Time-dependent dynamics of radio-frequency-bunched ions in an electrostatic ion beam trap

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The dynamics of ions in an electrostatic ion beam trap in the presence of an external time-dependent field is studied with a recently developed particle-in-cell simulation technique. The simulation technique, capable of accounting for space-charge effects, has reproduced all the experimental results on the bunch dynamics in the radio frequency mode. With simulation, the motion of ions is visualized in phase space and it is shown that the ion-ion interaction strongly affects the distribution of ions in phase space in the presence of an rf driving voltage.

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I. INTRODUCTION

Ion traps and storage devices are valuable tools used in various fields of atomic and molecular physics [1]. The electrostatic ion beam trap (EIBT) [2,3] is a unique trap where ion beams are trapped between two sets of mirror electrodes using electrostatic fields only, which has the advantage of trapping ions without any mass limitation [4]. Since its development, the EIBT has proven to be extremely versatile and has been used for the study of time-dependent processes in atomic, molecular, and cluster ions, mass spectrometry, as well as the study of the trapping dynamics itself. This has yielded interesting effects such as self-bunching of ions [5], demonstrating the critical importance of the ion-ion interaction. Simulations of the ion behavior in the EIBT have been made using both analytical models [6] as well as numerical ones, the latter allowing for the full integration of the ion-ion interaction [7] using particle-in-cell (PIC) numerical simulation. The comparison between these numerical simulations and the experimental results has enhanced the understanding of the behavior of trapped ions. The possibility to fully include the ion-ion interaction in numerical simulation is a major advantage and allows for a better understanding of various characteristics of ion storage in the EIBT.

In the present paper, we extend our numerical simulation studies of the EIBT [7] in order to include the application of a time-dependent external field. Applying such a time-dependent field on the trap electrodes allows for more sophisticated manipulation of the stored ions. For example, by applying an ac electric field with a frequency f_{rf} that matches the natural ion oscillation frequency f_{osc}, or its integer harmonics, nf_{osc}, to one of the trap electrodes, the ions can be bunched. This technique, known as radio frequency (rf) bunching, causes every ion to oscillate in the longitudinal phase space around the synchronous ion [8], as is well known in heavy ion storage rings [9,10]. The rf-bunching technique has various applications in the area of molecular dynamics spectroscopy [11,12], or for estimating the number of ions of a certain mass inside the trap. Moreover, one can generate a superposition of several frequencies and measure the dynamics of several different masses simultaneously [13]. The ion-bunch dynamics using the rf-bunching technique have been studied experimentally in the EIBT, and the sideband structure manifested by the dipole and quadrupole oscillations [11,12] has also been characterized analytically. Several new questions arise when adding an rf perturbation to the trap, such as its impact on bunch lifetimes and trapping efficiency. It has been found empirically that with a sufficiently large rf amplitude, ions are removed from the trap, shortening the measured lifetime [13]. Recently a frequency-chirped rf pulse was also tested, resulting in ion-bunch cooling [14] where the ion-ion collisions inside the bunch contribute to the thermalization of the ions. All these processes require an understanding of the role of ion-ion interaction and the interaction of ions with the external field, making the PIC simulation technique a useful tool to study the dynamics of ions in the EIBT.

In this paper, a systematic study of the dynamics of the ions in the rf-bunching mode of the trap is presented with a recently developed simulation technique based on a two-dimensional (2D) PIC code in cylindrical coordinates (2DCYLPIC) [7]. In the following, we demonstrate the surprising effect of ion-ion interaction on the bunch behavior where, unexpectedly, the repulsive interaction leads to a lack of filamentation of the phase space. Wherever possible, the simulation results are compared with the experimental data.

