Prospects of reaching the quantum regime in Li–Yb⁺ mixtures


View the article online for updates and enhancements.
Prospects of reaching the quantum regime in Li–Yb$^+$ mixtures

H A Fürst$^{1,7}$, N V Ewald$^1$, T Secker$^2$, J Joger$^1$, T Feldker$^1$ and R Gerritsma$^1$

$^1$ Institute of Physics, University of Amsterdam, 1098 XH Amsterdam, The Netherlands
$^2$ Eindhoven University of Technology, Post Office Box 513, 5600 MB Eindhoven, The Netherlands

E-mail: r.gerritsma@uva.nl

Received 26 July 2018, revised 2 August 2018
Accepted for publication 29 August 2018
Published 17 September 2018

Abstract

We perform numerical simulations of trapped $^{171}$Yb$^+$ ions that are buffer gas cooled by a cold cloud of $^6$Li atoms. This species combination has been suggested to be the most promising for reaching the quantum regime of interacting atoms and ions in a Paul trap. Treating the atoms and ions classically, we compute that the collision energy indeed reaches below the quantum limit for a perfect linear Paul trap. We analyze the effect of imperfections in the ion trap that cause excess micromotion. We find that the suppression of excess micromotion required to reach the quantum limit should be within experimental reach. Indeed, although the requirements are strong, they are not excessive and lie within reported values in the literature. We analyze the detection and suppression of excess micromotion in our experimental setup. Using the obtained experimental parameters in our simulation, we calculate collision energies that are a factor 2–11 larger than the quantum limit, indicating that improvements in micromotion detection and compensation are needed there. We also analyze the buffer-gas cooling of linear and two-dimensional ion crystals. We find that the energy stored in the eigenmodes of ion motion may reach 10–100 $\mu$K after buffer-gas cooling under realistic experimental circumstances. Interestingly, not all eigenmodes are buffer-gas cooled to the same energy. Our results show that with modest improvements of our experiment, studying atom–ion mixtures in the quantum regime is in reach, allowing for buffer-gas cooling of the trapped ion quantum platform and to study the occurrence of atom–ion Feshbach resonances.

Keywords: trapped ions, ultracold atoms, atom–ion collisions

(Some figures may appear in colour only in the online journal)

1. Introduction

In recent years, a novel field in atomic physics has developed in which ultracold atomic clouds are mixed with trapped ions [1–18]. These efforts aim at sympathetic cooling [16, 19, 20] of ions by atoms, and have potential applications in probing quantum many-body systems [21], quantum computation [22, 23] and quantum simulation [24]. Furthermore, Feshbach resonances are predicted to exist in atom–ion mixtures [25–29]. Such resonances play a pivotal role in neutral atom systems for the purpose of tuning the interactions between the atoms [30] and find applications in studies of quantum many-body physics [31]. However, up until now no atom–ion Feshbach resonances have been observed which is likely because the required ultracold temperatures have not been reached in these systems.

A crucial step towards realizing the applications described above is to reach the quantum (or s-wave) regime for atom–ion mixtures. While preparing atomic clouds at ultracold temperatures in the quantum degenerate regime is routinely performed in many groups using e.g. Li atoms [32–34], it turned out that the Paul or radiofrequency (rf) trap commonly employed for trapping the ions limits the attainable temperatures in atom–ion mixtures, and the s-wave regime has so far not been reached in this system. This limitation stems from the oscillating electric fields employed in the rf trap, which causes the ion to perform a rapid micromotion. During an atom–ion collision energy may be transferred from
the time-dependent trapping field into the atom–ion system [16, 35–44]. Even in the limit of zero atomic temperature and an ion sitting at the rf node of the trapping field, the attractive atom–ion potential leads to a drag on the ion to a region of non-negligible rf amplitude during a collision. In fact, even runaway heating may occur when the atom is heavier than the ion. Cetina et al [38] calculated that the lowest temperatures may be achieved for atom–ion combinations with large ion to atom mass ratios. They theorize that Yb$^+$–Li, which has the largest mass ratio of any atom–ion combination allowing straightforward laser cooling, may enter the quantum regime after improving control over the trapping voltages to slightly beyond state-of-the-art to compensate excess micromotion.

In this article, we perform Monte-Carlo simulations of the classical scattering events between individual free $^6$Li atoms and $^{171}$Yb$^+$ ions trapped in a Paul trap. The simulation includes the attractive atom–ion potential as well as the full trapping potential of the ion and potential excess micromotion fields using realistic experimental parameters that we obtain from our experimental setup and from parameters reported in the literature. The atom’s initial velocity is drawn from a thermal distribution of given temperature, starting on a random position on sphere around the ion. The sphere size is chosen carefully to take into account the potentially large ion orbits and account for realistic atomic densities. We track the ionic motion continuously while each atom is introduced after the previous atom has escaped a second sphere around the ion again, defining the end of a collision. Thus no back-action on the atomic cloud is assumed, justified by the large experimental atom numbers on the order of typically $10^5$–$10^7$. Using this approach, we calculate that the s-wave regime of Yb$^+$–Li should be in reach with current technology and considering all known sources of excess micromotion in the ion. We further investigate the prospects of collisional cooling of single ions and crystals of ions into the motional ground state using a cloud of ultracold Li, taking into account experimental imperfections. We give a limit to the remaining number of motional quanta that can be expected and compute the cooling rate. Motivated by the prospects of an ultracold atom–ion system to form a solid-state emulator [24] we study the classical cooling dynamics for multiple trapped ions forming a Coulomb-crystal within the cloud of atoms and show that the cooling dynamics is very similar to that of a single trapped ion.

This article is organized as follows: first, we give the theoretical background of ion trapping and micromotion as well as the model for simulating buffer-gas cooling in section 2. In section 3, we describe the experimental parameters and limitations in our experimental setup. We use these parameters in the calculations of section 4, where we study the thermalization of a single trapped ion experiencing each type of micromotion. In sections 5 and 6 we describe the buffer-gas cooling of linear ion crystals, while section 7 describes the results for two-dimensional ion crystals. Finally, we draw conclusions in section 8.

2. Simulating buffer-gas cooled ions

2.1. Ion trapping in a linear quadrupole trap

The potential of a Paul trap as a function of ion position $\vec{r}$ can be written as:

$$\Phi(\vec{r}, t) = \sum_{i=1}^{3} \alpha_i r_i^2 + \frac{1}{2} \cos(\Omega_{rf} t) \sum_{i=1}^{3} \alpha'_i r_i^2$$

with the positive, geometry- and voltage-dependent prefactors $u_{dc}$ and $u_{rf}$ and trap drive frequency $\Omega_{rf}$. To describe a linear Paul trap as it is used in our experiment we have [45]

$$\alpha_1 = \alpha_2 = -\frac{1}{2} = -\alpha_3, \quad \alpha'_1 = -\alpha'_3 = 1, \quad \alpha'_2 = 0.$$ (2)

For this choice, the confinement along the 3-axis is supplied by a time-independent harmonic trapping potential $\propto u_{dc}$, whereas the radial confinement is supplied by the oscillating field $\propto u_{rf}$. Note that in reality the $\alpha_{1,2}$ coefficients are chosen to slightly differ from each other to lift the degeneracy in the resulting radial trap frequencies. The electric field is given by

$$\vec{E}(\vec{r}, t) = -\nabla \Phi(\vec{r}, t)$$

$$= -u_{dc} r_3 \hat{e}_3 - \frac{1}{2} (r_1 \hat{e}_1 + r_2 \hat{e}_2) - u_{rf} \cos(\Omega_{rf} t) (r_1 \hat{e}_1 - r_2 \hat{e}_2),$$

with the unit vectors $\hat{e}_i$ in the $i$th direction. With that, the equation of motion for a single ion with mass $m_{ion}$ and positive charge $+e$ can be written as the Mathieu equation [46]

$$\dot{r}_i + (a_i + 2q_i \cos(\Omega_{rf} t)) \frac{\Omega_{rf}^2}{4} r_i = 0,$$ (4)

where $i \in \{1, 2, 3\} \equiv \{x, y, z\}$ and the parameters

$$a_1 = a_2 = -\frac{1}{2} a_3 = -\frac{2 e u_{dc}}{m_{ion} \Omega_{rf}^2},$$

$$q_1 = -q_2 = 2 e u_{rf} \frac{m_{ion} \Omega_{rf}}{m_{ion} \Omega_{rf}}, \quad q_3 = 0,$$ (5)

which are the stability parameters of the Paul trap [45]. Usually, Paul traps are operated at a region where $|q_i|$, $q_i^2 \ll 1$, which can be achieved by properly choosing a suitable combination of $\Omega_{rf}$ and the static and rf electrode voltages $\propto u_{rf}$, $u_{dc}$. An approximate solution in first order in $q_i$ can then be obtained by

$$r_i(t) \approx r_i^{(1)}(\omega_i t + \phi) \left(1 + \frac{q_i}{2} \cos(\Omega_{rf} t)\right),$$

where the phase $\phi_i$ and amplitude $r_i^{(1)}$ are determined by the initial condition at $t = 0$. The motion consists of a low frequency part, oscillating with the secular frequency $\omega_i \approx \frac{1}{2} \Omega_{rf} \sqrt{|a_1| + \frac{1}{2} q_1^2}$, thus requiring $a_i + \frac{1}{2} q_i^2 > 0$ for a stable solution. In the two radial directions, the rf field drives the so-called micromotion that oscillates in phase with the rf drive and whose amplitude depends on the secular motion amplitude $q_i$-parameters. Note that in a real ion trap
imperfections in the electrode alignment can lead to a small rf field component also in the axial direction, effectively setting \( q_z = 0 \). By averaging over the secular oscillation period \( T = \frac{2\pi}{\omega} \), one can obtain the average kinetic energy in each coordinate,

\[
E_{\text{kin},i} = \frac{1}{2} m_{\text{ion}} \bar{\mathbf{r}}_i(t)^2 \bar{\omega}_i^2 \\
\approx \frac{1}{4} m_{\text{ion}} \bar{\mathbf{r}}_i(0)^2 \left( \bar{\omega}_i^2 + \frac{1}{8} q_i^2 \Omega^2 \right),
\]

where the assumption \( \Omega \gg \omega \) was used.

