The decay of ion bunches in the self-bunching mode

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Abstract. The properties of ion bunches stored in an electrostatic ion beam trap (EIBT) have been investigated using the Cryogenic Trap for Fast ion beams (CTF). The extremely high vacuum used rendered the main ion loss mechanism, namely collisions with the rest gas, negligible. Aluminum dimer anions were photo-detached by a pulsed laser to measure the longitudinal ion distribution in the bunch, which for the first time revealed the presence of a dc ion beam component co-existing with the oscillating ion bunch after several hundreds of revolutions. Bunches stabilized by the so-called self-bunching mode of operation have been observed for times as long as 12 s (a factor of 100 longer than previous room-temperature experiments) using N₂⁺ and Al₂⁻ bunches at 6–7.1 keV beam energies after which the bunch abruptly decayed. The decay of the bunch was observed to be intensity dependent and is well reproduced by a model that includes the expansion of the bunch along the beam axis, intrabeam scattering and collisional losses between the bunch and the dc component. Radio-frequency bunching of the ions resulted in the extension of the bunch observation time to 600 s, placing upper limits on all other EIBT ion bunch and trap losses as well as supporting the newly developed decay model and EIBT-adapted bunch dynamics.

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1. Introduction

In the extensive field of ion trapping, the electrostatic ion beam trap (EIBT) [1], which is also known as a multi-pass time-of-flight (TOF) mass spectrometer [2, 3], is one of the most recent developments. These traps only use electrostatic fields, functioning on a principle quite similar to that of an optical resonator. Numerous traps such as these have been built in recent years, hosting a wide variety of experiments in cluster physics [4–10], molecular physics [11–13] and electron collision studies [14]. Experiments probing larger clusters and other heavy species exploit the mass invariance of ion trajectories in these electrostatic storage devices. Recent experiments have also used non-destructive methods in the form of capacitive electrodes to observe ion bunches and perform Fourier analysis [15] or a comb-sampling method [16] to probe the mass resolution of such devices. Nuclear physics experiments also employ EIBTs as mass separators [3, 17–19] as an EIBT’s resolving power linearly increases with observation time [20]. These experiments depend on small TOF differences, which accumulate over many revolutions in the trap, allowing the separation of ions with different masses. Such methods could be further developed via a more detailed understanding of bunch dynamics inside EIBTs. A recent proposal [21] has also suggested the use of short, stable ion bunches inside an EIBT to carry out fundamental interaction studies of the electron–neutrino correlations in the $\beta$ decay of $^6$He. The properties of ion bunches inside these traps are, therefore, not only of technical interest, but also have applications in atomic, molecular, nuclear and particle physics, as well as astrophysics.

The ability to keep a group of identically charged particles together in a coherent bunch without external intervention, despite the given velocity distribution from the ion source and the Coulomb repulsion force between the ions, has already been observed in both room-temperature EIBTs [15, 22–24] and storage rings [25]. In EIBTs, this can be achieved by ensuring that the trap potential in the ion reflection region is shallow (corresponding to a negative $\eta_E$
(22, 26, 27)), which ensures that higher-energy ions travel longer paths than slightly lower-energy ions, thus keeping the bunch with a finite energy distribution together over many trap revolutions. All the experiments presented here employ the so-called self-bunching trapping mode, also referred to as the negative mass instability [27], in an EIBT to maintain stable bunches.

First, the longitudinal ion bunch profile is examined by photo-detaching aluminum dimer anions, revealing the presence of a dc ion beam component simultaneously stored with the bunch.

Next, the amplitude of the bunch measured at a fixed frequency as a function of storage time is then investigated, extending the previously reported durations of around 90 ms [24] to about 12 s using the extremely high vacuum of the Cryogenic electrostatic Trap for Fast ion beams (CTF). This has allowed us to experimentally observe the decay of the bunch for the first time, which was not observable in previous experiments due to the loss of the beam itself by collisions with residual gas. The longer bunch observation time greatly increased our sensitivity to the ion bunching properties and trapping stability of EIBT-type devices. Aspects of storage ring physics are adapted to describe the EIBT dynamics of the ions within the bunch. A model is then developed to reproduce the decay, which is dependent on the expansion of the bunch, in combination with intrabeam scattering and a loss term attributed to collisions between the stored bunch and the dc beam component. Our model predicts a strong dependence on the initial bunch length, in agreement with our bunch decay measurements.

Finally, radio-frequency (RF) bunching measurements further extend the observed duration of the bunch until the trap was turned off 600 s after ion injection. The bunch decay model is used to examine the decay, demonstrating that bunch expansion is, as expected, almost entirely prevented. The results are also in agreement with the expected suppression of the dc beam component and the resulting decrease in bunch loss. A reduction in the magnitude of intrabeam scattering is also observed, in agreement with the EIBT-adapted bunch dynamics.

2. Bunching in an electrostatic ion beam trap

Two different trapping modes exist for EIBTs: normal and self-bunching. When an ion bunch is injected and stored in normal mode, the bunch disperses within a short time due to the Coulomb repulsion force between the ions, the source-dependent kinetic energy distribution and the differences in the populated ion orbital trajectories. In contrast, the same bunch using the so-called self-bunching mode will be preserved over thousands of trap oscillations, exhibiting only minimal bunch-length expansion as a function of time. The physical difference lies in the utilized mirror potentials (in particular the gradient near the reflection region), which determines the trap’s slip factor $\eta_E$. This causes slightly higher energetic ions to travel longer paths within the EIBT, while slightly slower ions are decelerated earlier. The resulting negative frequency dispersion counteracts the effects of velocity and longitudinal position spread as well as space-charge repulsion, and hence ensures that the bunch remains coherent.