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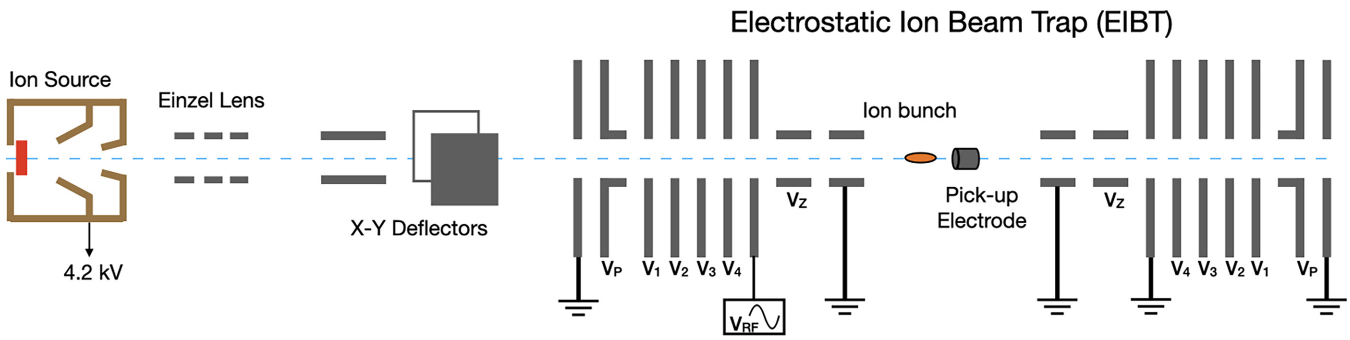

FIG. 1. Schematic view of the experimental setup, including the EIBT with pickup electrode and rf.

II. EXPERIMENTAL SETUP

The EIBT used for the present paper is schematically depicted in Fig. 1. More details about the EIBT and its characteristics can be found in [15–20].

Bunches of SF_5^+ are produced in a pulsed (20-μs pulse duration) Even-Lavie supersonic expansion source [21]. The gas injected into the valve consists of SF_6 (≈1%) mixed with argon as a carrier gas (≈99%) at a total pressure of ≈180 psi. The mixture is supersonically expanded through the pulsed nozzle and is ionized by an electron pulse accelerated to an energy of about 200 eV, impacting the expanding gas ≈1 mm downstream from the nozzle. The source is mounted on a high-voltage platform, which accelerates the ions to $E_k =$ 4.2 keV, which then pass through a skimmer, an Einzel lens, and a pair of orthogonal deflectors prior to being injected and trapped in the EIBT operated in dispersive mode [7]. The settings of the electrode potentials used in the experiment were $V_p = 5750$ V, $V_1 = 6500$ V, $V_2 = 4875$ V, $V_3 = 3250$ V, $V_4 = 1625$ V, and $V_z = 3400$ V. The other electrodes were grounded. The opening of the entrance mirror was achieved by shortly lowering the V_p voltage to ≈1600 V. After each injection, the ion beam was trapped for about 800 ms. The pressure in the trap was $\approx 3 \times 10^{-10}$ Torr and the average lifetime was on the order of 1000 ms [13]. In such a mode, the ion bunches disperse within a few milliseconds and fill up the trap, and their second harmonic oscillation frequency is measured to be 187 635 Hz, obtained from the Fourier transform (FT) of the pickup signal without time-dependent fields. The time evolution of the ion bunch can be observed using a cylindrical pickup electrode [13] that is slightly shifted from the center of the trap towards the exit mirror and connected to a digitizer (see Fig. 1). This shift in the position of the pickup is useful to determine the direction in which the beam is traveling. The pickup is connected to the gate of a junction field transistor whose drain is fed to a charge sensitive amplifier. The amplified signal is recorded as a function of time with a digital oscilloscope at a sampling rate of 10–50 MHz. The signal from the pickup electrode representing the time trace of a bunch of SF_5^+ is recorded by a computer and analyzed in real time.

The rf bunching of ions in the experiment is achieved by applying a sinusoidal rf field (of amplitude between 0.5 and 5 V) to one of the electrodes of the entrance mirror (see Fig. 1). This electrode is usually grounded when experiments without rf bunching are conducted. The time-dependent external voltage applied to this electrode is given by

$$V(t) = V_{\mathrm{rf}} \sin(2\pi f_{\mathrm{rf}} t + \phi) \quad (1)$$

where V_{rf} is the applied rf amplitude, $f_{\mathrm{rf}} = 187\,635$ Hz is the second harmonic of the oscillation frequency of the ions, and ϕ is the phase of the rf.