### 2.2. Excess micromotion

Besides the intrinsic micromotion of the ion caused by the radiofrequency drive, stray charges on the trap electrodes, imperfections of the trap assembly and electrical connection as well as finite-size effects can lead to various types of so-called excess micromotion [46] that affects the average kinetic energy of the ion and prevents reaching ultracold temperatures. Below, we will briefly describe the three different kinds of excess micromotion that occur in a linear Paul trap, and in section 3 we will describe how these can be detected and compensated in our experiment.

Stray electric fields \( E_{\text{rad}} \) in the radial direction may push the ions away from the rf null, where they experience the presence of the radiofrequency field even without any secular energy. This type of excess micromotion we will call radial micromotion. The modified Mathieu equation of the system including \( E_{\text{rad}} \) reads [46]

\[
\frac{\mathbf{r}}{t} + (a_i + 2q_i \cos(\Omega_{\text{rf}} t)) \frac{\Omega_{\text{rf}}}{4} \mathbf{r} = \frac{eE_{\text{rad},i}}{m_{\text{ion}}},
\]

with \( i \in \{x, y\} \approx \{1, 2\} \). To lowest order in \( q \), the solution is given by

\[
r_i(t) \approx (r_i(0) + r_i(1) \cos(\omega_i t + \phi_i)) \\
\times \left( 1 + \frac{q_i}{2} \cos(\Omega_{\text{rf}} t) \right),
\]

with the equilibrium position of the secular motion being shifted by \( r_i(0) \approx eE_{\text{rad},i} / (m_{\text{ion}} \omega_i^2) \). For both radial directions this additional shift leads to an energy

\[
E_{\text{emm},i} = \frac{1}{16} m_{\text{ion}} (q_i E_{\text{rad},i} \Omega_{\text{rf}})^2 \\
= \frac{4}{m_{\text{ion}}} \left( q_i E_{\text{rad},i} \Omega_{\text{rf}}^2 \right)^2 / 8 \omega_i^2,
\]

in first order. Typically this micromotion can be compensated by applying an external static electric field to cancel the stray field at the position of the ion. Note that a stray field component in axial direction only changes the ion’s axial equilibrium position, not the kinetic energy of the system.

Axial excess micromotion is mainly caused by the finite size of the trap leading to a radiofrequency pickup on the dc end caps. This pickup leads to an additional, position-independent, oscillating field with amplitude \( E_{\text{ax}} \) in axial direction that modifies the axial Mathieu equation to

\[
\frac{\mathbf{r}}{t} + a_z \mathbf{r} = \frac{eE_{\text{ax}} \cos(\Omega_{\text{rf}} t)}{m_{\text{ion}}},
\]

leading to the analytic solution of a driven harmonic oscillator,

\[
r_z(t) = \frac{eE_{\text{ax}} \cos(\Omega_{\text{rf}} t)}{m_{\text{ion}} (\omega_z^2 - \Omega_{\text{rf}}^2)} + r_z(1) \cos(\omega_z t + \phi_z),
\]

thus increasing the average kinetic energy by the term

\[
E_{\text{emm},z} = \frac{(eE_{\text{ax}} \Omega_{\text{rf}})^2}{4 m_{\text{ion}} (\omega_z^2 - \Omega_{\text{rf}}^2)^2}.
\]

While it is hard to minimize this pickup by trap design, it can be reduced by appropriate low-pass filters connected to the end cap electrodes or injecting an rf field with opposite phase at one of the end cap electrodes [47].

Phase- or quadrature micromotion [48] is caused by a phase difference \( \delta_{\text{rf}} \) between the radiofrequency voltages on the opposing rf-electrodes, e.g. in \( x \)-direction. The phase micromotion can be approximately described by an additional homogeneous oscillating field in the direction of the electrodes [46], \( \mathbf{E}_{\text{ph}} \approx \frac{1}{4} q_i m_{\text{ion}} \delta_{\text{rf}} \Omega_{\text{rf}}^2 R_{\text{trap}} \sin(\Omega_{\text{rf}} t) \mathbf{e}_x \), where \( R_{\text{trap}} \) is half the distance between the two rf-electrodes. The field leads to the modified Mathieu equation

\[
\frac{\mathbf{r}}{t} + (a_x + 2q_x \cos(\Omega_{\text{rf}} t)) \frac{\Omega_{\text{rf}}}{4} \mathbf{r} = \frac{1}{4} q_x R_{\text{trap}} \delta_{\text{rf}} \Omega_{\text{rf}}^2 \sin(\Omega_{\text{rf}} t),
\]

The solution in first order approximation then reads

\[
r_x(t) = r_x(1) \cos(\omega_x t + \phi_x) \left( 1 + \frac{1}{2} q_x \cos(\Omega_{\text{rf}} t) \right) \\
- \frac{1}{4} q_x R_{\text{trap}} \delta_{\text{rf}} \sin(\Omega_{\text{rf}} t),
\]

leading to an additional term in the average kinetic energy in the \( x \)-direction

\[
E_{\text{emm},x} = \frac{1}{64} m_{\text{ion}} (q_x R_{\text{trap}} \delta_{\text{rf}} \Omega_{\text{rf}})^2.
\]

Compensation of the quadrature micromotion is possible but technically challenging, for example by using two coherent rf drives with an adjustable phase difference between their respective outputs.

### 2.3. Modeling atom–ion collisions

We numerically simulate the classical atom–ion scattering process by launching individual atoms on a sphere of constant radius \( r_0 \) centered around the equilibrium position of the trapped ion, one at a time. The radius of the sphere is chosen to be large enough to allow for potentially large orbits of the ionic motion and to resemble realistic atomic densities. During the whole propagation, we simulate the full radiofrequency potential of the Paul trap, including potentially present excess micromotion. A collision event is finished after the atom has left a second sphere with a slightly bigger radius.
ions $B$, cell from the interval meaning that the velocity components at a given temperature
The initial velocity change, or to Langevin collisions where atom and ion are
where mainly the momentum direction of the partners slightly change, or to Langevin collisions where atom and ion are spiraling into each other, enabling for a large energy and momentum transfer. Langevin collisions occur when the impact parameter $b$ is less than the Langevin range $b_C = (2C_4/E_{col})^{1/4}$ [49]. Notably, the Langevin collision rate $\Gamma_C = 2\pi\rho_a\sqrt{C_4/\mu}$ is only dependent on the atomic density $\rho_a$ and the $C_4$ potential as well as the reduced mass $\mu$ of the two body system but not the collision energy $E_{col}$.

To fairly sample the flow of atoms, the atom launching coordinates are obtained from a uniform distribution on the sphere surface at the beginning of each collision event. To get a starting position, two points $p$ and $q$ are randomly picked from the interval [0, 1]. The azimuthal angle $\phi$ is then given by $\phi = 2\pi \cdot p$ and the polar angle $\theta = \arccos(2q - 1)$ [50], from which the Cartesian coordinates are derived,

\[ r_{a1} = r_0 \cos(\phi) \sin(\theta), \]
\[ r_{a2} = r_0 \sin(\phi) \sin(\theta), \]
\[ r_{a3} = r_0 \cos(\theta). \]

The initial velocity $\dot{v}_a$ of the atoms is then sampled from the probability distribution $P_b(\dot{v}_a, T_a)$ of the flux of thermal atoms

\[ \Phi(\dot{v}_a) = \rho_a 4\pi r_0^2 \hat{e}_r \cdot \dot{v}_a, \]

at a given temperature $T_a$ and density $\rho_a$ through the sphere,

\[ P_b(\dot{v}_a, T_a)d\dot{v}_a = \Phi(\dot{v}_a) \rho_a 4\pi r_0^2 2\pi(k_B T_a)^2e^{-\frac{m_2^2}{2m_1^2k_B T_a}v_a^2} \]
\[ = \frac{m_2^2}{2\pi(k_B T_a)^2}v_a e^{-\frac{m_2^2}{m_1^2k_B T_a}v_a^2} \]
\[ \times e^{-\frac{m_2^2}{m_1^2k_B T_a}v_a^2}f_{\dot{v}_a,r}d\dot{v}_a, \]

meaning that the velocity components $\dot{v}_{a,\perp}$ and $\dot{v}_{a,\parallel}$ tangential to the sphere surface are picked from one-dimensional Gaussian distributions with a standard deviation of $\sigma = \sqrt{k_B T_a/m_2}$ each, whereas the perpendicular velocity $\dot{v}_{a,\parallel}$ is picked from a Weibull-distribution with shape parameter $k = 2$ and scale parameter $\lambda = \sqrt{2k_B T_a/m_1}$. Only atoms flying towards the center of the sphere will have a chance to collide with the ions, therefore it is enforced that $v_{a,\parallel} < 0$. The Cartesian components of the velocity are then obtained by a coordinate transformation of the spherical components. We check the functionality of the random number generator in appendix A.

After the atom is introduced, the full trapped-ion-atom system is propagated forward in time by an adaptive step-size Runge–Kutta algorithm of fourth order [51], including the full radiofrequency ion trap potential and if desired additional of excess micromotion. The algorithm maintains a desired relative accuracy in each coordinate $p_{\text{tol}}$ in each coordinate as explained in appendix A. This allows for a fast propagation when atom and ion are far away from each other and an accurate propagation when the interaction is strong. At the end of a collision event, when the atom passes the second sphere of radius $r_1$, all ion’s coordinates at this time are intermediately stored as initial coordinates for the next collision event and the energy of the ion is determined. For this, the atom is removed from the simulation and the ion motion is propagated further for a fixed amount of time $t_{\text{ion}} \gg 2\pi/\omega_i$ using $N_{\text{ion}}$ fixed time steps of duration $\Delta t_{\text{ion}}$, sufficiently small to resolve micromotion. During this additional propagation all ion trajectories are stored. From the velocities $\dot{v}_{i,n}$ at each point in time the average kinetic energy

\[ E_{\text{kin}} = \frac{1}{N_{\text{ion}} \times 2} \sum_{k=1}^{N_{\text{ion}}} \sum_{n=1}^{N_{\text{ion}}} \dot{v}_{i,n}(t_k)^2 \]

is computed, which can be used to determine the collision energy in the relative coordinate frame of the two body atom–ion system. The expression contains both the energy of the secular harmonic oscillator motion of the ion as well as the micromotion energy, caused by intrinsic- and potentially present excess micromotion. Here, we defined the quantity kinetic temperature $T_{\text{kin}}$ that includes both micromotion and secular motion energy, thus it is technically not a temperature but just the average kinetic energy scaled with $3N_{\text{ions}}/2k_B$, whereas in literature the ion temperature often refers only to the secular motion. The decomposition into micromotion and vibrational energy will be discussed in section 5. After the energy of the ion has been determined in the simulation, the time and ion coordinates are reset to the stored values from the end of the collision event and a new atom is introduced.

