

## Ion Motion Synchronization in an Ion-Trap Resonator

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Using a new type of ion trap, we demonstrate that the length of a packet of charged particles oscillating between two electrostatic mirrors will remain constant under special conditions. The effect can be understood in terms of phase synchronization, where, in a rather counterintuitive way, the repulsive Coulomb interaction between the ions actually holds the packet together. Application of this effect for mass spectrometry is discussed.

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Eight years after Huygens invented the pendulum clock, he observed that the pendula of two clocks hanging on the same wall were phase locked [1]. It is now understood that such synchronization occurs when there is some nonlinear coupling mechanism between two systems oscillating at nearby frequencies. Modern examples of synchronization are to be found in laser [2] and Josephson junction [3] arrays, magnetic resonance processes [4], and a large variety of chemical and biological systems [5–11]. In this Letter we describe the synchronized oscillations of a large number of ions forming a bunch in an electrostatic ion trap, where the coupling mechanism is the repulsive Coulomb interaction between them. The absence of dispersion, despite the finite energy spread of the ions, can be exploited to transform the trap into a mass spectrometer with extremely high resolution.

The experimental system allows trapping of a bunch of fast ions between two identical electrostatic “mirrors” [12,13]. A schematic view of the trap is shown in Fig. 1. Each mirror is made of eight cylindrical electrodes. The distance between the grounded innermost electrodes is 228 mm, thus creating a large field-free region in the center. The ion bunch, produced in an external ion source (not shown), can be injected into the device through the entrance (left) electrodes which are initially grounded. Once the bunch is inside the trap, the potentials ( $V_1$  to  $V_5$ ) of the entrance electrodes are switched on, the potentials of the exit electrodes being always on. The stability conditions for this system [12,13] are analogous to the stability condition of an optical resonator. For a symmetric resonator, the criterion is

$$\frac{L}{4} < f < \infty, \quad (1)$$

where  $L$  is the distance between the mirrors and  $f$  is their focal lengths. In the electrostatic case,  $f$  is determined by the potential configuration of the electrodes. A similar ion trap was developed by Benner [14] for mass spectrometry of single ions produced by an electrospray source. In practice, ions are lost from the trap because of collisions

with the residual gas (typical pressure  $< 10^{-9}$  Torr), leading to charge exchange and multiple scattering processes so that the number of trapped ions decreases exponentially with time. In the present study, we used a bunch of 4.2 keV  $\text{Ar}^+$  ions having an oscillation frequency of 340 kHz. The lifetime of the beam was monitored by measuring the number of neutral atoms exiting the trap and impinging on a microchannel plate (MCP) detector [12,13]. The length of the injected bunch was determined by a fast electric chopper located near the ion source and was set at 130 ns (giving a bunch length of 18 mm and a density of  $6.7 \times 10^4$  ions/cm<sup>3</sup>). The image charge induced in a short metal cylinder (length = 7 mm; ID = 18 mm) located at the center of the trap (see Fig. 1) was used to detect the passage of the ions. An example of the observed signal for a bunch of about  $\sim 10^5$   $\text{Ar}^+$  ions 100  $\mu\text{s}$  after injection is shown in Fig. 2(a). The width  $W$  of the peak is proportional to a convolution of the length of the bunch and that of the pickup, while the area under the peak is proportional to the number of ions in the bunch.

Assuming that the Coulomb interaction between the ions is negligible, the time evolution of the bunch length during storage depends on two factors, both of which lead to spreading (debunching) of the bunch. First, the ion velocity  $v$  is distributed around a mean value. The width of this

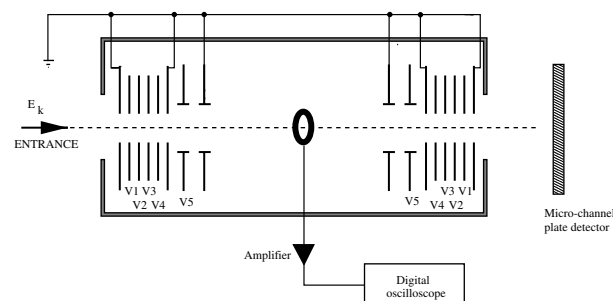


FIG. 1. Schematic view of the ion beam trap. The bunch is injected through the left-hand side of the trap (entrance electrodes). The central ring represents the pickup electrodes used to measure the induced charge produced by the bunch.