The self-bunching trapping mode in EIBTs has been studied in depth both experimentally [15, 22, 23, 28, 29] and theoretically [26–32] by measuring, for example, the bunch dispersion rate or simulating the shape of the bunch throughout its movement inside an EIBT. This self-bunching effect, while relatively new to the ion-trapping field of physics, was first introduced as the negative mass instability in 1959 for relativistic circular accelerators and storage rings [33]. An introduction to the extensive experimental and theoretical treatment of
this effect for such larger storage devices was given by Laclare [25]. For both EIBTs and storage rings, the $\eta_E$ parameter has to be negative in order to achieve self-bunching.

To describe self-bunching inside an EIBT, it is instructive to derive the $\eta_E$ parameter. The oscillation frequency $f$ of the ions in the trap is described by $f = \frac{\bar{v}}{T}$, where $\bar{v}$ is the time-averaged velocity which accounts for the deceleration and reflection of the ions in the mirrors and $L$ is the orbit length (twice the distance between the turning points). It follows that the frequency variation $\Delta f$ can be expressed as

$$\frac{\Delta f}{f} = \frac{\Delta \bar{v}}{\bar{v}} - \frac{\Delta L}{L}. \quad (1)$$

At a fixed beam energy, the path length variation $\Delta L$ is a function of the trap potential at the reflection points. At the point of reflection $x_r$ for an energy $E_0$, the change in the orbit length due to an energy shift $\Delta E$ can be linearly approximated as

$$\Delta L = 4 \frac{1}{q} \frac{\partial \phi(x_r)}{\partial x} \Delta E, \quad (2)$$

where $q$ is the ion charge and $\partial \phi(x_r)/\partial x$ is the gradient of the potential at the point $x_r$. The slip factor $\eta_E$ has a strong influence on the temporal development of an initially well-localized cloud of ions or ion bunch. Depending on whether $\eta_E$ is positive or negative, the bunch will disperse or retain its initial longitudinal shape, respectively. Combining $\eta_E$ with equation (1) yields

$$\eta_E = \frac{\Delta f/f_0}{\Delta E/E_0} = \frac{\Delta \bar{v}/\bar{v}_0}{\Delta E/E_0} - \frac{\Delta L/L_0}{\Delta E/E_0}. \quad (3)$$

Note that when $\eta_E = 0$, the frequency variation $\Delta f$ has to vanish as well, meaning that ions with any kinetic energy $E$ (close enough to $E_0$ that the linear approximation in equation (2) holds) will have the same oscillation frequency; thus the bunch will also retain its initial shape. Ions with higher energy must, therefore, travel a longer path as shown in equation (2). From equations (2) and (3), it is clear that $\eta_E$ becomes negative as the gradient approaches smaller values (corresponding to a shallow potential slope).

Since some EIBT papers on self-bunching [24, 27, 28] have defined the slip factor $\eta_T$ as a function of momentum and oscillation period, it is useful to note the conversion factor. Combining $f = 1/T$ and $E_0 = \frac{p_0^2}{2m}$ where $\Delta f/f_0 = -\Delta T/T_0$ and $\Delta E/E_0 = 2\Delta p/p_0$, the slip factor $\eta_E$ can be expressed with this change of variables as

$$\eta_E = -\frac{\Delta T/T_0}{2\Delta p/p_0} = -\eta_T/2. \quad (4)$$

Consider now the coupling of an RF voltage into an EIBT, and the alteration of EIBT bunching behavior. Storage ring ion bunching physics can be further adapted to EIBTs by considering that an ion revolution in an EIBT can be reduced to a standard single gap problem by recognizing that the entry and exit of an ion from the pickup electrode consists of two single gap passes. Note that the term single gap is commonly used in accelerator/storage ring physics and refers to a gap between two electrodes where RF is applied to an electrode to facilitate particle bunching, acceleration or deceleration. Since an ion enters and exits the pickup twice per revolution period $T$, this is equivalent to passing through four single gaps, allowing one to apply the concepts from storage ring physics with an appropriate transit time factor $T_t$ to account for the additional gaps. The change in ion energy $\Delta E$ with respect to the so-called synchronous
particle located by definition in the center of the bunch with the average ion velocity can, therefore, be given as

$$\Delta E(\Delta t) = Q U T_t(v) \sin(\omega \Delta t),$$

(5)

where $Q$ is the total charge of an ion, $U$ is the RF voltage applied to the electrode, $\Delta t$ is the time shift between an individual particle and the synchronous particle, $\omega = 2\pi / T$ and $T_t$ is the transit time factor, which is dependent on the ion velocity $v$. While it is possible to apply the RF to either the pickup or one of the trap electrodes, it is more convenient to apply the RF to a trap electrode and observe the ion bunch with the pickup electrode. The location chosen will only effect the magnitude of the transit time factor and thereby the required voltage amplitude.

Now consider the longitudinal bunch phase space shown in figure 2. Since the synchronous particle is located by definition in the center of the bunch, all phase differences in the longitudinal projection of phase space (i.e. energy and time displacement) are described with respect to this position. The separatrix is the line describing the largest possible phase differences from the synchronous particle in which the bunched ions still oscillate within closed, stable orbits. The so-called bucket refers to the entire phase space contained within the separatrix. Over many hundreds of trap oscillations, individual ions are expected to travel along the paths shown, oscillating in position from the front to the back of the bunch. Ions with energy differences or time displacements larger than that of the separatrix will no longer be contained within the bunch even if the phase space acceptance of the trap still permits them to be stored. Note that the direction of ion oscillation around the synchronous particle shown in figure 2 is valid only for $\eta_E > 0$ (normal trapping mode) and ignores the effects of space charge. The orbits of bunched ions in an EIBT operating with $\eta_E < 0$ oscillate in the opposite direction (see equations (3) and (5)). The maximum width of the bunch is one trap oscillation $T$, which defines the maximum width of the bunch phase space. The maximum bucket height scales as $\Delta E_{\text{max}} \propto \sqrt{U}$, where $U$ is the RF voltage applied to either the pickup or a trap electrode. The space charge of the ion beam, however, modifies the bucket height and is a function of $\eta_E$. We expect that for $\eta_E > 0$, the maximum bucket height $\Delta E_{\text{max}}$ is reduced by the ion bunch space charge, whereas for $\eta_E < 0$, the maximum bucket height is increased. This agrees with existing EIBT observations demonstrating that bunches are not stable when $\eta_E > 0$, but stable without the application of RF voltage when $\eta_E < 0$ [15, 22, 23, 28]. The ion bunch space charge in the self-bunching mode defines the non-zero bucket height needed for bunch stability. It logically follows that for positive $\eta_E$, the application of RF is required for the existence of stable bunches as the RF must overcome the threshold given by the space charge of the ion bunch.