2.4. Collision energy and s-wave limit

The $s$-wave limit of $^{171}\text{Yb}^+ / ^6\text{Li}$ is reached at a collision energy of [18]

\[ T_s = \frac{\hbar^2}{2k_B \mu^2 C_4} = 8.6 \, \mu\text{K}. \]

The collision energy is given by the energy in the relative atom–ion coordinate. In the experimentally relevant situation in which the ion has a much larger kinetic energy than the
3. Micromotion detection and compensation

The experimental setup is described in detail in [52]. The linear Paul trap is made out of four blade electrodes with a distance of $R_{\text{trap}} = 1.5 \text{ mm}$ to the trap center. End caps with a spacing of 10 mm are used to confine the ion along the axial ($z$) direction. Two sets of additional electrodes can be used for compensation of stray electric fields. Oscillating voltages at a frequency of $\Omega_{d} = 2\pi \times 2 \text{ MHz}$ and an amplitude of $V_{0} = 75 \text{ V}$ are applied to the blades and dc voltages of $V_{\text{dc}} \approx 15 \text{ V}$ to the end caps. This results in radial and axial trap frequencies of $\omega_{\text{rad}} \approx 2\pi \times 150 \text{ kHz}$ and $\omega_{\text{ax}} \approx 2\pi \times 42 \text{ kHz}$. Below, we describe how we detect and compensate micromotion in our setup and give limits on the attainable experimental parameters. More details on the micromotion detection and compensation can be found in [52].

A radial stray field component $E_{\text{rad}, d}$ leads not only to excess micromotion but also to a shift in equilibrium position as shown in figure 1.

Within the horizontal direction (1), this can be detected by tracking the ions position for different radial trap frequencies, shifting the ions position by $r_{h}^{(0)} \approx \frac{E_{\text{rad}, d}}{\omega_{\text{rad}}^{2}}$, for $\omega_{\text{rad}} \approx \omega_{h} = \omega_{b}$. We measure the shift on the ion by imaging from the top. We extract the ion’s horizontal position from averaging over five camera images at each radial trap frequency setting and fitting a Gaussian function. From these measurements we conclude that $E_{\text{rad}, h} < 0.5 \text{ V m}^{-1}$ under optimal circumstances.

The ion’s vertical position cannot be obtained with the camera as the imaging system and vacuum system was designed to only image the ions from the top. Instead, we use the magnetic field dependence of the $(^{2}\!\!\!\!\!_{3}S_{1/2}, F = 0, m_{F} = 0) \leftrightarrow (^{2}\!\!\!\!\!_{3}S_{1/2}, F = 1, m_{F} = 1)$ hyperfine splitting in $^{171}\text{Yb}^{+}$ [53] for a determination of position shifts as a function of trap frequency. To do so, we apply a vertical magnetic field gradient of $g_{v} = 0.15 \text{ T m}^{-1}$ as shown in figure 1, which leads to a frequency shift of 2.1 kHz for a determination of position shifts as a function of trap frequency.

By comparing the frequency shift at radial confinements of $\omega_{\text{rad}} = 2\pi \times 80 \text{ kHz}$ and $\omega_{\text{rad}} = 2\pi \times 230 \text{ kHz}$ using microwave Ramsey spectroscopy, we measure a dc electric field of $E_{\text{dc}} = 0.29(2) \text{ V m}^{-1}$ for 1 V applied to the compensation electrodes. As the set compensation voltages typically need to be adjusted by less than 1 V from day to day, we estimate that $E_{\text{rad}, v} < 0.3 \text{ V m}^{-1}$ starting at optimal compensation during an experimental run.

The axial micromotion is obtained by measuring the line broadening of the 4.2 MHz wide $^{2}\!\!\!\!\!_{3}D_{3/2} \leftrightarrow ^{2}\!\!\!\!\!_{3}D_{3/2}$ transition at $935 \text{ nm}$ wavelength in $^{171}\text{Yb}^{+}$ [53]. For this, we use a laser beam aligned along the trap axis [46, 52]. We obtain an upper bound to the amplitude of the oscillating electric field in the trap center of $E_{\text{ax}} \leq 15 \text{ V m}^{-1}$, limited by the observed linewidth of the $^{2}\!\!\!\!\!_{3}D_{3/2} \leftrightarrow ^{2}\!\!\!\!\!_{3}D_{3/2}$ transition at optimal compensation. By measuring the axial micromotion at various ion positions along the trap axis, we obtain $q_{i} = 0.0023$.

Aligning the beam under 45° with respect to the trap axis allows us to also check for quadrature micromotion, but none was detected. The observed transition linewidth results in the limit $b_{\text{eff}} < 0.65 \text{ mrad}$. Using a transition with a narrower linewidth, e.g. the 22 Hz wide $^{2}\!\!\!\!\!_{3}S_{1/2} \leftrightarrow ^{2}\!\!\!\!\!_{3}D_{3/2}$ clock transition at $411 \text{ nm}$ in $^{171}\text{Yb}^{+}$ [54], could improve these limits significantly.

4. A single ion in the cold buffer gas

In this section, we present the simulation results for collisions between a single trapped ion in a Paul trap using parameters that can be achieved with the ion trap used in our experiment. We investigate the influence of atomic bath temperature as well as the different kinds of micromotion on the ion’s average kinetic energy for realistic parameters. For simplicity, we start our calculations with an ion that has no energy and observe how this ion thermalizes with the atomic bath in a similar way as described in [16, 38]. Although chosen for convenience, this situation is also of experimental relevance, as the ion may be laser-cooled close to its ground state of motion before the atoms are introduced [16].
4.1. Influence of the atomic bath temperature

We simulated collisions for $T_a$ between 0 and 50 $\mu$K. The ion’s averaged kinetic energy after equilibration in units of $T_{\text{kin}}$ and typical $1/e$ number of collisions to equilibrate $N_{\text{col}}$ were determined by fitting an exponential function of the form

$$T(N_{\text{col}}) = (T_{\text{kin}} - T_0)(1 - e^{-N_{\text{col}}/\tau_{\text{eq}}}) + T_0,$$

(24)

to the results obtained by averaging at least 300 individual runs. The results are shown in figure 2. The errors given in the plot correspond to the standard errors of the fit parameters. The average kinetic energy of the ion (left) in units of $T_{\text{kin}}$ shows a strictly linear dependence with a slope of 1.79 (2) and offset of 7.60 (14) $\mu$K. The dashed line shows the hypothetical case in which each secular mode of the ion equilibrates with the temperature of the atom, according to the approximate prediction of equation (7). Its slope reads 1.68 using the trap parameters of the simulation. In particular, the deviation from unity slope is given by the extra energy stored in the micromotion amplitude, which is approximately $1/2 k_B T_a$ extra per radial direction [46], such that the energy of the atomic bath excites five degrees of freedom instead of three, explaining the slope of approximately 5/3. The deviation in slope of the simulated points with respect to the prediction is expected to be caused by the approximations made to obtain the prediction (i.e. $[\alpha]$ and $q_{\perp}^2 \ll 1$, see section 2.1). The offset can be seen as the direct influence of micromotion-induced heating, transferring energy from the trap drive rf field into the secular motion of the ions, mediated by the atoms. The number of collisions required to equilibrate (right) follows a square root function, which is to be expected, since the thermalization rate $\Gamma_{\text{eq}} = 1/N_{\text{col}}$ should be directly proportional to the fraction of events that lead to thermalization, namely Langevin collisions, divided by the number of total events, $\Gamma_{\text{eq}} \propto \frac{\Gamma_L}{\Phi(u)}$, with $\Gamma_L$ the Langevin rate and $\Phi(u)$ the flux into the sphere on which the atoms start as defined in equation (19). Thus, $N_{\text{col}} \propto \Phi(u)^{-1} \propto \sqrt{T_0}$. From the fit, we obtain a proportionality factor of $412(8)/\sqrt{\mu K}$.

4.2. Influence of radial excess micromotion

In this paragraph, we investigate the influence of radial excess micromotion caused by a stray electric field $E_{\text{rad}}$ on the average kinetic energy of a single ion when immersed in a cold atomic bath of 2 $\mu$K. We scanned $E_{\text{rad}}$ over a range of 0.0-0.6 V m$^{-1}$ and determined the ion’s average kinetic equilibrium energy in units of $T_{\text{kin}}$ and the typical number of collisions required to equilibrate $N_{\text{col}}$ according to equation (24) by averaging over at least 300 individual runs for each point. We additionally checked the influence of the radial direction of $E_{\text{rad}}$—the results are shown in figure 3. The temperatures (blue) were calculated using a radial electric field in x-direction only. The results were fit with a quadratic function (solid blue line),

$$T_{\text{kin}} = T_1 + \theta_{\text{rad}} E_{\text{rad}}^2,$$

(25)

leading to a quadratic rise factor of $\theta_{\text{rad}} = 2680(15)$ $\mu \text{K} (\text{V/m})^{-2}$. The dashed blue curve represents the approximate theoretical amount of kinetic energy due to the presence of excess micromotion, according to equation (10), with a quadratic rise factor of 2360 $\mu$K (V/m)$^{-2}$. Also shown is the average kinetic energy for an ion without an atomic bath present, initialized at zero temperature (red points) along with a quadratic fit (red dashed line). The difference between the solid blue curve and the dashed red curve corresponds approximately to the amount of energy stored in the intrinsic micromotion and the secular motion. The point at $E_{\text{rad}} = 0.3 \text{ V m}^{-1}$ was simulated once with a factor 10 smaller tolerance parameter $\beta_{\text{rad}}$ in the propagator to check for numerical errors. The values in orange were taken using a dc field with equal components $E_{\text{rad},x} = E_{\text{rad},y}$ in both radial directions, the values in green (behind the orange points) with a dc field with opposite components, $E_{\text{rad},x} = -E_{\text{rad},y}$, to check the influence of the direction of $E_{\text{rad}}$, showing no deviation from the fitted curve. The number of collisions required to equilibrate (right) seems to slightly decrease with increasing field amplitude.