III. 2DCYLPIC SIMULATIONS

The 2DCYLPIC simulation has been described in detail earlier [7]. The simulation is based on the principle of PIC technique [22] that has been widely used in studying plasmas, gravitational systems, geodynamics, etc. The concept of 2DCYLPIC simulation for an EIBT originated from the 3DCYLPIC simulation [23] particle-in-cell code originally written to simulate ion trap and ion transport devices using a three-dimensional (3D) cylindrical coordinate system. It has since been updated to support other 2D and 3D coordinate systems and is a natural choice in studying the ion dynamics in the EIBT. Recently, this technique has been employed to study the advanced cryogenic gas stopper [24]. The idea behind the 2DCYLPIC simulation is based on solving Poisson's equation numerically on a computational grid, in order to obtain the electric field at each grid point. The positions and velocities of simulation particles representing ions are then updated based on their location on the grid. Next, the new particle locations are used to update the charge density on the grid, at which point the electric field is recalculated. The simulation continues in this fashion until complete. In all the simulations described here, the time step $\Delta t = 5$ ns was used. The general practice is to choose the largest value of Δt that does not significantly change the simulation results. The simulation will continue in steps of Δt until the end condition is satisfied, or the simulation is manually terminated.

The present simulations were carried out in the dispersive mode with an external time-dependent force applied to one of the electrodes in a setting that is identical to the experimental setup. The optimized potentials as given in Sec. II of [7] were used for all of the simulations in the present paper. The rf frequency used to bunch the ions for the current set of mirror electrode potentials is $f_{\mathrm{rf}} \approx 187\,748$ Hz, which was obtained from the FT of the pickup signal from the simulation without external force applied. The small discrepancy of 113 Hz between the experimental and simulated value of f_{rf} is mostly due to the differences in the exact trap geometry as well as the

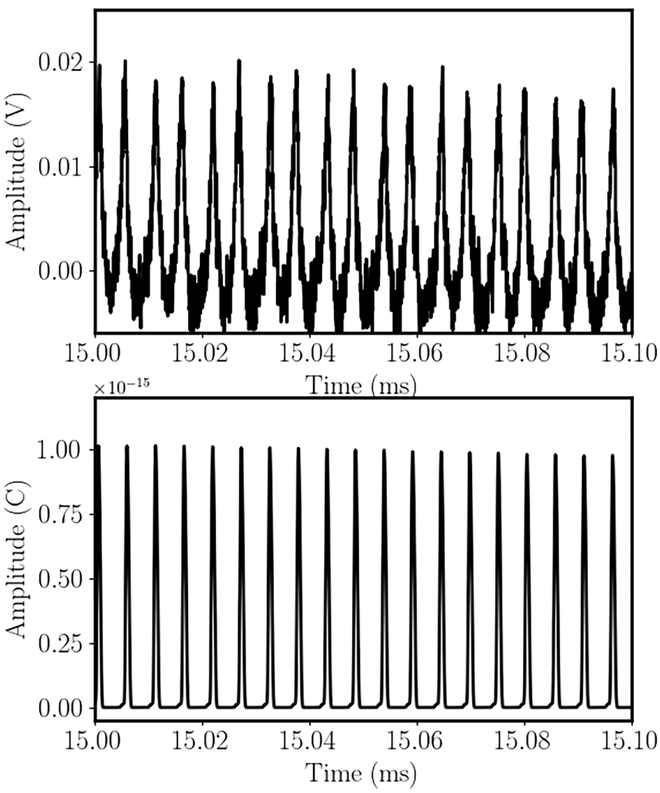

FIG. 2. Typical time trace of the pickup signal for 100 μs from 15 to 15.1 ms from the experiment (top) and simulation (bottom) (100 000 ions) in the rf-bunching mode of the trap with the rf amplitude $V_{\mathrm{rf}} \approx 2.5$ V.