The determination of the transit time factor $T_t$ as well as the exact relationship describing the dependences of the bucket height is beyond the scope of this paper. The actual ion phase space paths (in particular at larger phase differences) as well as the separatrix most likely differ from those plotted in figure 2. Detailed simulations, which include the bunch space charge, and possible RF voltage are required for a more accurate picture of the EIBT separatrix, in particular for such long ion bunches. The preceding cursory discussion does, however, give an intuitive understanding of EIBT bunching. As the number of ions increases, the resulting larger space charge will expand the bucket (for $\eta_E < 0$), permitting more ions to be stored within the bunch. The stable bunch phase space can be further expanded in the self-bunching mode by applying an RF voltage at the cost of increasing the energy width of the stored ion bunch. Alternatively, stable bunch phase space can be created for positive $\eta_E$ values with the application of RF. The phase space of the trap, however, places an upper limit on the effectiveness of RF bunching, as some ions will no longer be trapped if the energy width becomes too large.
Figure 1. A schematic cross section of the CTF electrostatic mirror electrodes used for trapping the ions. The pickup electrode used to non-destructively observe the oscillating ion bunch as well as the perpendicularly crossed pulsed laser beam are also indicated. The potential along the trap axis that was employed in self-bunching mode is plotted below for cations with an injected kinetic energy of 6 keV.

3. Experimental setup

The bunching experiments were performed using the CTF (see figure 1), which is a liquid-helium-cooled linear EIBT mounted inside a 3 m long cryogenic vacuum chamber assembly [8]. Injected ions are trapped between two electrostatic mirror stacks, each of which employs an einzel lens to confine the ions in the radial direction. Rest gas densities of 2000 particles per cm${^3}$, equivalent to a pressure of $8 \times 10^{-14}$ mbar at room temperature, have already been demonstrated during cryogenic operation using 1.8 K liquid helium [8]. The excellent vacuum dramatically reduces the main mechanism of ion loss from the trap, namely collisions with residual gas, extending observed beam lifetimes to over 300 s for keV ions. The unique ability to cryogenically cool the CTF vacuum system and trap electrodes allowed the first investigations of cluster anion decay in an EIBT as a function of ambient temperature [10].

Two different ion sources were used in the experiments described in this paper. A Penning ion source was used to produce $N_2^+$ ions, while a cesium sputter source was used to produce $Al_2^-$ anions. The aluminum dimer was chosen as it was produced in large quantities and could be readily detached by our laser. A beam chopper with a chicane in the ion trajectory created by four electrostatic deflectors was employed, where a variable-length ion pulse was created by switching one of the deflectors. Low-level mass selection to prevent the trapping of heavier aluminum clusters or atomic ions was achieved by setting the trap closing time and exploiting the TOF differences between different masses along the injection beam line. The measurement
Figure 2. Schematic diagram of the bunch phase space inside an EIBT for \( \eta_E > 0 \), ignoring the effects of space charge based on equation (5). All ions in the bunch must exist and travel along the closed orbits within the stable region of bunch phase space. The synchronous particle is located in the center with which all other orbits as well as phase differences (\( \Delta E \) and \( \Delta t \)) are defined. The outer limits of bunch phase space are defined by the separatrix and the bucket refers to the entire phase space contained within the separatrix. See text for more details.

The procedure consisted of injecting the ions and trapping them by quickly applying the high-voltage potentials to the entry-side electrostatic mirror (mirror 1 in figure 1) after the ions were inside the trapping region. The trap potentials were set to self-bunching mode, corresponding to the axial potential plotted in figure 1, where electrodes A, C and H are grounded. The employed \( \eta_E \) was determined to be \( \approx -0.2 \) by varying the ion source kinetic energy while measuring the oscillation frequency of the ions (see equation (3)).

Observation and mass-identification of the trapped ion bunch was achieved by measuring the image charge induced on a cylindrical pickup electrode located slightly off the geometric center of the trap. This signal was read out using a low-noise FET amplifier (NF SA-220F5), providing a gain of 200, and observed using a 500 MHz oscilloscope (LeCroy LC374A) and a spectrum analyzer (Advantest R3131A). While the oscilloscope sampled the pickup signal in the time domain with high time resolution, its maximum observation time of a single injection was limited. Conversely, the spectrum analyzer was capable of observing the bunch amplitude in the frequency domain (typically at the second harmonic of the ion’s oscillation frequency) throughout the entire injection cycle. It was also possible to observe the neutrals escaping from the trap by counting signals from a micro-channel plate (MCP) located at room temperature behind the trap electrodes along the beam axis. Such signal pulses were amplified, discriminated, and a personal computer equipped with a timing card (National Instruments PCI-6602) was used to record each event and the corresponding time after the injection sequence had been started. The vacuum chamber temperatures were measured with an array of sensors, where the chambers containing the ion trap were cooled to temperatures below 15 K.