4.3. Influence of axial micromotion

In this paragraph, we investigate the influence of a homogeneous oscillating electric field along the axial direction of the trap on the average kinetic energy of a single ion when immersed in a cold atomic cloud at 2 $\mu$K. We scanned the...
field amplitude $E_{ax}$ from 0 to 15 V m$^{-1}$ and determined the ion’s average kinetic equilibrium energy in units of $T_{kin}$ and the typical number of collisions required to equilibrate $N_{col}$ according to equation (24) by taking the average over at least 300 individual runs for each point. The results are shown in figure 4. The temperatures (blue) were fit with a quadratic function (solid blue curve),

$$T_{kin} = T_i + \theta_{E_{ax}} E_{ax}^2,$$  \hspace{1cm} (26)

leading to a quadratic rise factor of $\theta_{E_{ax}} = 7.44(3) \, \mu K/(V/m)^2$. The dashed blue curve represents the approximate theoretical amount of kinetic energy due to the axial oscillating electric field, according to equation (13), with a quadratic dependence of 6.92 $\mu K/(V/m)^2$, in agreement with the points in red, showing the average kinetic energy of a crystal at zero secular energy without atoms present.

Due to the large axial oscillation amplitudes at high values of $E_{ax}$, a fixed starting sphere causes the atoms to occasionally launch very close to the ion, thus introducing unrealistic jumps in the potential energy that can lead to unstable behavior. Therefore, the blue points were not obtained using a starting sphere with fixed origin at the ion’s equilibrium position, but a comoving sphere around the ion’s immediate position. As a consequence, there are events where the ion is moving away from the introduced atom such that the atom is immediately registered as having escaped, leading to an increased number of required collisions, (figure 4 right, blue points) as compared to the non-comoving case (green points). This effect seems to increase with field amplitude. A comoving starting sphere means that especially very slow atoms that would usually cause a Langevin collision are overseen. Therefore, the average contribution of the atom to the collision energy increases. Since the ion temperature in this regime is dominated by the micromotion energy anyways, this effect can be ignored.

### 4.4 Influence of quadrature micromotion

The effect of phase micromotion on the equilibrium average kinetic energy of a single ion in an atomic gas of 2 $\mu K$ is investigated. We scanned the phase difference $\delta\phi_{rf}$ from 0 to 0.65 mrad, corresponding to the expected experimental upper limit from the linewidth broadening measurement as discussed in 3. We determined the resulting equilibrium average kinetic energy in units of $T_{kin}$ as well as $N_{col}$ according to equation (24) by averaging over at least 300 individual runs per point. The results are shown in figure 5. The temperatures (blue) were fit with a quadratic function (solid blue line),

$$T_{kin} = T_i + \theta_{\delta\phi_{rf}} \delta\phi_{rf}^2,$$  \hspace{1cm} (27)

leading to a quadratic rise factor of $\theta_{\delta\phi_{rf}} = 3980(15) \, \mu K \, \text{mrad}^{-2}$. Also shown is the approximate theoretical amount of kinetic energy stored in the phase micromotion (dashed blue), according to equation (16), with a quadratic increase of $3 \, 651.2 \, \mu K \, \text{mrad}^{-2}$ for the parameters used in the simulation. As in the case for axial micromotion, the red points show the average kinetic energy of an ion without an atomic bath present, in agreement with the dashed blue line. All points of the plot were simulated using a comoving start and escape sphere for the atoms to prevent numerical instabilities, thus leading to an increasing number of collisions required to equilibrate (right).

### 4.5 Summary

We have seen that the intrinsic micromotion-induced heating of the ion in the simulated system leads to an ion temperature of $T_{kin} = 7.60(14) \, \mu K$ for the limit of $T = 0 \, \mu K$ and rises linearly with the atomic bath temperature as expected, redistributing the atomic bath temperature on the motional degrees of freedom of the ion. From this, a collision energy of $E_{col}/k_B = 0.38(1) \, \mu K$ can be deduced, much lower than the s-wave limit of $T_s = 8.6 \, \mu K$ for the system. From the excess micromotion scans, we obtain the same result when the excess micromotion is is set to 0 and otherwise a quadratic dependence of the scanned parameter, as expected. The quadratic rise is about 7.5%–13.6% larger than the intrinsic micromotion energy of an isolated ion, indicating additional heating due to the collisions with the much colder atoms. This effect is found to be strongest in the radial case and weakest in the axial case. We will discuss the accessibility of the desired values for each excess micromotion parameter in section 8.
5. Ion crystals

In this section, we briefly introduce the theoretical and numerical framework to describe the normal modes of oscillations in an ion crystal. We present and test a numerical method to extract the energy stored in the secular motion of each individual mode. The section is structured as follows.

First, we present the underlying physics and a method to numerically find the ion’s equilibrium positions. Starting from these positions, we expand the potential energy in second order and express the motion in a normal mode basis, diagonalizing the approximate potential energy. Last, we present a numerical method to extract the secular energy stored in each mode from the stored trajectories of each ion by Fourier transformation of the normal mode coordinates.

By treating the mutual Coulomb interaction of the ions as well as the trapping itself in harmonic approximation, the ion crystal can be described as a system of coupled harmonic oscillators. This system can be decomposed into normal mode coordinates and frequencies. This procedure is described in detail in e.g. [55] for a linear ion crystal. For a given set of secular trap frequencies and number of ions, the equation of motion reads

\[ F_n = m_{\text{ion}} \ddot{r}_n + e^2 f_{\text{D}} \sum_{m \neq n} \frac{\ddot{r}_n - \ddot{r}_m}{|\ddot{r}_n - \ddot{r}_m|^3}, \]  

where the first term describes the three-dimensional trapping of each ion with the trap frequency matrix \( \omega = \text{diag}(\omega_x, \omega_y, \omega_z) \) and the second term is the mutual Coulomb interaction of the ions. This system can be decomposed into normal mode oscillators. This system can be transformed numerically using a standard Cooley–Tukey fast Fourier transform algorithm [56]. The Fourier spectra of the normal coordinates then contain only a peak at the respective mode frequency along with peaks at the micro-motion sidebands. To obtain the energy stored in each mode, we compute the average kinetic energy of each normal coordinate \( q_m \),

\[ \bar{E}_{m, \text{tot}} = \frac{1}{2} m_{\text{ion}} \sum_{k=1}^{N_{\text{fit}}} \left| q_m(t_k) \right|^2, \]

where \( m \) is the mode index and \( k \) the time index of the Fourier time grid of spacing \( \Delta t_{\text{fit}} \). Since this energy still contains micromotion, we make use of the Fourier relation for time derivatives,

\[ (F q_m)(f) = -i 2 \pi f q_m(f), \]

where \( q_m(f) = (F q_m)(f) \) is the Fourier transform of the normal coordinate \( q_m(t) \), and Parseval’s theorem for the discrete Fourier transformation,

\[ \sum_{k=1}^{N_{\text{fit}}} \left| q_m(t_k) \right|^2 \Delta t_{\text{fit}} = \sum_{k=1}^{N_{\text{fit}}} \left| -i 2 \pi f_k q_m(f_k) \right|^2 \Delta f_{\text{fit}} \]

\[ = (2 \pi)^2 \Delta f_{\text{fit}} \sum_{k=1}^{N_{\text{fit}}} f_k^2 \bar{q}_{m}(f_k) \bar{q}_{m}^*(f_k), \]

with which we can replace the expectation value of the squared normal mode velocity \( q_m(t) \) and obtain

\[ E_{m, \text{tot}} = \frac{1}{2} m_{\text{ion}} (2 \pi)^2 \Delta f_{\text{fit}} \sum_{k=1}^{N_{\text{fit}}} f_k^2 \bar{q}_{m}(f_k) \bar{q}_{m}^*(f_k) \]

\[ = \frac{1}{k_B T_{\text{m}}}, \]

using the identity for the Fourier frequency grid spacing \( \Delta f_{\text{fit}} = (N_{\text{fit}} \Delta t_{\text{fit}})^{-1} \).
the micromotion frequency, the high frequency parts of the spectrum can be cut off easily by reducing the limit of the sum in equation (35) to a value $N_{\text{cut-off}} = f_{\text{sec}} / \Delta f_{\text{fin}}$, where $f_{\text{sec}}$ is the desired cut-off frequency. To obtain only the secular energy part for each of the modes $E_{m,\text{sec}}$, the cut-off frequency should be chosen centered between the highest normal mode frequency and the lowest micromotion sideband. We define the temperature of each mode by $T_{m,\text{sec}}$ and the total secular temperature as $T_{\text{sec}}$ as

$$E_{\text{sec}} = \sum_{m=1}^{N_{\text{ions}}} E_{m,\text{sec}} = \sum_{m=1}^{N_{\text{ions}}} \frac{1}{2} k_B T_{m,\text{sec}}$$

$$= \frac{3N_{\text{ions}}}{2} k_B T_{\text{sec}}. \quad (37)$$

The approximate eigenmode frequencies $f_{m}$ can be found by searching the peak position of the Fourier spectrum for the respective mode within an accuracy of the Fourier frequency grid size $\Delta f_{\text{fin}} = 1/(N_{\text{fin}} \Delta t_{\text{fin}})$ leading to a relative error typically on the order of $\frac{1}{2} \Delta f_{\text{fin}} / f_{m}$.

A typical spectrum of the Fourier amplitudes for a linear four-ion crystal is shown in figure 6. The Fourier spectra of all spatial coordinates (left) show each multiple peaks at the twelve different mode frequencies. The spectrum also contains the micromotion sidebands around the trap drive frequency of $f_{\text{trap}} = 2 \text{ MHz}$ and a possible cut-off value (gray bar) for the secular energy determination. While some of the peaks at around 130 kHz are too close to be distinguished, the Fourier spectra of the normal mode coordinates (right) show only one peak each, allowing for the numerical frequency and energy determination within each mode. Note that the plots are cut off at the relevant eigenmode frequency scale, not showing the micromotion sidebands around the trap drive frequency $f_{\text{trap}} = 2 \text{ MHz}$. The twelve normal modes of the four-ion crystal are visualized in figure C2 in appendix B, along with their respective frequencies obtained from the diagonalization of the secular case as presented in this section and the frequency peak positions of the Fourier spectra. Typically, these modes are assigned with the names given in the right column [57].