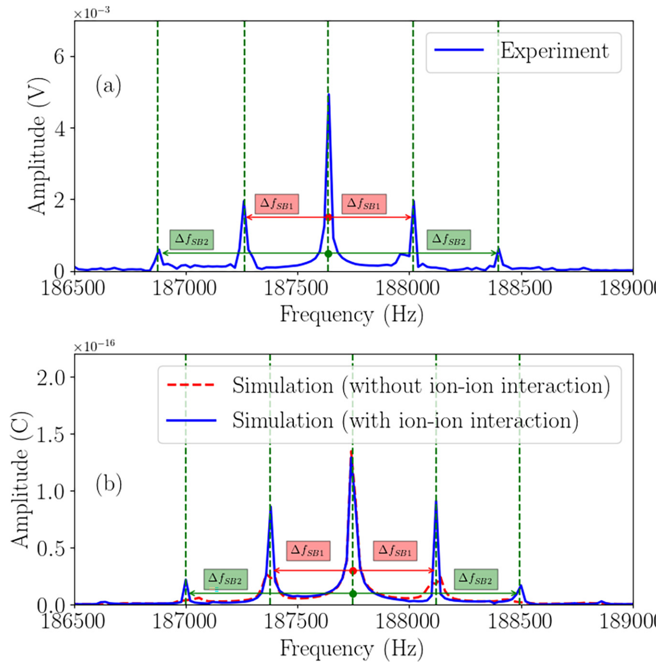

FIG. 3. FT of the pickup signal for an rf-bunched SF_5^+ beam ($V_{\mathrm{rf}} = 2.5$ V) around the second harmonic of the oscillation frequency of the ions. (a) From the experiment, measured for the single injection into the EIBT, and (b) from the simulation for 100 000 ions for 50 ms, with (blue line) and without (dashed red line) ion-ion interactions.

experimental uncertainty of the beam energy. The amplitude of the V_{rf} has been varied from 0.5 to 5 V. The kinetic energy of the beam was set at 4.2 keV with an initial energy spread of 3 eV and an initial bunch width of 1 μs, closely matching the initial experimental conditions.

IV. rf BUNCHING

A typical time trace of the pickup signal from both the experiment and simulation is shown in Fig. 2. The displayed data consist of a narrow slice of time, from 15 to 15.1 ms, when an external rf force with an amplitude of 2.5 V is applied.

Figures 3(a) and 3(b) show the FT of the time trace of the pickup signal integrated over 50 ms with an rf signal applied with an amplitude of 2.5 V, for the experiment and the simulation, respectively. Both plots are zoomed around the second harmonic of the oscillation frequency of the ions. Besides the main peak, sideband peaks are visible, related to the expected dipole and quadrupole oscillations. The dipole sidebands are related to the motion of the mean position of ions around the ideal synchronous ion [11]. The quadrupole sidebands can be ascribed to the oscillation of the width and amplitude of the ion bunches with 180° phase difference [12].

For the experimental data [Fig. 3(a)], the frequency differences Δf_{SB1} and Δf_{SB2} of the sideband structure from the main peak closely follow $\Delta f_{\mathrm{SB2}} = 2\Delta f_{\mathrm{SB1}}$ with $\Delta f_{\mathrm{SB1}} =$ 377 Hz and $\Delta f_{\mathrm{SB2}} = 763$ Hz approximately.

The FT of the time trace of the simulated pickup data is shown in Fig. 3(b) with (blue line) and without (red dashed line) ion-ion interactions for a total of 100 000 trapped ions. The FT from the simulations (with ion-ion interaction) reproduces similar features (main peak and sharp, prominent sidebands) as observed in the experimental results shown in Fig. 3(a). In good agreement with the experiment, the first sideband peak Δf_{SB1} is approximately at a distance of 374 Hz, and the second sideband peak Δf_{SB2} is at 747 Hz. The frequency differences Δf_{SB1} and Δf_{SB2} of the sideband structure from the main peak follow $\Delta f_{\mathrm{SB2}} = 2\Delta f_{\mathrm{SB1}}$, similar to the experimental observation. For the case without ion-ion interaction, it is seen that the first and second sidebands related to the dipole and quadrupole oscillation are significantly suppressed, whereas, for the case taking the ion-ion interaction into account, the sidebands are sharp and higher in amplitude. The amplitude of the sidebands depends on many parameters such as bunch size, rf amplitude, rf phase, and ion density. The subject is now under study, but some additional information can be found in [11].