In combination with the aluminum dimer anions, a laser setup was installed permitting the photo-detachment of the negative ions. A pulsed Nd:YAG laser was used to pump a dye laser to produce ~10 ns pulses with an energy of 6 mJ per pulse at a wavelength of 600 nm and a repetition frequency of 10 Hz. The laser beam was expanded in a telescope and then guided via a series of mirrors through the middle of the trap, perpendicularly crossing the ion beam axis. The laser shots were synchronized with the data acquisition system and the measured jitter between
the injection timing and laser activation was less than 10 ns, thus ensuring a reproducible ion bunch location for each laser shot in each injection cycle.

The CTF trapping efficiency was determined to be approximately 12% by comparing the amplitude of the injected and stored bunch signals on the pickup, which are proportional to the amount of current passing through it. We suspect that this value is primarily limited by a radiation shield inside the beam tube, which is installed between the cryogenic and warm vacuum chambers to reduce the amount of 300 K radiation entering the cryogenic region. This tube also partially obstructs the injected ion beam, limiting the initial ion trap phase space that can be accessed and thereby reducing the trapping efficiency.

4. Longitudinal ion bunch profile

The CTF’s crossed laser and ion beam geometry permitted the study of the longitudinal ion bunch profile, revealing new aspects of the self-bunching mode. A stable 2.1 µA (±5%) Al\textsubscript{2}\textsuperscript{−} beam was chopped into bunches by applying a 3.3 µs long rectangular pulse to the deflectors of the beam chopper. This chopper pulse width corresponds to the minimum width required for ion bunch production and trapping in the current setup (already at a width of 3.2 µs, no bunch signal was visible). Due to the finite chopper switching times, it is very likely that the initial ion bunch length was approximately 100 ns. The fast dispersion of this very short ion bunch in comparison to the following section can most likely be explained by the much shorter bunch length and correspondingly low bunch charge (small bucket size) as shown and discussed using the model below. The ions were stored for 300 ms with an injected kinetic energy of 6.0 keV. The laser beam was then used to photo-detach the Al\textsubscript{2}\textsuperscript{−} ions (which have a 1.50 eV electron affinity in the ground state [34]) at a well-defined delay after ion injection via the trigger setup described above. TOF mass selection was used to ensure that only aluminum dimer anions were stored, as confirmed by the TOF between the laser pulse and the observed neutral particles on the MCP. The delay between the closing of the trap and the first laser shot was increased in 1 µs steps, covering just over one complete trap oscillation, allowing the longitudinal ion density to be probed. Only neutrals that were heading towards the MCP (as opposed to the trap injection side) after laser excitation were observed. In the absence of the stored ions, no discernable signal was seen, confirming that cross-talk from the laser was negligible. The laser was operated at a repetition frequency of 10 Hz, corresponding to three laser pulses per storage cycle.

The neutral counts on the MCP as a function of time after ion injection are shown in figure 3. The pulse of neutral particles formed in the injection beam line due to residual gas collisions is followed by the decay of highly excited Al\textsubscript{2}\textsuperscript{−} and the collisional background rate between the stored ions and the residual gas in the trap. The neutrals from the photo-detached aluminum dimers are seen 1.4 ms after ion injection as a point far above the background. The height of this peak varied as the laser delay with respect to ion injection was changed. A maximum was observed if the ion bunch was at the laser crossing at the time of the laser pulse. Such maxima were found at laser delay times spaced by the Al\textsubscript{2}\textsuperscript{−} oscillation period $T$ of about 12 µs. When an additional delay of 7 µs was introduced, corresponding to approximately half an oscillation period, a much lower signal rate due to missing the bunch was observed (see blue triangles in figure 3). A close examination, however, shows that this signal rate is almost a factor of 4 larger than our collisional background due to residual gas, indicating a non-zero ion density at this time. By scanning the delay between the trap closing time and the
Figure 3. Two sample neutral decay rate measurements of $\text{Al}_2^-$ employing different laser delays around 1.4 ms after injection, causing the laser to strike or miss the ion bunch. Neutral fragments from the injection beam line are seen initially, followed by the decay of excited aluminum dimers and ending with the collisional background in the trap. A non-zero count rate (significantly above the background) due to the presence of a dc ion beam was also observed.

laser pulse train, the longitudinal bunch profile was measured as shown by black squares in figure 4. A bunch with well-defined flanks and a width of 2.6 $\mu$s (full-width at half-maximum, FWHM) is clearly visible with a count rate more than an order of magnitude larger than the rate in the time between the oscillating bunch. Due to the laser’s repetition frequency, a second shot 100 ms later (see the red circles in figure 4) demonstrated that the bunch at this point had completely dispersed, homogeneously filling the entire longitudinal phase space of the trap, resulting in a nearly constant signal rate at all times. The third shot (not shown) occurred 201.4 ms after ion injection and confirmed the homogeneous distribution observed in the second shot by also yielding a constant signal rate, albeit at a reduced count rate (factor of 10) due to the depopulation of the stored ions by the previous laser shots and the gradual change in the rovibrational population of the ions.

The observed ion bunch length is shorter than the employed beam chopper duration primarily due to the finite switching time of the chopper power supplies. The changing electric fields within the chopper and the corresponding variation in ion trajectories could also result in a mismatch between the beam emittance and phase acceptance of the trap for portions of the chopped ion bunch. Since the injected bunch does not have a dc component, the non-zero signal amplitude between the ion bunch peaks strongly suggests that in just over 100 trap revolutions, a small portion of the injected bunch uniformly filled the remaining longitudinal phase space of the trap. Based on the relative intensity of the signals, this dc component has approximately 4% of the bunch’s peak charge density. An integral of the counts over one oscillation demonstrates that less than 20% of the total stored ion charge is in this dc component. The corresponding bunch loss rate and explanation for the fast decay of this short bunch is discussed using longer ion bunches in the following section.
Figure 4. Average neutral count rate per injection of photo-detached 6.0 keV Al$_2^-$ versus laser delay after ion injection (black squares), showing the longitudinal profile of the ion bunch. A second laser scan probed the bunch profile 100 ms later (red circles) when the bunch had already dispersed, filling the entire trap. Lines are drawn to guide the eyes and sample statistical errors are plotted at 1.391 and 1.401 ms delays.

