of the pseudopotential and its ease of use.

Even with this additional numerical step and using the least possible number of generalized coordinates, the improved pseudopotential might be too difficult to derive for systems with many more degrees of freedom, for instance in the range 6-10. In comparison, an analytical expression for the standard pseudopotential can be written down for any number of variables, since it is simply the sum of the pseudopotentials for each of the $N$ ions and their mutual Coulomb interactions. Even here, however, finding the positions of the $N$ ions in this standard pseudopotential is equivalent to finding the minima of a $3N$-dimensional potential, which can indeed be performed numerically for few-ion systems, but becomes increasingly difficult for larger numbers of ions. Analyzing this $3N$-dimensional standard pseudopotential to find crystal-region boundaries also becomes very difficult.

Thus, both the standard and improved pseudopotentials encounter the problem of scalability, the latter more so than the former. This is a problem in several many-body systems and can perhaps be resolved by adopting a macroscopic view of the system. Instead of starting with few ions and then increasing $N$, we could instead begin at high $N$, adopting a liquid or cloud picture, and then look at the collective modes of these $N$-ion clouds. Instead of solving coupled Newton’s equations for steady-state particle positions, we might use a differential equation that treats the system as a continuous medium (such as the Vlasov equation) and solve for some steady-state density $\rho$, allowing us to consider “exotic” density distributions. Naturally, in this macroscopic view, we would expect the minute details of ion positions to be lost. However, we might also expect better scalability, and thus greater pertinence to quantum computing applications that seek to exploit systems of thousands to millions of ions. Work on this line of research is in its nascent stages and currently being pursued.
A second issue with the pseudopotential is its inability to predict crystal-free regions in systems of 2, 3, and 4 ions in the Paul trap. Our discretized model of the two-ion Paul trap, as laid out in Chapter 4, together with numerical evidence, gives us reason to believe in the existence of this region, though an analytical treatment is still lacking. One possible explanation of this failure of the pseudopotential is that in our derivation of the pseudopotential, we assume that the macromotion coordinate changes negligibly over the course of a trap cycle. While this assumption may hold true for ions in a crystalline state, this may break down when considering energetic states in the so-called crystal-free region. In this case, a time-averaged picture of the system may be insufficient to predict the existence of the crystal-free region.

An unexpected result of the two-ion model developed in Chapter 4 is the discovery of a new kind of stability in the Paul trap. Here, relative motion between ions is stable, but the center-of-mass motion is unstable. Thus, the ions may preserve some relative spatial structure, while being ejected from the confines of the trap. Numerically, this can be confirmed by initializing the ions in a crystal state for some $q$ within the Mathieu stability region, slowly increasing $q$ beyond the Mathieu stability boundary, and then observing the stable relative motion of the ions as they are ejected from the trap. Perhaps this procedure can be used as the basis for experiment, for instance, as a source of spatially-correlated ions. From an analytical standpoint, we expect that this new kind of stability is a consequence of the discrepancy between the stability region of the regular Mathieu equation, describing the center-of-mass motion, and the modified Mathieu equation possessing an additional Coulomb term, describing the relative motion.

### 6.2 Outlook

One strength of the improved pseudopotential is that it applies to any nonlinear, periodically-driven system described by equations of the form (3.14) and may be used to analytically capture qualitatively new behaviour in such systems. In Chapter 4, we
showed this, by applying our general framework to the case of two ions in a linear Paul trap. Reference [58] claims that two-ion tilted crystals do not exist in the linear Paul trap. Our numerical simulations provide evidence that they do, but does not prove that they do. The improved pseudopotential we derive for this system actually allows us to analytically prove the existence of such crystals. Work is also being done on finding other systems to which the pseudopotential may be applied.

Linear ion chains have, thus far, been the primary candidates for trapped-ion quantum computing architectures, already having successfully performed quantum logic gates [17, 39] and formed entangled states [60, 62]. However, the 1-dimensional structure of these crystals limits the number of ions that can be stored in this way and thereby, the scalability of this architecture [59, 63]. Entangling any two arbitrary qubits in a linear ion chain experimentally also presents significant difficulties for more than tens of ions [65]. As a potential solution to these scalability issues, quantum computation with 2- and 3-dimensional crystals has been gaining interest [19–21].

Given these increased capabilities, it makes sense to explore other “exotic” crystal configurations as well. A scalable pseudopotential would allow for the description of ion positions in these structures. New computational and analytical tools for predicting novel crystal configurations may also be of interest. Machine learning techniques may be applied to automatically sort through and identify novel crystal configurations from numerical simulations, though it is preferable to develop a scalable scheme that predicts crystal morphology without such computationally-intensive procedures. Research efforts to accomplish this goal are already under way in our research group.
Bibliography


[65] Y. S. Nam, private communication.