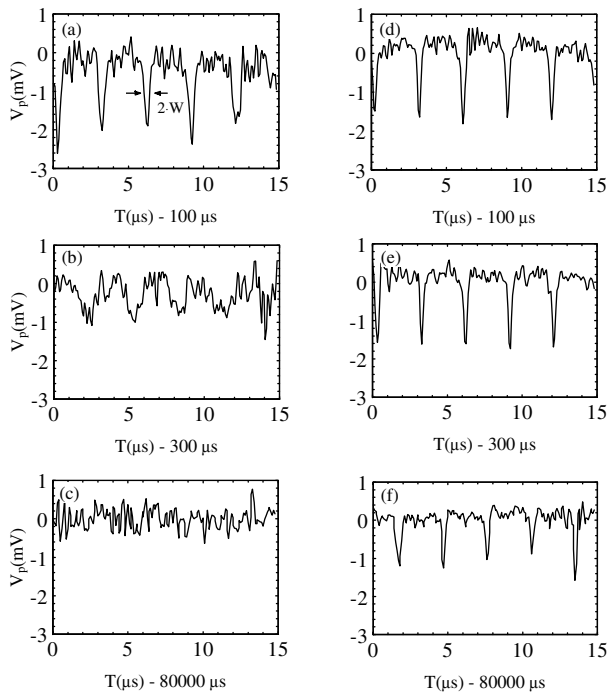


FIG. 2. Signal as observed from the pickup electrode of the trap, for different time windows and different potential configurations: (a), (b), and (c) are for the configuration where  $\Delta T = 3.57$  ns, and (d), (e), and (f) are for  $\Delta T = 0.18$  ns. The time span for each plot is  $15 \mu\text{s}$ .

distribution,  $\Delta v$ , is mainly due to the characteristics and mode of operation of the ion source. We used an electron impact ion source with a relatively narrow distribution  $\Delta v/v < 0.1\%$  (manufacturer's specifications). Such a velocity spread yields a time spread for each oscillation which we denote as  $\Delta T_v$ . This value is both a function of the length of the trap and the shape of the electric field within the mirrors. Second, the ensemble of stable trajectories in the trap has an intrinsic distribution of oscillation times: Ions stored with small transverse velocities, and located close to the axis, have slightly shorter oscillation periods than particles with larger transverse velocities, or are located further from the axis of the trap. The time spread per oscillation due to this intrinsic property of the trap is designated by  $\Delta T_i$  and is a function of the field configuration.

Between these two factors, the intrinsic width  $\Delta T_i$  is generally dominant. Both can be estimated using a simulation program [15] in which the electrode potentials and the ions' initial velocities and distances from the axis are parameters, and one can analyze the trajectory evolution for as many oscillations as desired. Indeed, it is possible to change both  $\Delta T_i$  and  $\Delta T_v$  by choosing different configurations of the potentials for which Eq. (1) is still valid. Assuming that  $\Delta T_i$  and  $\Delta T_v$  are independent, the total time spread for each oscillation of the bunch in the trap is given by  $\Delta T = \sqrt{(\Delta T_v)^2 + (\Delta T_i)^2}$ . For noninteracting ions, where each ion preserves its initial oscillation period,

it can be shown [16] that the bunch length  $W_n$  increases with the number of oscillations  $n$  in the trap as

$$W_n = \sqrt{W_0^2 + n^2 \Delta T^2}, \quad (2)$$

where  $W_0$  is the length of the bunch at the beginning of storage.

The bunch length was measured as a function of storage time for two different potential configurations. In the first, the electrode potentials were adjusted such that the calculated [15] time spreads were  $\Delta T_i = 3.56$  ns and  $\Delta T_v = 0.18$  ns, yielding  $\Delta T = 3.57$  ns. The maximum beam radius for which Eq. (1) is valid for this configuration is 3.6 mm [15]. Figure 2 shows the measured signal for three different time windows: (a)  $100 < t < 115 \mu\text{s}$ , (b)  $300 < t < 315 \mu\text{s}$ , and (c)  $80000 < t < 80015 \mu\text{s}$ . Clearly, the signal broadens and disappears after a few hundred microseconds. It is important to point out that the disappearance of this signal is substantially shorter than the lifetime of the ion beam, which was measured using the MCP to be  $\sim 160$  ms. Figure 3(a) shows the evolution of the bunch length ( $W_0 = 130$  ns) obtained by fitting the peaks of the measured signal to a Gaussian function. Also shown in Fig. 3(a) (solid line) is the bunch length given by Eq. (2), and the calculated value of  $\Delta T$ . It can be seen that the calculated values are in very good agreement with the measured values. After about  $400 \mu\text{s}$ , the signal could no longer be distinguished from the noise [see Fig. 2(b)].