5. Bunch decay

5.1. Experimental parameters

In this section, we describe measurements of the pickup signal amplitude due to the image charge induced by the stored ion bunch as a function of the storage time. To observe this signal, the spectrum analyzer was set to measure a fixed frequency corresponding to the second harmonic ($h = 2$) of the ion oscillation frequency. This frequency provided the strongest signal amplitude due to the ions passing through the pickup twice per oscillation period. The trap was set to self-bunching mode as described above and short N$_2^+$ and Al$_2^-$ ion bunches given by the experimental parameters in table 1 were observed.

5.2. Results

The N$_2^+$ bunch amplitude measured at a fixed frequency is plotted as a function of time after injection in figure 5(a). Multiple ion injections with various initial intensities have been plotted together, all demonstrating a similar bunch decay pattern. The observed duration of the bunch was extended by over two orders of magnitude to $\approx$ 12 s in comparison with previously published observations of 90–100 ms [23, 24]. This corresponds to extending the number of trap revolutions from tens of thousands to about 1.5 million. This effectively demonstrates that EIBT-type devices with extremely low residual gas densities can provide stable bunch storage over an ion flight path of at least 1000 km without external RF fields. After a maximum time of 12 s, however, the ion bunch rapidly decays. The corresponding ion beam lifetime in the trap, which was measured by counting the neutral fragments with an MCP behind the trap, was
Table 1. Relevant experimental parameters: the injected kinetic ion energy $E_{\text{kin}}$, initial bunch width $x_0$, bunch oscillation period $T$, ion velocity $v$ and atomic mass number $A$.

| Species | $E_{\text{kin}}$ (keV) | $x_0$ ($\mu$s) | $T$ ($\mu$s) | $v$ (m $\mu$s$^{-1}$) | $A$ |
|---------|------------------------|----------------|-------------|-----------------|-----|
| Al$^-_2$ | 6.0                    | 4.05(2)        | 12.30(1)    | 0.15            | 54  |
| N$^+_2$  | 7.1                    | 2.05(2)        | 8.12(1)     | 0.22            | 28  |

Figure 5. Pickup signal amplitude at 246 kHz ($\hbar = 2$) of trapped N$^+_2$ ion bunches versus (a) injection time and (b) time after bunch decay. Multiple background-subtracted injections are superimposed.

over 300 s [8]. A comparison between the beam lifetime and the much shorter bunch decay time scale indicates that a mechanism independent of residual gas-induced ion loss must lead to the decay of the bunch. The bunch duration is shown to be dependent on the initial pickup amplitude and repeatedly demonstrates a similar decay shape. This is shown in figure 5(b) by setting the origin to the time when the bunch amplitude is just above the background rate. Higher intensity bunches are additionally observed to systematically exhibit non-exponential losses at early times. Since the bunch decays at roughly the same pickup amplitude throughout all injections, it is likely that a minimum amount of charge is required for bunch stability. These measurements thus provide the first experimental support for the idea that stable bunches require a minimum ion density [26, 31].

Measurements using Al$^-_2$, despite differences in the kinetic energy, bunch length and mass, yielded qualitatively similar bunch decays. A difference was observed, however, in the duration of the stable bunch with the anion bunches dispersing after somewhat shorter time periods as shown in figure 6. Here, the frequency amplitude (measured at twice the oscillation frequency of the ions) as well as a decay model (see below) are plotted as a function of time after injection. As discussed later, the longer initial bunch length of the anions is likely responsible for the observed change in bunch duration.

The initial length of the trapped ion pulse was measured during each injection using the pickup electrode and an oscilloscope. The results are summarized in table 1. The charge difference of the two ion species is not expected to influence the bunching dynamics. The error in the data points is indicated by the representative error bars in figure 6 and is given by the relative fluctuations between neighboring data points before the data were re-binned for clarity.
Figure 6. Sample superimposed, background-subtracted pickup signal amplitudes from multiple injections measured at twice the oscillation frequency of the ions ((a) 7.1 keV N$_2^+$; (b) 6.0 keV Al$_2^-$) in the trap as a function of time after ion injection. The ion bunch decays have been fitted with equation (14). Sample error bars are shown at later times.

These fluctuations were constant throughout the entire measurement at a level of 0.008 and 0.02 mV for N$_2^+$ and Al$_2^-$, respectively.

5.3. Bunch expansion

Our model is based on the common assumption that the longitudinal charge distribution in the bunch, and thereby the pickup signal, can be approximated by a rectangular distribution (similar to the measured experimental distribution shown in figure 4). This distribution is normalized where $A_0$ is the product of the total bunch charge and the calibration factor of the pickup system. A rectangular charge distribution was chosen to facilitate the calculation and fitting of the data, although a parabola-shaped distribution yielded similar results. As the bunch is observed to have completely dispersed after about 10 s, the bunch must expand along the beam axis with respect to time. A second-order Taylor expansion has been used to approximate the functional relation of $x$ and $t$ such that the bunch width $x(t)$ is given by

$$x(t) = x_0 + \beta t + \beta_2 t^2,$$

where $x_0$ is the initial bunch length, and $\beta$ and $\beta_2$ are the linear and quadratic expansion coefficients, respectively. A Fourier transformation converts the bunch signal amplitude into the frequency domain in order to compare the measured spectrum analyzer amplitude at a given frequency with the model amplitude $\hat{I}_h(t)$,