6. Ion crystals in the cold buffer gas

In this section, we investigate the influence of the number of ions as well as that of all types of micromotion in an ion crystal. We further analyze the case where an additional oscillating electric quadrupole field in axial direction is present, leading to a non-vanishing $q_e$-parameter, which is typically the case under realistic experimental conditions. In this section, we assume that the entire crystal is immersed in the atomic cloud, and each ion is equally likely to collide with an atom. In particular, we dice the ion at which the atom is introduced before calculating each collision event.

6.1. Influence of the number of ions

First, the influence on the achievable temperature of the crystal $T_{\text{kin}}$ (see equation (21)) and typical number of collisions required to equilibrate $N_{\text{col}}$ as defined in equation (24) was investigated. The results for one to six ions trapped using no axial or excess radial micromotion is shown in figure 7. For one and two ions at least 300 runs were averaged, whereas due to the computational effort for three to six ions, only 40 runs each were simulated, thus leading to worse statistics and thus larger errors. For the final temperature of the crystal (left) a weak dependence on number of ions can be observed. The results were fit with a heuristic fit function (blue line),

$$T_{\text{kin}} = T_{1} + \theta_{1}(N_{\text{ions}} - 1)^2, \quad (38)$$

leading to $T_{1} = 11.4(2) \mu \text{K}$ and a quadratic rise factor of $\theta_{1} = 0.17(2) \mu \text{K}$. The number of collisions required for thermalization (right) is strictly linear in number of ions. The linear fit (solid line) leads to an increase of 626(20) collisions per additional ion. The behavior is to be expected since the number of modes of the crystal that need to be cooled increases linearly as well. While in the simulation only one atom is introduced at a time, in the experiment the density of atoms ideally is the same all along the ion crystal, thus increasing the actual collision rate by the factor $N_{\text{ions}}$. Consequently, the thermalization time for an $N_{\text{ions}}$-crystal is expected to be the same as for one ion.

6.2. Influence of excess micromotion

Similar to the single ion case, the effect of radial excess micromotion as well as axial micromotion and quadrature
micromotion was investigated. Additionally, the dependence of the secular energy was studied. The obtained results can be found in appendix C. The behavior of the final average kinetic energy versus the scanned micromotion parameter is in perfect agreement with the single ion case.

6.3. Influence of a non-vanishing axial rf-gradient \((q_z \neq 0)\)

To study the effect of a non-vanishing \(q_z\), the parameter was scanned from 0 to \(5 \times 10^{-3}\). The value in our ion trap is around \(q_z^{opt} = 2.3 \times 10^{-3}\) for similar trapping parameters as used in the simulation. The resulting equilibrium Temperatures \(T_{kin}\) and \(T_{sec}\) are shown in figure 8 (blue). The points were obtained by averaging over at least 30 individual runs for each value of \(q_z\) and fitting the averages according to equation \((24)\). The results for the average kinetic energy (left) were fit using a quadratic function with offset (solid line), \(T_{kin}(q_z) = T_1 + \theta_q q_z^2\), leading to a quadratic rise factor of \(\theta_q = 8.29(6) \times 10^3 \mu K\) with offset \(T_1 = 13.0(6) \mu K\). The approximate theoretical dependence of the average kinetic energy according to equations \((9), (10)\) is shown as a dashed line. The quadratic rise of the theoretical curve is given by \(\theta_{theo} = 7.80 \times 10^3 \mu K\). The points in red show the average kinetic energies due to the influence of \(q_z\) in the non-interacting case where the ions were initialized without secular energy. A quadratic fit of the red points lead to a rise factor of \(\theta_q^{00} = 7.81(1) \times 10^3 \mu K\), in good agreement with the prediction from the approximate solution, which is to be expected as the approximation holds for \(q_z^2 \ll 1\). The secular temperature (right) shows an almost linear dependence on \(q_z\) and resembles the actual influence of the additional micromotion-induced heating due to a non-vanishing \(q_z\).

6.4. Micromotion-induced heating on the individual modes

In this section, we analyze the effect of each type of micromotion on the individual modes of a four-ion crystal. The secular temperature of each mode was obtained as described in section 5 from the simulations of the linear four-ion crystal in sections 6.2 and 6.3. The resulting temperatures for the twelve individual modes as presented in figure C2 are shown in figure 9 for radial excess micromotion (left top) axial micromotion (right top) and quadrature micromotion (left bottom). In each of the three cases the radial modes equilibrate to a slightly higher temperature than the axial modes, when the scanned excess micromotion parameter is low. For high values the temperature of the modes with excess micromotion dominate, which is the \(x\)-direction (red) for both radial and quadrature micromotion and the \(z\)-direction (black) in the case of axial micromotion. A further sub-separation of the radial and axial modes is not resolved.

Interestingly, for a non-vanishing axial gradient, expressed by \(q_z\), the situation is quite different, as it is shown in figure 9. In this case, the modes separate for high \(q_z\) into different groups, starting with the \(x\) and \(y\) zigzag modes (red and blue crossed circles) at the lowest temperature for \(q_z = 0.05\). The next group is formed by the \(x\) and \(y\) center-of-mass modes (red and blue squares) along with the drum modes (red and blue triangles) and the \(z\) anti-stretch mode (black triangles). Approximately located at average mode temperature the two tilt modes (red and blue circles) are found. At higher temperature, the three remaining axial modes, i.e. Egyptian (black crossed circles), center-of-mass (black squares) and stretch (black circles) are located. This behavior is mainly caused by the participation of the outer ions to these modes, since these can exchange the largest amount of energy during a collision due to their large micromotion amplitudes. While the contribution of the outer ion’s motion to the zigzag modes is lowest and the mode is moving perpendicular to the micromotion direction, the radial center-of-mass and drum modes show larger and equal coupling as indicated by the arrow length in the mode visualization insets and in figure C2. The anti-stretch mode shows less coupling strength for the outer ions but moves in the direction of micromotion, thus enhancing the probability for a high energy exchange within a collision. The strongest radial contribution of the outer ion’s motion is to the two tilt modes, leading to the highest radial mode temperatures. As in the case of an homogeneous oscillating axial field, the highest temperatures are found within axial modes, dominated by the one with the largest contribution of the outer ion’s motion, the stretch mode.

7. Two-dimensional ion crystals

By adjusting the axial and radial trapping fields, it is possible to change the shape and dimensionality to form two-dimensional ion crystals \([58–61]\). Even with perfect micromotion compensation, there are always ions within any nonlinear crystal that have their quasi-equilibrium position outside the radiofrequency node axis, thus experiencing a non-vanishing oscillating electric field, leading to additional, unavoidable micromotion. Therefore, immersing the complete ion crystal in a cloud of ultracold atoms will always lead to micromotion-induced heating of the normal modes. To avoid this effect, one can utilize the large spacing between the ions, enabling the experimental possibility to overlap a dense and small atomic cloud only with a single ion sitting at the axial radiofrequency node within a larger ion crystal.
To simulate a stable 7-ion hexagonal ion crystal, we change the trap parameters to $f_z = 95.459$ kHz, $q_x = q_y = 0.261$, and $\alpha_x = 1.0, \alpha_y = -2.0$ to achieve $f_r = 211.002$ kHz and $f_y = 127.229$ kHz as radial secular trap frequencies, all within experimental reach with the ion trap used in our experiment. Due to the stronger confinement in the $x$-direction, the crystal forms in the $y$–$z$ plane. Its geometry along with its approximate (secular) mode structure is depicted in figure 10. Notably, in contrast to a linear crystal, the mode with the highest frequencies are not center-of-mass modes, but the two planar blink modes, where the ion density oscillates in $y$ and $z$ direction respectively. Also the mode with the lowest frequency is not a center-of-mass mode but the $x$ rotate mode, where all six ions defining the hexagon oscillate in phase clockwise/counterclockwise around the central ion within the crystal plane.

To simulate the thermalization of the secular modes, we initialize the ion crystal with negligible secular energy by first switching on a strong velocity-dependent damping force as defined in equation (29) that is adiabatically turned to zero. To give the ion crystal an initial secular energy, we add to each ion’s velocity components a velocity sampled from a Maxwell–Boltzmann distribution at a given temperature before the first collision occurs. We only let the central ion collide with atoms at $T_a = 2 \mu$K. We obtain the secular temperatures for each mode as in the case for the linear ion crystal by integrating over the Fourier spectra of the normal mode coordinates. Due to the orders of magnitude larger micromotion sidebands around the trap frequency of 2 MHz, it is necessary to increase the frequency resolution by a factor of four and only take a narrow range around the respective peaks for the integrals into account. Otherwise, the integrals suffer from a non-negligible micromotion floor of the Fourier spectra even around the secular frequencies that can only be

---

**Figure 9.** Individual secular temperatures $T_{sec,m}$ of each normal mode of a linear four-ion crystal colliding with atoms at $T_a = 2 \mu$K in the case of a radial dc electric field $E_{rad}$ in $x$-direction (left top), a homogeneous axial oscillating field (right) with amplitude $E_{ax}$ (right top) or in the presence of a rf phase shift $\varphi_{rf}$ between the rf electrodes (left bottom). The results depicted in red and blue were obtained from the four modes oscillating in $x$- or $y$-direction respectively, whereas the results in black were obtained from the four axial modes. The plot on the lower right shows the behavior of the twelve secular mode temperatures for a non-vanishing $q_i$ parameter. The insets illustrate the respective modes, in which the arrows indicate the direction and relative amplitude of motion.

**Figure 10.** Visualization of the normal mode movement for a trapped planar seven-ion crystal. The arrows indicate the direction and amplitude of the respective mode within the plane (black) and perpendicular to the plane (red). For each mode the respective eigenfrequency $f_i$ obtained from diagonalization of the secular approximation is shown.
suppressed by further increasing the Fourier resolution towards unfeasible computational effort. To compensate for the already large increase in computation time due to the large micromotion amplitudes and increased number of particles compared to the four-ion linear crystal, the atom start sphere size was chosen to be fixed and only $r_0 = 0.3 \, \mu m$ around the central ion, thus increasing the likelihood of Langevin collisions but also cutting down the propagation times during a collision. The results for all 21 modes of a planar seven-ion crystal initialized at 25 $\mu K$ are shown in figure 11. The values were averaged over 120 individual runs. The thermalization of the modes can be classified into three different groups.