The same measurement was performed with a different potential profile at the mirror electrodes, chosen to give a much smaller  $\Delta T$ . Such a reduction is mainly achieved by using potentials for which the optical aberration is large, so that Eq. (1) holds only for ions located close to the axis of the trap ( $< 2$  mm), thereby reducing the intrinsic width  $\Delta T_i$ . The potentials were configured to focus the ion beam inside the mirrors where the ions are slowed down. As a result, the density of ions was strongly increased in the mirror regions.

The potential configuration was also designed to have velocity compensation properties; i.e., ions with larger velocities penetrate deeper into the mirrors and spend more time there than slower ones, while they obviously spend less time in the central, field-free region. It is possible to adjust the profile of the potential so that the round-trip time is independent of velocity, i.e.,  $\Delta T_v \approx 0$ . Such compensation is well known in time-of-flight mass spectrometry, where Reflectron mirrors are used to improve mass resolution [17]. Using this potential profile, a total calculated time spread of  $\Delta T = 0.18$  ns is achieved [15]. Figures 2(d)–2(f) show the signal observed with the pickup for three different time windows:  $100 < t < 115 \mu\text{s}$ ,  $300 < t < 315 \mu\text{s}$ , and  $80000 < t < 80015 \mu\text{s}$ , respectively. It is clearly seen that the length of the bunch remains constant for much longer times, while its area decreases. The area decrease is due to the loss of ions by collisions with the residual gas leading to a finite lifetime of 164 ms.

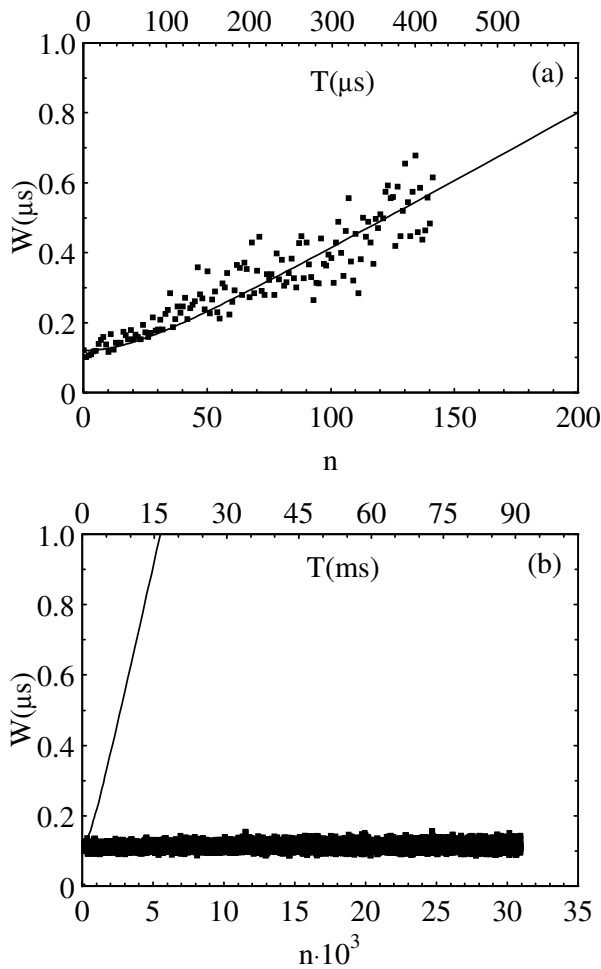


FIG. 3. Measured bunch width as extracted from the pickup signal as a function of the number of oscillations (bottom scale) or time (upper scale) for the two potential profiles in the mirror electrodes (a)  $\Delta T = 3.57$  ns and (b)  $\Delta T = 0.18$  ns. The solid lines represent the expected behavior from Eq. (2).