$$\hat{I}_h(t) = \frac{4}{T} \int_{t_0-x(t)/2}^{t_0+x(t)/2} \frac{A_0}{x(t)} \cos(h \omega t) \, dt,$$

where $h$ refers to the measured harmonic of the ion oscillation frequency (i.e. $h = 2$ is twice the oscillation frequency of the ions in the trap), $T$ is the bunch oscillation period and $\omega = 2\pi / T$. Note that the Fourier transform is multiplied by two since the bunch passes through the pickup twice per oscillation period and $x(t)$ is treated as a constant in this integral as it changes very
slowly with respect to the measured frequency. The simplified resulting pickup amplitude is given by
\[ \hat{I}_2(t) = \frac{A}{x(t)} \sin(x(t)\omega), \] (8)
where the new constant \( A \) is still proportional to the total charge and is given by
\[ A = \frac{4A_0}{h\pi} \cos(h\tau_0\omega). \] (9)

5.4. Particle loss from the bunch

The observation of non-exponential, intensity-dependent ion loss from the bunch at times immediately after injection, which is exhibited in the injections with the highest pickup amplitudes, strongly suggests the presence of intrabeam scattering. Intrabeam scattering refers to the process during which ions in the stored beam collide with each other, resulting in the expansion of the ion cloud (increase in the populated phase space). While particles in the bunch oscillate around the synchronous particle in longitudinal phase space, particles outside this phase space will be lost from the bunch. This does not, however, necessitate that the ions are lost from the trap as demonstrated by the dc beam component discussed throughout our results. They will only be lost from the trap if the collisions force the particles out of the trap’s acceptance. Intrabeam scattering is important for higher ion densities, which may occur in EIBTs near the turning points in the mirrors when the ion velocity is very small and the beam is compressed longitudinally, or along the field-free region if a particularly intense beam is stored. The rate for this loss process scales with the square of the number of ions. Due to the complex contributions of multiple ion-loss processes such as residual gas collisions at the relevant time scales in a room-temperature EIBT, this particular loss process has, however, not yet been clearly studied in an EIBT. Assuming that this process is present, the change in the number of ions in the bunch \( \dot{N}(t) \) is given by
\[ \dot{N}(t) = -k N(t) - \alpha (N(t))^2, \] (10)
\[ k = k_c + k_s + k_{dc} + k_x, \] (11)

where \( \alpha \) parameterizes the intrabeam scattering losses and is dependent on the bucket height described in section 2 in addition to the ion mass and velocity, while \( k \) incorporates all other exponential bunch loss processes. The sign of the species’ charge is not expected to affect \( \alpha \). Note that the contributions from collisional detachment \( k_c \) and, to a lesser extent, multiple scattering \( k_s \) by residual gas typically dominate in EIBTs at room temperature where trap pressures are about \( 10^{-8}–10^{-10} \) mbar. During CTF cryogenic operation, however, both of these terms are significantly suppressed. The presented data, therefore, permit the investigation of other normally negligible processes such as collisions between the ion bunch and the stored dc beam \( k_{dc} \), and possibly other unknown loss mechanisms \( k_x \) become important. Collisions between the stored dc beam and the bunch are assumed to scale with the product of the two separate populations \((N_0-N(t))N(t)\). Since collisional losses from the trap, which are given by \( k_c \ll 1 \), occur on much longer time scales (>300 s) than the bunch duration (<12 s), the total
number of ions stored in the trap can be assumed to remain constant. Note that the quadratic portion of this loss term has been combined into $\alpha$. Solving this differential equation leads to

$$N(t) = N_0/[e^{\alpha t} - c_1 k^{-1} + c_1 k^{-1} e^{\alpha t}],$$

where $c_1 = \alpha N_0$.

### 5.5. Bunch decay model

Since intrabeam scattering reduces the number of ions in the bunch, this is expected to only affect the total bunch charge and not its length $x$. Substitution of equation (12) in the amplitude $A$ of equation (8), therefore, yields the following formula to reproduce the observed bunch intensity at the second harmonic frequency as a function of time:

$$I_2(t) = \frac{-B \sin(x(t)w)}{x(t)[e^{\alpha t} - c_1 k^{-1} + c_1 k^{-1} e^{\alpha t}]}.$$

where $B$ is the new amplitude constant. To avoid the double $N_0$ dependence of both the $B$ and $c_1$ factors, a change of variables is performed replacing $c_1$ with $B\alpha'$. The expected pickup amplitude is now given by

$$I_2(t) = \frac{-B \sin(x(t)w)}{x(t)[e^{\alpha t} - B\alpha k^{-1} + B\alpha k^{-1} e^{\alpha t}]}.$$

The decay of the pickup signal is dominated by intrabeam scattering $\alpha'$ at short times (particularly for higher-amplitude ion bunches), exponential particle loss $k$ due to the dc beam at intermediate times and the expansion of the bunch ($\beta$ and $\beta_2$ contained in $x(t)$) at late times.

### 5.6. Decay model discussion

The reproduction of the measured decay curves over two to three orders of magnitude using equation (14) is very good for both ion species, as shown by the sample fits in figure 6. The average of the parameters obtained from a least-squares fit for all 51 injections (34 $N_2^+$ injections and 17 $Al_2^+$ injections) is summarized in table 2.