- The modes where the central ion’s motion is not participating at all do not show significant cooling dynamics (left), besides the $y$ drum (orange dashed) and $y$ wave (dark red dashed) mode, showing a relatively slow cooling and heating, possibly due to enhanced nonlinear Coulomb interactions between the ions in these two modes.
- The modes where the central ion participates rather weakly (right, black dashed), as indicated by the length of the vectors in figure 11, show a slow cooling dynamic over the observed number of collisions.
- The modes where the central ion participates most (right, solid red), $x$, $z$ blink, $z$ drop, $z$ pendulum and $x$ double rotation, thermalize the fastest.

The different initial temperatures of each mode are caused by the different coupling strength and number of modes each ion is involved in and could in principle be corrected for, but this is not necessary for the qualitative analysis of the behavior. Remarkably, the achieved minimum temperatures of the planar crystal $T_{\text{kin}} = 700 \, \mu K$ is five orders of magnitude larger due to the large micromotion amplitudes of the outer ions.

8. Conclusions

In this article we have presented numerical simulations of classical Yb$^+$/Li collisions for ions trapped in a Paul trap. We presented and tested a numerical framework to simulate and analyze the collisions using parameters that can be achieved in our experiment, including all types of micromotion that are observable in real ion traps. We analyzed the effect of the micromotion on the achievable average kinetic energy of a single ion. For an ion in an ideal Paul trap and in the limit where $T_a \to 0$, this energy is found to be at $T_{\text{kin}} = 7.60(14) \, \mu K$. Owing to the large mass ratio, this leads to a collision energy of $T_{\text{rel}} = 0.4 \, \mu K$ which lies well below the $s$-wave temperature limit. In this situation, the ion is cooled close to its ground state of motion with $\hat{n} = 1.2$ motional quanta remaining in the secular motion on average.

For the limits for all types of excess micromotion found in our experiment, the determined collision energies are a factor of 2–11 higher than the $s$-wave temperature limit, as it is shown in table 1. This indicates that better micromotion detection and compensation is required there. In particular, using a narrow linewidth laser would allow to put better limits on the axial and quadrature micromotion amplitudes. Another option may be to use the atoms themselves for accurate micromotion detection as described in [13].

The limits for each experimental parameter that lead to $s$-wave collisions energies are also given in table 1. Although all lie beyond the limits of our current setup, they are not excessive, as e.g. Härter et al [13] report a field of $E_{\text{rad}} \lesssim 0.02 V \, m^{-1}$ and $E_{\text{ax}} \lesssim 2.1 \, V \, m^{-1}$ in a similar system. For the quadrature micromotion, we expect the given experimental limit of $b_{\text{q,rf}} = 0.65 \, \text{mrad}$ to be overestimated by at least an order of magnitude due to the limitations of our detection techniques, as we show in section 3. The rf phase shift mainly results from unequal length of the connectors, which is approximately less than $\Delta \varphi_{\text{rf}} \approx 0.5 \, \text{mm}$. Thus, we expect a 2$\times$ phase mismatch on the order of $b_{\text{q,rf}} \approx \frac{\Delta \varphi_{\text{rf}}}{v_{\text{prop}}/2 \approx 0.04 \, \text{mrad}}$. We suggest an assumed signal propagation velocity of $v_{\text{prop}} \approx c_{\text{light}}/2$ half the speed of light. Similarly, we expect that the true axial micromotion amplitude lies significantly below the experimental limit stated. We conclude that Yb$^+$/Li may reach the quantum regime with state-of-the-art micromotion compensation. We note however that our present analysis is based on classical theory. For excellent micromotion compensation, a quantum description such as the one developed in [20] should be generalized to include excess micromotion and used to predict thermalization in the ultracold regime.

We found that a buffer-gas cooled linear ion crystal behaves similar as a single ion and the presence of more than three modes of ion motion does not significantly influence the achievable collision energies and thermalization rates. A non-vanishing axial gradient expressed as a $q_z$-parameter leads to a collision energy of $T_{\text{col}} = 26.3 \, \mu K$ for a four-ion crystal and the experimental value of $q_z^{\exp} = 0.0023$. Also shown in table 1 are the mean secular energies of the single ion and four-ion case along with the mean thermal occupation numbers for the mode with the lowest frequency (center-of-mass).

Within all simulations, we do not observe runaway heating of the ion, as expected, since the mass of the ion is much larger than the mass of the atom [35]. In the simulations it takes around $N_{\text{col}} \approx 550–600$ collisions for a single ion.
to equilibrate within an atomic cloud with a density of $\rho_s = \frac{1}{4 \mu_3 r_0^3} \approx 1.1 \times 10^{18} \text{ m}^{-3}$ (i.e. one atom within the interaction sphere at a time). Within a simulation run using a non-comoving sphere we observe an average flux $\Phi_n$ of 10,000 collisions within 120 ms propagation time, which translates into

$$\Gamma_{1 \text{col}} = 2\pi \rho_s \sqrt{\frac{C \mu_2}{\mu}} \frac{N_{\text{col}}}{\Phi_n} \approx 35-38.$$  

(39)

Langevin collisions that are required for reaching the equilibrium temperature. As only Langevin collisions lead to a significant change in momentum and energy of the colliding pairs [49], the back-action on the atomic cloud temperature in a real experiment with atom numbers typically on the order of $10^2 - 10^3$ can be safely neglected. Luckily, the chance for an inelastic collision happening during the interaction time, leading to charge transfer or molecule formation is less than 0.76% as we recently measured [52]. The cooling rate for a linear ion crystal is comparable to the single ion case, under the assumption of a homogeneous atomic density all along the ion crystal. Interestingly, the secular modes of a linear ion crystal equilibrate to slightly higher temperatures than average when moving in a micromotion direction.

We have shown that collisional cooling of a planar seven-ion crystal by a localized atomic cloud interacting with only the central ion should be possible. The technique enables cooling of all the ten modes where the colliding ion participates in. The achieved temperatures of these modes are all below 12 $\mu$K, corresponding to mode occupation numbers of $n_{\text{m}} = \frac{\hbar E_{\text{coll}}}{\hbar a_{\text{coll}}} = 2-11$ phonons. Shuttling the ion crystal to overlap one of the outer ions with a small atomic cloud at the position of optimal micromotion compensation should in principle increase the number of cooled modes up to 18 out of the 21 total modes. Such localized micro-clouds could be implemented by using a dimple trap as it is described in [62]. There, the atomic cloud is trapped by a strongly focused laser beam with a waist of $\leq 1.8 \mu$m, thus trapped in a volume much smaller than the interionic distance, e.g. $14.6 \mu$m for the ion crystal investigated in this article.

Our results show that with modest improvements in the functionality of the random number generation, the accuracy of the simulation algorithm can be achieved in our experiment, enabling buffer-gas cooling of the trapped ion system. The micromotion compensation and detection, reaching the quantum regime of atom-atom collisions, by using a dimple trap as it is described in [52]. The cooling rate for a linear ion crystal is comparable to the single ion case, under the assumption of a homogeneous atomic density all along the ion crystal.

### Acknowledgments

This work was supported by the EU via the ERC (Starting Grant 337638) and the Netherlands Organization for Scientific Research (NWO, Vidi Grant 680-47-538 and Start-up grant 740.018.008) (R.G.). We gratefully acknowledge fruitful discussions with Antonio Negretti.

### Appendix A. Reality checks of the simulation

In this section, we check the accuracy of the simulation algorithm in detail using realistic trapping fields that can be achieved in our experiment. A summary of the parameters used in the simulations unless noted otherwise can be found in table A1.

The functionality of the random number generation was checked by analysis of the distributions of initial atom coordinates for 10,000 events sampled at $T_a = 2 \mu$K on a sphere of $r_0 = 0.6 \mu$m. By definition, the spatial coordinates automatically lie on the sphere. It is therefore sufficient to check that each coordinate is uniformly distributed in the interval $[-r_0, r_0]$. For the velocities, the distributions of equation (20) must be obtained. As an example, distributions for $r_{a,i}$, $v_{a,i}$, and $v_{a,i}$ are shown in figure A1.

Having a method for the energy determination at hand, it is of importance to check the negligible influence of the start- and escape sphere sizes for the atoms. If the sphere radii are picked at the same order as the range of the atom–atom interaction, the immediate change in potential energy after the insertion and extraction of an atom leads to unrealistic kicks in the force on
the ion. To check the influence of the sphere radii on the ion temperature, the inner sphere radius \( r_0 \) was scanned between 0.2 and 1.8 \( \mu \)m. The thermalization of a single trapped ion initially left at rest with a thermal cloud of atoms at 2 \( \mu \)K was simulated. The outer sphere radius \( r_1 \) was chosen to be 0.5% bigger than \( r_0 \). An example for a thermalization curve (blue points) is shown in figure A2 (left). The curve was obtained by averaging over 656 individual runs and fitted with an exponential (see equation (24)) (black line) leading to an equilibrium temperature of \( T_{\text{kin}} = 11.4(1) \mu \)K on the characteristic timescale of \( N_{\text{col}} = 607(2) \) collisions, using \( T_0 = 0 \) as the initial ion’s temperature. The ion’s energy distribution after thermalization is shown in figure A2 (right). The blue points were obtained from all ion energies of the 656 runs between collision 5000 and 10 000 and fitted with a thermal distribution (red, dashed) leading to a temperature of 9.4(2) \( \mu \)K and a thermal distribution with fixed temperature (purple) obtained from the exponential fit (left). The ion’s energies deviates quite a bit from the thermal distributions, showing a longer tail towards high energies, which is a well known behavior [16, 39, 41, 43] caused by the additional kinetic energy due to the micromotion of the ion.

### Table A1. Parameters used for the numerical simulation of the atom–ion collisions, unless given otherwise in the text. If varied, the last column refers to the respective section where it is investigated.