According to the analytical model described in [11], which is well reproduced experimentally, the frequency difference Δf_{SB1} shows a square root dependence on the applied rf voltage $\Delta f_{\mathrm{SB1}} = C\sqrt{V_{\mathrm{rf}}}$ where the parameter C depends on the exact experimental conditions. We tested this dependency with the simulation. The results, together with the measurement performed with the current experimental setup, are shown in Fig. 4. The simulation results agree very well with the measured values within the uncertainty of the experiment and reproduce not only the square root dependence of the

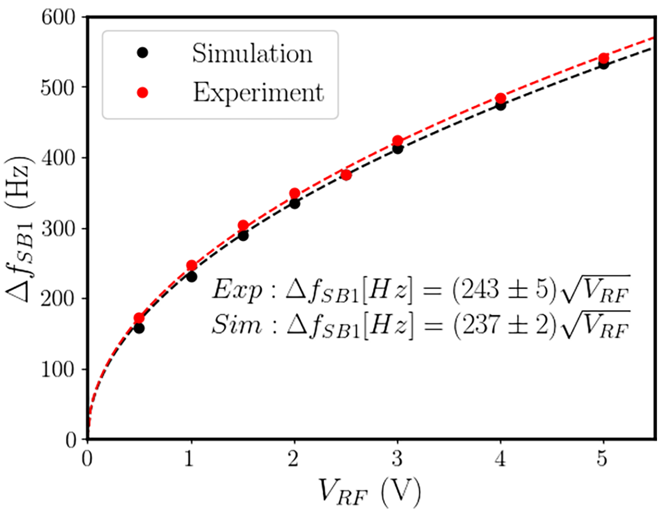

FIG. 4. Comparison of the frequency difference Δf_{SB1} between the first sideband peak and the main peak as a function of the applied rf-voltage amplitude from the experimental measurement and simulation (100 000 ions used).

Δf_{SB1} as a function of applied rf voltage but also the coefficient C described above.

V. MOTION OF IONS IN THE RF BUCKET

The dipole and quadrupole oscillation of ions in the EIBT, with an applied rf field, has been studied extensively [11,12]. The dipole peaks are attributed to the longitudinal oscillation of ions around the synchronous ion and are characterized by the oscillation of the mean position of the bunch relative to the position of the synchronous ion. For a nonuniform distribution of ions in the rf bucket, the dipole peak will be enhanced in the FT spectrum. The quadruple oscillation represents the oscillation of the height and the width of the bunch with 180° phase difference. In Fig. 3(b), dipole and quadrupole oscillation of the bunch is enhanced when the ion-ion interaction is present and suppressed when there is no ion-ion interaction. The effect can be understood more clearly with the motion of the ion bunch in the rf bucket. The rf bucket corresponds to the phase space area contained within the separatrix. The separatrix is the line in phase space where the ions with maximum phase difference from the synchronous ion will oscillate in a closed orbit. A schematic of the rf phase space is shown in Fig. 5. The synchronous ion is located at the center of the bucket. The ions arriving earlier than the synchronous ions will lose velocity and vice versa and oscillate within the bounds of the separatrix. The PIC simulation grants the ability to follow the evolution of the phase space, both with and without the ion-ion interaction, to gauge its impact.

The injection of an ion bunch smaller than the rf bucket size causes phase space filamentation [25]. In the simulation, a 1-μs bunch of ions is injected in the trap, and a constant rf of 187 748 Hz (bucket size approx 2.7 μs) is applied for 50 ms. In Figs. 6(a) and 6(b), the phase space distribution of ions without and with ion-ion interaction is shown, respectively. When there is no ion-ion interaction, the ions disperse in the rf bucket, resulting in phase space filamentation. How-

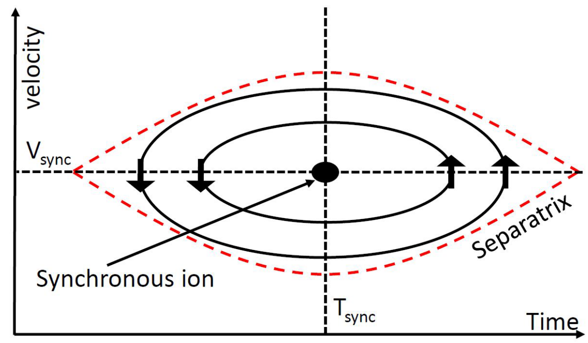

FIG. 5. A schematic of rf phase space showing the separatrix (dashed red line), synchronous ion, and direction (arrows) of motion of other ions in the bucket.