The fitted $\alpha'$ values for both ion species describing the intrabeam scattering agree within their error bars, and no dependence on any other parameters within the parameter range investigated was found. Further studies, however, are needed to clearly determine any possible
The measured bunch loss rate $k$ for both ion species exhibits a large error due to the spread of the fitted $k$ values. All these values are, however, about an order of magnitude larger than the ion trap loss rate due to residual gas collisions ($k_c = 2.9 \pm 0.5 \times 10^{-3} \text{ s}^{-1}$ [8]). The decay of the bunch within less than 12 s in addition to the fitted $k$ values clearly indicates that a process independent of residual gas dominates bunch loss because the ions are seen to remain stored long after leaving the bunch. This stringent requirement agrees very well with the result of the photo-detachment measurement, which demonstrated the formation of a dc beam component stored simultaneously inside the trap with the bunch. Collisions between the dc beam and the ion bunch ($k_{dc}$) most likely dominate $k$ (see equation (11)) as explored in more detail using RF bunching experiments in the following section. Note that the bunch loss rate $k$ observed using the very short ion bunch in section 4 can be estimated from the portion of total charge remaining in the bunch after 1.4 ms. This loss rate ($k > 1$) is significantly larger than the $k$ values from the few $\mu$s ion bunches (in table 2). Such high bunch loss rates would easily explain the fast bunch decay and suggest that $k$ depends on the initial bunch width $x_0$. This also supports the idea of minimum charge requirements in the formation of stable ion bunches as discussed above.

The fitted $\beta$ and $\beta_2$ values yield self-consistent results, where the calculated bunch duration (or time required for the bunch to decay and longitudinally fill the trap) and the observed bunch duration agree. The $\beta$ values for both species span a similar range, while those for $\beta_2$ differ by a factor of 10, which significantly exceeds the experimental error bar. This could suggest a mass or velocity dependence, but further studies are required for confirming this.

Model simulations employing the $N_2^+$ values from table 2 while varying either the initial bunch length or the bunch amplitude highlights the influence of both parameters on the pickup signal. As shown in figure 7(a), the bunch duration or expansion terms ($\beta$ and $\beta_2$) are strongly dependent on the initial bunch length. Note that while the charge density is relatively constant, the longer bunches contain by definition more charge, thus explaining the change in bunch amplitude. A revival of the bunch is also predicted to occur at later storage times, albeit at much lower intensities near our background level. This revival is, however, most likely an artifact.
of our model as intuitively one does not expect the homogeneous charge distribution to bunch again after achieving such an entropic state. The predicted bunch length dependence is also in agreement with the observed bunch decays as the bunch lengths of Al$^-$ were twice as long as those from N$^+_2$. The much shorter bunch employed in the bunch-profile measurement in section 4 also clearly demonstrates that such short bunches containing little total charge are not stable over long storage times in comparison to the longer bunches employed in section 5. The dependence of the bunch decay on the initial charge density is explored in figure 7(b), where the number of charges clearly not only changes the initial amplitude, but also changes the form of the decay as intrabeam scattering losses begin at higher charge densities. As explored above, however, the shorter bunches likely exhibit larger $k$ values as opposed to a constant value, which would significantly reduce the observed bunch duration.

6. Radio-frequency bunching

Continuous RF pulses can also be applied to a trap or pickup electrode [7, 14], thereby forcing the bunching of the beam and ensuring that they maintain a given oscillation frequency as long as the RF is active, despite other processes such as ion–ion space-charge interactions, which lead to the dispersion of the bunch. RF bunching ensures that the ion bunch remains coherently together by slightly accelerating or decelerating particles located behind or in front of the synchronous particle, respectively. While still employing the trap in the self-bunching mode, 10 V$_{pp}$ sinusoidal RF at a frequency of 123.15 kHz was applied to the exit-side einzel lens electrode (electrode B of mirror 2 in figure 1). This frequency corresponds to the oscillation frequency of the stored 7.1 keV N$^+_2$ bunch in the trap. The amplitude of the second harmonic frequency ($h = 2, f = 246$ kHz) was then observed as a function of time after ion injection as shown in figure 8(a). The initial decay of the bunch can be seen from 0 to 11 s after which the bunch was dispersed, having filled the entire trap along the beam axis. A fit of this decay yields results consistent with those presented for N$^+_2$ without RF bunching in table 2. Continuous RF was applied after 15 s of storage, resulting in the rapid re-bunching of at least a portion of the ions, which then exhibited a much slower bunch decay until the trap was emptied. A fit of this second slower decay was only possible after the initial losses due to the sudden application of RF and re-bunching of the ions were finished. The unknown bunch length prevents a precise determination of the model parameters, but values similar to the measurements below were observed.

The RF bunching of ions inside an EIBT was further investigated by employing a 6.0 keV N$^+_2$ ion bunch (see other parameters in table 1). A continuous 10 V$_{pp}$ RF signal at the ion bunch’s oscillation frequency (113.05 kHz) was once again applied, where the two ion injections can be seen in figure 8(b). The bunch intensity after background subtraction is plotted as a function of time after ion injection. Two sample error bars in each curve around 500 s after injection are shown. The bunch is clearly visible throughout the entire trapping period exhibiting a slow, exponential decay below a pickup amplitude of 0.1 mV until the trap was turned off after 600 s. The higher pickup amplitude injection also demonstrates non-exponential loss at earlier times due to intrabeam scattering. Since the RF bunching is only expected to inhibit the expansion of the bunch, the bunch decay model in equation (14) can again be used to fit the observed decays. While small deviations from the bunch decay model are visible 300 s after ion injection, the decays are still well reproduced, particularly for early storage times. The only difference between the two injections is the initial number of ions in the bunch and the relative phase of
Figure 8. (a) Demonstration of the re-bunching of 7.1 keV N$_2^+$ by applying RF after the bunch had dispersed in self-bunching mode, where the pickup amplitude is plotted versus time after injection. (b) Two injections of 6.0 keV N$_2^+$ bunches and their signal amplitudes versus storage time in self-bunching mode with permanent RF bunching after background subtraction. The trap was turned off after 40 or 600 s, respectively, and equation (14) has been fitted to the bunch decay data.

the RF with respect to the injected bunch. These two injections were fitted using equation (14) and are summarized in table 2. While the model can very accurately reproduce the first 300 s of the decay, an acceleration in the bunch decay is observed in both curves around 300 s. This effect may be an instrumental artifact and is not yet understood. The 600 s trapping time corresponds to an ion path length of over 91 000 km or almost 68 million ion oscillations.