<table>
<thead>
<tr>
<th>Param.</th>
<th>Value</th>
<th>Comment</th>
<th>Sections</th>
</tr>
</thead>
<tbody>
<tr>
<td>( f_c )</td>
<td>42.426 kHz</td>
<td>Axial trap freq.</td>
<td>—</td>
</tr>
<tr>
<td>( f_\text{ff} )</td>
<td>2 MHz</td>
<td>rf-drive freq.</td>
<td>—</td>
</tr>
<tr>
<td>( q_x )</td>
<td>0.219</td>
<td>rad. ( q )-param.</td>
<td>—</td>
</tr>
<tr>
<td>( q_y )</td>
<td>(-q_x + q_y)</td>
<td>rad. ( q )-param.</td>
<td>—</td>
</tr>
<tr>
<td>( q_z )</td>
<td>0</td>
<td>ax. ( q )-param.</td>
<td>6.3</td>
</tr>
<tr>
<td>( T_{\text{bc}}(0) )</td>
<td>0 ( \mu )K</td>
<td>init. temp.</td>
<td>—</td>
</tr>
<tr>
<td>( T_\text{a} )</td>
<td>2 ( \mu )K</td>
<td>Atomic bath temp.</td>
<td>4.1</td>
</tr>
<tr>
<td>( r_0 )</td>
<td>0.6 ( \mu )m</td>
<td>Launch</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( r_1 )</td>
<td>1.005 ( r_0 )</td>
<td>Escape sphere rad.</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( p_{\text{tol}} )</td>
<td>( 5 \times 10^{-10} )</td>
<td>rel. num. tol.</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( N_{\text{col}} )</td>
<td>2</td>
<td>Fourier grid size</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( \Delta t_{\text{ff}} )</td>
<td>50 ns</td>
<td>Fourier resolution</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( \Delta t_{\text{kin}} )</td>
<td>5 ns</td>
<td>Time grid for ( E_{\text{kin}} )</td>
<td>—</td>
</tr>
<tr>
<td>( C_\text{t} ) (m ( \text{m}^2 ))</td>
<td>( 5.607 \times 10^{-17} )</td>
<td>attr. int. coeff</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( C_\text{t} ) (m ( \text{m}^2 ))</td>
<td>( 5 \times 10^{-19} )</td>
<td>rep. int. coeff</td>
<td>Appendix A</td>
</tr>
<tr>
<td>( E_{\text{cob}} )</td>
<td>[ (0, 0, 0) \text{ V} \text{ m}^{-1} ]</td>
<td>dc offset field</td>
<td>4.2 and 6.2</td>
</tr>
<tr>
<td>( E_{\text{ax}} )</td>
<td>( 0 \text{ V} \text{ m}^{-1} )</td>
<td>Axial rf ampl.</td>
<td>4.3 and 6.2</td>
</tr>
<tr>
<td>( \delta \theta_{\text{M}} )</td>
<td>0 mrad</td>
<td>Phase mismatch</td>
<td>4.4 and 6.2</td>
</tr>
</tbody>
</table>

### Figure A1. Spatial (left) and velocity (right) distributions of atoms picked on a sphere with radius 0.6 \( \mu \)m and a temperature of 2 \( \mu \)K along with the expected probability densities (black).

### Figure A2. Average kinetic energy of an ion colliding with atoms at 2 \( \mu \)K averaged over 656 runs to obtain the ion’s temperature (left) from an exponential fit (black) leading to a value indicated by the dashed line and distribution of the ion’s energies in units of \( T_{\text{kin}} \) as defined in equation (21) after thermalization (right) along with a fitted thermal distribution (red, dashed) and a distribution where the temperature was fixed to the value obtained from the exponential fit (purple).

### Figure A3. Equilibrium temperature \( T_{\text{kin}} \) as defined in equation (21) (left) and characteristic number of collisions \( N_{\text{col}} \) (right) for an ion colliding with atoms at 2 \( \mu \)K versus the starting distance \( r_0 \) between atom and ion. While the equilibrium temperature does not depend on \( r_0 \) in the scanned regime, the characteristic number of collisions increases quadratically. The lines show a constant (left) and quadratic fit (right).

The final temperatures and characteristic number of collisions \( N_{\text{col}} \) required for equilibration for the different starting radii are shown in figure A3 and were obtained using the exponential fit model given by equation (24). For each point, at least 300 runs were averaged. The equilibrium temperature \( T_{\text{kin}} \) of the ion shows no dependence on the starting sphere size \( r_0 \), whereas \( N_{\text{col}} \) shows a quadratic behavior over the scanned range. This behavior can be qualitatively explained by the nature of Langevin collisions. For a given collision energy \( E_{\text{cob}} \) every atom with an impact parameter smaller than \( b_0 = (2C_\text{t}/E_{\text{col}})^{1/4} \) undergoes a Langevin collision and can therefore cause a large energy and momentum transfer that contributes to the thermalization process. The fraction of atoms that undergo a Langevin collision \( P_L \) and therefore fly into the solid angle element defined by \( b_0 \) is then given by \( P_L = (1 - \cos(b_0/r_0)) \approx 1/2(b_0/r_0)^2 \). For an increasing \( r_0 \) this automatically demands for a quadratic increase in the required number of total collisions to equilibrate. Unless noted otherwise, \( r_0 = 0.6 \mu \)m is used in all further simulations as a trade-off between simulation time and realistic atomic densities (see e.g. [63]). Demanding only one atom at a time inside the sphere around one of the ions results in a density of \( \rho_n = 1 / (4/3\pi r_0^3 N_{\text{col}}) < 1.1 \times 10^{18} \text{ m}^{-3} \).

To realistically model the atom–ion interaction, one needs to check as well that the temperature of the ion does not strongly depend on the choice of the hard-core radius parameter \( C_\text{t} \) as introduced in equation (17). In reality, a repulsive
The red points were obtained using a higher numerical precision as a relative accuracy parameter for the size of the time steps in order to stay below a given tolerance. The points were obtained using a higher numerical precision as explained in the text.

Barrier is expected to be at a distance, where the electronic wavefunctions of the atom and ion begin to significantly overlap, typically in the range of hundreds of picometers to a few nanometers. The parameter $C_0$ was therefore scanned in a range between $5 \times 10^{-14}$ and $5 \times 10^{-21}$ m$^2$, effectively varying the position of the classical turning point $r_{hc} = \sqrt{2C_0}$ between 0.1 nm and 316 nm. The results for both final ion temperature $T_{kin}$ and collisions required for equilibration $N_{col}$ are shown in figure A4. The points were obtained by averaging the ions’ average kinetic energy over at least 300 runs and fitting it according to equation (24). For a broad range of barrier radii the final temperature of the ion remains at the same level. For values bigger than $r_{hc} = 10$ nm the potential is more and more dominated by the repulsive term proportional to $C_0$, preventing Langevin collisions and therefore the ion from micromotion-induced heating as we describe it in our work [44], where a repulsive barrier is utilized to prevent exactly this heating mechanism. For the smallest value of $r_{hc} = 0.1$ nm, the ion temperature seems to be a factor of 1.5 higher than in the regime between 0.3 and 10 nm, which can be explained by numerical errors due to the increasing steepness of the hard-core barrier for low values of $r_{hc}$, leading to large changes in acceleration in a hard-core collision. Therefore, this point was simulated again with a five times smaller tolerance in the adaptive step-size Runge–Kutta propagator, leading to the red points, in agreement with the values for larger $r_{hc}$. The number of collisions required for thermalization $N_{col}$ seems to first slightly decrease for higher values of $r_{hc}$ but shows a dramatic increase by around a factor of two at $r_{hc} = 77$ nm. Note that at this point the potential energy minimum caused by the attractive $C_4$-term of the potential becomes comparable to the collision energy, dominated by the atom temperature of 2 $\mu$K. Therefore, the immediately released kinetic energy during a Langevin collision becomes negligible. For even higher values of $r_{hc}$ the thermalization process speeds up again due to the quadratically increasing geometric cross section for repulsive collisions. For all further simulations, $r_{hc} = 1$ nm is used, which is around three times larger than the classical turning point of the Li–Yb$^+$ system [29, 52] but still produces similar results with less numerical effort due to the weaker forces involved.

During propagation, the Runge–Kutta propagator adjusts the size of the time steps in order to stay below a given relative accuracy parameter $p_{tol}$. It therefore propagates the system once by a full time step and once by two half time steps and compares the relative difference in propagated coordinates between both methods. If the maximum relative difference between one of the coordinates (including velocity) is bigger than the desired tolerance, the propagation step is repeated using an adjusted time step. To ensure a sufficiently small tolerance $p_{tol}$, further tests were performed. Firstly, the allowed tolerance was scanned from $p_{tol} = 10^{-15}$ to $10^{-12}$ as a parameter for the propagation of a single ion starting at a randomly chosen kinetic energy sampled from a thermal distribution at $T_{kin} = 13$ $\mu$K, leading to $E_{kin}/k_B = 20.5$ $\mu$K in the presented case. The trajectories including the velocities for the individual runs were stored to compute the relative deviation in kinetic energy for each tolerance with the one from the smallest value, $p_{tol} = 10^{-15}$,

$$\delta E_{kin}(p_{tol}) = \left| \frac{E_{kin}(p_{tol}) - E_{kin}(10^{-15})}{E_{kin}(10^{-15})} \right|.$$ (A.1)

Because collisions with atoms can cause a dramatically different change in trajectory for each tolerance, no atoms were introduced in this test. The ions were propagated for 120 ms, a timescale that typically corresponds to 10 000 collisions in the simulation. Due to the large amount of data, the trajectories were stored only during the last millisecond of propagation.

The resulting relative deviations $\delta E_{kin}(p_{tol})$ are shown in figure A5 (left). Due to the adaptive step-size algorithm, it is not possible to have the trajectories for each tolerance stored at the exact same time steps each, therefore the kinetic energy $\delta E_{kin}(10^{-15})$ was interpolated using cubic polynomials to match the time grid of the other tolerances, possibly leading to a small amount of interpolation noise. For clarity, only the values for $p_{tol} = 10^{-6}$ (green), $10^{-10}$ (blue) and $10^{-14}$ (red) are shown along with their time averages (straight lines). In figure A5 (right) the time averaged deviations for the other values of $p_{tol}$ are shown, approximately following an exponential behavior (solid line) with exponent $n \approx 0.78$. While for a tolerance of $p_{tol} = 10^{-6}$ the time averaged relative deviation is $< 11\%$, $p_{tol} = 10^{-8}$ delivers acceptable values of $\langle \delta E_{kin}(p_{tol}) \rangle \leq 0.2\%$ already.