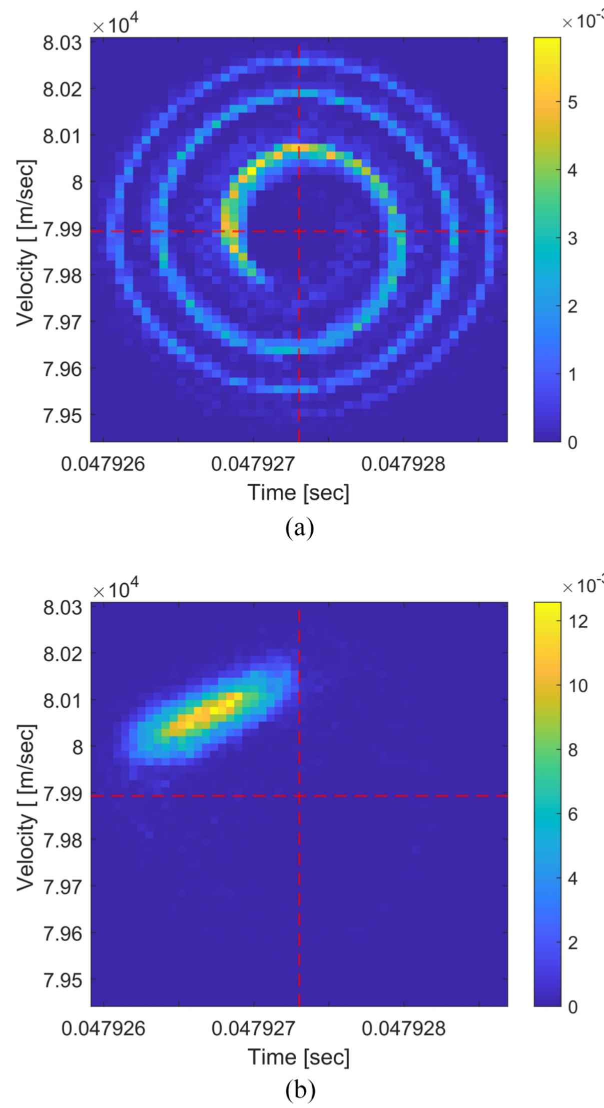

FIG. 6. Position of ions in the rf bucket after approximately 48 ms. (a) Without ion-ion interaction. (b) With ion-ion interaction. The false color bar represents the normalized intensity of ions. Red dashed lines show the position of the bucket center.

ever, surprisingly, when ion-ion interaction is present, the ions are kept in a bunch, moving within the rf bucket, in a striking difference when compared to the no ion-ion interaction case. The fact that the ions stay, in this case, "localized" within the rf bucket is also demonstrated by the enhancement of the dipole peaks as shown in Fig. 3. These results demonstrate that, unexpectedly, the ion-ion interaction leads to a lack of filamentation of the phase space.

The synchrotron oscillation in the rf bucket is nonlinear and depends on the oscillation amplitude [25]. This nonlinearity causes filamentation of the phase space and is clearly seen in Fig. 6(a) when ion-ion interaction is not present. Such filamentation is well known and observed in storage ring setups, where ion-ion interaction is present. However, in the present ion trap, unlike in storage rings, the ion density itself strongly oscillates, since the ions are compressed at the turning point of the trap. As calculated in [6], such an increase in the ion density can be up to three orders of magnitude. Moreover, and similar to the self-bunching effect (i.e., without rf) as demonstrated in [5], the dynamics of the ions in the trap are strongly influenced by the slope of the potential at the trap edges, yielding positive or negative slip factors.

VI. SUMMARY

In this paper, we have studied the time-dependent dynamics of rf-bunched ions trapped in an EIBT. The experimental results are very well reproduced by the simulation. Without ion-ion interaction, filamentation of the phase space is observed, while the ion bunch maintains its shape when the ion-ion interaction is included. Although more experiments and simulations are needed to fully understand this effect, it is clear that the possibility to keep the phase space density compact under rf-bunching conditions for the whole trapping time may lead to some interesting opportunities, such as the manipulation of the phase space to achieve kinematical cooling of the stored ions. A complete study of this effect and its implications is currently underway, but still requires numerical optimization of the simulation in order to reduce the computation time.

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