While the pickup signal amplitudes were similar to the bunch decay results presented above, all other model parameters exhibited differences. As expected, the presence of RF bunching dramatically reduced the longitudinal increase of the bunch length, as shown in the reduction of $\beta$ and $\beta_2$ by five and four orders of magnitude, respectively, in comparison with the bunching experiments also summarized in table 2.

The decrease in the intrabeam scattering term $\alpha'$ by an order of magnitude can most likely be attributed to the increase in the bucket height via the application of RF. The resulting larger bunch phase space reduces intrabeam scattering losses from the bunch by requiring larger energy exchanges for bunch loss. This observation is, therefore, also consistent with the adapted EIBT bunch dynamics discussed above.

The bunch loss rate $k$ is now of the same order as the collisional ion loss rate due to residual gas ($k_c = n \langle \sigma_c v \rangle$). It is also possible that losses at the same order of magnitude due to phase noise from the RF generator also contribute to this low $k$ value. The most likely explanation for the observed decrease of $k$ is a strong reduction in the number of collisions between the bunch and the dc beam component due to the suppression of the dc beam component by the RF bunching. The suppression of this component is expected to eliminate any corrections to $k$ due to collisional ion losses ($k_c$) throughout the 600 s storage time. An estimate of the Coulomb collisional cross section based on Rutherford scattering yields values at least an order of magnitude larger for small incident angles ($<1.2^\circ$) or four orders of magnitude larger at angles below $0.2^\circ$ than that of residual gas collisions ($\sigma_c \approx 23 \times 10^{-16}$ cm$^2$ [35, 36]). An estimation of this newly identified EIBT bunch loss process can be given by $k_{dc} = 2n_{dc} \langle \sigma_{dc} v \rangle$, assuming that
the collisions occur between ions (in the bunch and dc beam) traveling in opposite directions. Using the known rest gas density \( n = 2000 \) particles per cm\(^3\), an estimation of the number of required ions in the dc beam can be made. Taking a very conservative value of \( \sigma_{dc} = 10 \sigma_c \) with a 1 cm diameter beam yields \( \approx 5000 \) ions stored for the CTF’s effective trap length of 66 cm. Considering that injected ion currents of over 2 \( \mu \)A were used, corresponding to over \( 1 \times 10^6 \) stored ions using the above stated trapping efficiency and bunch lengths, a mere 5000 ions lost from the bunch to form the dc beam is very realistic. Collisions between the stored ion bunch and a dc beam of stored ions in the absence of RF bunching as well as intrabeam scattering between ions in the bunch are, therefore, proposed as new EIBT ion bunch loss mechanisms.

The low \( k \) values achieved both with RF bunching (0.0060(8) s\(^{-1}\)) and without (0.0029(5) s\(^{-1}\)) [8] also establish an upper limit on all other ion bunch and ion beam loss processes in an EIBT. Small losses, magnified over the course of the employed trapping time, could conceivably arise from local electric field imperfections, imperfect trap alignment, electrode voltage stability and local magnetic fields. All of these loss processes, however, must be smaller or of the same order of magnitude as residual gas collisional losses. An upper limit, therefore, on these other possible EIBT loss processes, at least for the CTF trap geometry and environment, has been established.

7. Summary

The extreme vacuum of the CTF has permitted the investigation and thereby a better fundamental understanding of ion bunch decays in EIBTs by providing a trapping environment almost independent of collisional beam loss.

A laser pulse directed orthogonally with respect to the ion beam axis was employed to measure the longitudinal ion bunch profile by photo-detaching aluminum dimer anions. This measurement simultaneously demonstrated the presence of a dc ion beam component in the trap after a hundred ion bunch revolutions, co-existing with the oscillating ion bunch.

The time span over which stable bunches have been observed in the self-bunching trapping mode has been extended by more than two orders of magnitude, to times as long as 12 s. The decay of these bunches was observed for the first time, and was also found to be intensity dependent. These decays provide the first experimental support to theoretical models, suggesting that a minimum charge is required for stable bunches. A semiempirical bunch decay model was developed, based on the expansion of the bunch with respect to time in combination with intrabeam scattering and bunch loss attributed to small-angle collisions between the stored bunch and the quickly arising dc ion beam. Our model was able to very accurately reproduce the observed decays of both \( \text{Al}_2^- \) and \( \text{N}_2^+ \) ion bunches. It also predicts a strong dependence of the bunch expansion on the initial bunch length, in agreement with our decay experiments. To our knowledge, these results are also the first confirmed observation of intrabeam scattering in an EIBT.

RF bunching of the ions resulted in the extension of the bunch observation to 600 s. The bunching concepts from storage ring physics were adapted to EIBTs and supported by RF bunching observations. Parameter fits of these bunch decays with our model demonstrate that both intrabeam scattering and the expansion of the bunch were reduced (by up to five orders of magnitude) as expected in the presence of RF. The bunch loss rate \( k \) was also observed to decrease, almost to the rate of collisional losses due to residual gas in the CTF, which constitutes the ultimate limit for bunch loss. This reduction has been attributed to a newly observed ion
bunch loss mechanism, namely collisions between the ion bunch and a dc ion beam component, which is stored simultaneously in the absence of RF bunching. This measured decay constant also places an upper limit on all other EIBT bunch and beam loss processes.

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