Similar to the tests for $C_0$ and $r_{hc}$, also the influence of the tolerance parameter $p_{tol}$ on the final ion temperature $T_{kin}$ and required collisions $N_{col}$ to equilibrate was investigated. The results are shown in figure A6. Each point was obtained from taking the average of $E_{kin}$ over at least 300 individual runs and fitting the curves according to equation (24). Both observables do not change significantly from $p_{tol} = 10^{-12}$ to $10^{-6}$, only the point at $p_{tol} = 10^{-5}$ shows a dramatic increase in both $T_{kin}$ and $N_{col}$ due to increasing numerical errors. For all further simulations $p_{tol} = 10^{-10}$ is used (unless noted otherwise) as a trade-off between precision and computational effort.

A final check for both energy conservation of the propagator during collisions as well as physical behavior of the system is to investigate the secular case, where the time-
of collisions required to equilibrate exponential scanned tolerances are shown on the right, along with an along with the time averages (lines). The time averages for the other scanned tolerances are shown on the right, along with an exponential fit.

Figure A6. Final ion temperature $T_{\text{kin}}$ (left) and characteristic number of collisions required to equilibrate $N_{\text{coll}}$ (right) for an ion colliding with atoms at $2 \mu K$ versus tolerance parameter $p_{\text{tol}}$ used in the adaptive step-size propagator. The inset shows a magnified version of the plot from $p_{\text{tol}} = 10^{-12}$ to $10^{-6}$.

dependent trapping potential of the Paul trap is replaced by a 3D harmonic oscillator potential with the secular trap frequencies of the Paul trap. From a thermodynamic point of view, the ion should then thermalize to the same temperature as the atomic bath and the total energy during each collision should be conserved since no micromotion energy can be transferred to the secular oscillation. The resulting thermalization curve, averaged over 608 individual runs along with a histogram of the energy distribution is shown in figure A7. The histogram was taken from all points between collision 3000 and 5000 and is in perfect agreement with a thermal distribution (solid line) at $2 \mu K$, the same temperature as the atomic bath. Also the exponential fit of the thermalization curve (left) leads to the same value, thus indicating a correct physical behavior of the numerical model.

To finally investigate the energy conservation of the collisions, the energy transfer between atom and ion in each collision was investigated by comparing the atom and ion energies before and after a collision, at the points in time $t_0$ when an atom is introduced on the sphere with radius $r_0$ with the point in time $t_1$ when that atom escapes the sphere defined by $r_1$. The energy transfer on an ion trapped in the harmonic oscillator potential is shown in figure A8 (right), taken from one of the 608 individual runs from the simulation used for figure A7 (left). The plot shows the ion’s energy transfer $\Delta E_{\text{ion}} = E_{\text{ion}}(t) - E_{\text{ion}}(t_0)$ for each collision and ranges on scales limited by the atom energies. For the atom, a corresponding curve can be obtained. In figure A8 (right) the level of energy conservation $|\Delta E_{\text{ion}} + \Delta E_{\text{atom}}|$ is shown. The averaged error in total energy in each step is less than 0.12 nK (dark blue line) and therefore negligible on the typical energy scales of the simulations. Note that this error is mainly caused by the sudden but tiny jump in potential energy when the atom is introduced and extracted. With reasonable effort this could be corrected in the energy determination and atom injection scheme, but is only of interest for much higher densities and lower temperatures that may anyways require a quantum mechanical treatment. We therefore conclude that the employed propagator produces physical results with reasonable precision.

Appendix B. Reality checks of the Fourier method

In this section, we check the accuracy of the presented Fourier method for determining the average kinetic energy of an ion crystal. Unless stated otherwise, we use a linear chain of four ions at around $100 \mu K$ and let them thermalize by collisions with a cloud of atoms at $2 \mu K$.

To test the Fourier analysis method for obtaining the temperature of an ion crystal, we compare the temperature $T_{\text{fit}}$ of equation (36) with the temperature obtained from the average kinetic energy $T_{\text{kin}}$ (equation (21)) as shown in figure B1. For a step-size of $\Delta t_{\text{fit}} = 50 \text{ ns}$, sufficient to resolve frequency components of up to $f_{\text{max}} = 20 \text{ MHz}$, there is no significant improvement when increasing the number of steps from 16384 (red) to 32768 (black), the relative deviation from $T_{\text{fit}}$ to $T_{\text{kin}}$ is at around 2.5% on average over a broad range of temperatures. This leads to the conclusion that a frequency resolution of $\Delta f_{\text{fit}} = 1/(N_{\text{fft}} \Delta t_{\text{fit}}) \approx 1.2 \text{ kHz}$ is a good choice.
Figure B1. Average kinetic energy of a four-ion crystal colliding with thermal atoms at 2 μK (left) obtained by the Fourier method (see equation (35), (36)). The ions start at an initial temperature of around 100 μK. The Fourier spectra were obtained at different \( N_{\text{fft}} \) and a constant \( \Delta t_{\text{fft}} = 50 \) ns, thus effectively varying the frequency spacing. As a reference, the temperature obtained from \( E_{\text{kin}} \) (see equation (21)) is shown (yellow), averaging over 8 ns in steps of 5 ns. The curves for \( N_{\text{fft}} = 16384 \) (red) and 32768 (black) steps are almost on top of each other, as it can be also seen in the relative deviation from \( T_{\text{kin}} \) (right).

Figure B2. Average kinetic energy of a four-ion crystal colliding with atoms at 2 μK (left) obtained by the Fourier method for several combinations of Fourier grid sizes \( N_{\text{fft}} \) and grid spacings \( \Delta t_{\text{fft}} \), resembling a variation of the maximally resolvable frequency \( f_{\text{max}} \). All curves besides for \( N_{\text{fft}} = 2048 \) (gray) lie on top of each other, deviating from the average kinetic energy \( T_{\text{kin}} \) by less than 5% (right).

To find a sufficient number of grid points while leaving the frequency resolution constant, we vary \( \Delta t_{\text{fft}} \) inversely with \( N_{\text{fft}} \), as shown in figure B2. For all combinations with \( \Delta t_{\text{fft}} \leq 200 \) ns, the relative deviation from \( T_{\text{kin}} \) is approximately the same. At \( \Delta t_{\text{fft}} = 400 \) ns (gray) the maximum resolvable frequency is \( f_{\text{max}} = 2.5 \) MHz, being too close to the micromotion sidebands at around \( f_{\text{side}} = 2.0 \) MHz and therefore leading to a much lower energy, dominated by only the low frequency parts. To be on the safe side, we chose the combination \( \Delta t_{\text{fft}} = 50 \) ns and \( N_{\text{fft}} = 16384 \) for our system.

While during the collision processes very fast dynamics demanding for an adaptive step-size algorithm may occur, the fastest timescale during the temperature determination is set by the micromotion oscillation at \( f_{\text{side}} = 2 \) MHz in the case for \( q < 1 \). Therefore, a fixed step-size propagator with \( \Delta t_{\text{kin}} \ll 1/f_{\text{side}} \) is sufficient. In order to save computation time, we use the same fixed step-size propagator for the Fourier transformation energy determination as for obtaining the average kinetic energy. We therefore choose the time grid to be integer subdivisions of the Fourier grid, \( \Delta t_{\text{kin}} = \Delta t_{\text{fft}}/n, n \in \mathbb{N} \). To find a sufficiently small \( \Delta t_{\text{kin}} \) to resolve the micromotion oscillations at \( f_{\text{side}} = 2 \) MHz, we compare the kinetic temperature as defined in equation (21) for different propagation time steps \( \Delta t_{\text{kin}} \) with the temperature obtained using the smallest step size 2.5 ns as a reference. For time steps up to 40 ns we obtain relative deviations of less than 0.05% from the kinetic energy derived using time steps of 2.5 ns when averaged over 8 ns propagation time over the whole temperature range. To be on the safe side, we chose \( \Delta t_{\text{kin}} = \Delta t_{\text{fft}}/10 = 5 \) ns.

Appendix C. Excess micromotion in a linear four-ion crystal

The obtained results for average kinetic energy and secular energy are shown in figure C1. Each point was fit by averaging over at least 30 individual runs. The resulting average kinetic energies expressed as \( T_{\text{kin}} \) (left) follow approximately the same quadratic behavior as in the case of a single ion, indicating that the main part of the kinetic energy is stored in the micromotion. The quadratic fits (solid blue lines) lead to the increase parameters \( \theta_{\text{rel}} = 7.45(3) \) μK (V/m)\(^{-2} \), \( \theta_{\text{rel}} = 2705(15) \) μK (V/m)\(^{-2} \) and \( \theta_{\text{rel}} = 4005(16) \) μK mrad\(^{-2} \), in almost perfect agreement with the single ion case. For all three cases the theoretical average excess energies due to the micromotion is shown as dashed blue lines. To verify the validity of these

Figure C1. Average kinetic energy of a linear four-ion crystal colliding with atoms at 2 μK (left, blue) expressed as \( T_{\text{kin}} \) and part of the average kinetic energy is shown on the right. The insets show the difference between the solid and dashed blue curves, resembling the micromotion-induced heating. Green points were chosen over the whole temperature range. To be on the safe side, we chose \( \Delta t_{\text{kin}} = \Delta t_{\text{fft}}/10 = 5 \) ns.
curves, the average kinetic energy for a crystal without atoms, initialized at zero secular temperature was simulated as well (red points). Only for the case of radial excess micromotion the theoretical prediction (dashed blue) deviates significantly from the red points, indicating the approximate nature of the prediction at high radial micromotion amplitudes.

To quantify the micromotion-induced heating effect on the secular motion, the secular temperature was extracted as described in section 5. In all three cases the dependence on the scanned parameter seems to be a bit weaker than quadratic. The temperature dependence of the individual modes is discussed in section 6.4. The modes and their respective secular frequencies are depicted in figure C2.

In all three cases, the number of collisions required to equilibrate (not shown in the figures) show a similar behavior as in the single ion case, besides the fact that they are $N_{\text{ions}} = 4$ times higher because of the reduced effective density of atoms.

**References**


**Figure C2.** Visualization of the normal mode movement for a trapped linear four-ion crystal in descending order with respect to the eigenmode frequency. The arrows indicate the direction and amplitude of the respective mode. The modes are shown along with their respective eigenfrequencies $ff_t^{(n)}$ obtained from the diagonalization of the secular approximation and the values $ff_q^{(n)}$ obtained numerically from Fourier analysis of the mode spectra. Typical names of the modes are shown on the right column. The center-of-mass (c.o.m.) modes represent the upper and lower limit of the frequencies.

**ORCID iDs**

H A Fürst https://orcid.org/0000-0002-6811-5248