Figure 3(b) shows the evolution of the measured bunch length [note the change in the abscissa compared to Fig. 3(a)] under these new conditions. Also shown (solid line) is the expected bunch length calculated using Eq. (2) and  $\Delta T = 0.18$  ns. The calculated bunch length shows an increase with time which is much slower than for the first potential configuration [see Fig. 3(a)], but which is very much faster than the measured bunch length that remains constant over the 100 ms shown. The experimental result shown in Fig. 3(b) is totally unexpected: Although  $\Delta T$  has been reduced by a factor of 20 (from 3.57 to 0.18 ns), the complete lack of time dependence of the bunch length is in glaring contrast with the prediction for a system of noninteracting ions [Eq. (2)]. This behavior indicates that the ion motion is now synchronized, and we interpret this behavior as analogous to the clock synchronization phenomenon described earlier. If we consider each ion stored in the trap as an oscillator which interacts through Coulombic collisions with all the other ions (oscillators),

there are two reasons why synchronization is more likely in the second case than in the first. First, the density of ions at the turning points increases by a factor of 100 in the latter configuration (up to  $7.8 \times 10^8$  ions/cm<sup>3</sup>) enhancing the collision probability at these points. Second, the distribution of revolution frequencies is also much narrower; for the first potential configuration, the frequency spread of the ions was 0.12%, while it was reduced to 0.0061% in the second. Without Coulomb interactions, the particles would dephase, suggesting that it is the repulsive Coulomb interaction itself [that was assumed to be negligible in our previous description, Eq. (2)] which provides the coupling needed for the synchronization.

Convincing evidence that the Coulomb interaction is responsible for the observed effect was obtained by measuring the onset of synchronization as a function of the time the ion bunch spends at the turning points. For this purpose, we used a new set of potentials for which it is possible to change this time by varying  $V_1$  (see Fig. 1), without significantly altering the overall potential profile, except at the edges of the mirrors. Decreasing  $V_1$  from 6.5 to 4.3 kV lead to an increase of 160 ns or 6% in the revolution time. Simulations [15] demonstrated that the additional oscillation time was spent close to the turning points, where the ion density is the highest. While changing  $V_1$  the intrinsic time spread remains essentially constant  $\Delta T_i = 1.7$  ns, while the time spread originating from the velocity spread varies from  $\Delta T_v = 0.28$  ns at  $V_1 = 6.5$  kV to  $\Delta T_v = 0.46$  ns at  $V_1 = 4.2$  kV with a minimum  $\Delta T_v \sim 0$  ns at  $V_1 = 4.8$  kV. Hence, the major contribution to the total time spread is also  $\Delta T_i$  for these configurations. The transition to synchronization was observed when the value of  $V_1$  decreased below 4.9 kV, corresponding to an additional time spent close to the turning points of 82 ns. Basically, when  $V_1 < 4.9$  kV, enough time was given to the particles to interact, demonstrating that it is the repulsive Coulomb force which is responsible for the locking effect. It should be stressed that velocity compensation ( $\Delta T_v \sim 0$ ) is not essential for the existence of the synchronization phenomena.

The ion motion synchronization reported here is fundamentally different from the collective motion observed in Paul and Penning traps [18–20] since no parametric excitation is applied. In the present case, synchronization ensues from the collisional coupling between the ions in the mirror regions. As opposed to the pendulum clocks observed by Huygens, there is not a fixed phase relation between the synchronized ions; rather, their phases vary stochastically around a mean value.

A detailed description of the synchronization observed is outside the scope of this Letter. However, in a simplified picture the synchronization can be understood as a competition between two opposing effects. First, the ions inside the packet will tend to dephase according to the time spread for noninteracting ions ( $\Delta T$ ). Second, upon collisions in the mirror regions the ions will exchange energy

and momenta, and the oscillation times of the ions will be altered. The net effect of the ion-ion collisions is to redistribute the oscillation periods among the ions. Synchronization can occur if the collisional redistribution is much faster than the dephasing, i.e.,

$$P_c \gg \frac{\Delta T}{T}, \quad (3)$$

where  $P_c$  is the collision probability per oscillation and  $T$  is the average oscillation time.

An alternative approach to describe the observed synchronization is to consider the mean field approximation, where the average motion of the ion packet acts as an external force on each ion (oscillator) [21]. We have performed numerical calculations, where the classical equations of motion for a one-dimensional system made of a large number of Coulomb interacting particles are solved inside a flat-bottomed potential well bounded by walls with various slopes. The results of these calculations [16] show that phase synchronization between the particles occurs under certain conditions related to the density of ions, their relative initial velocities, and the slope of the walls, in qualitative agreement with the experimental results. However, if the Coulomb interaction between the particles is neglected, the packet of ions debunches much as in Fig. 3(a).

A major implication of the absence of dispersion observed in this experiment is to use the ion trap as a mass spectrometer [22]. It is well known that time-of-flight mass spectrometers (TOFMS) have a resolution which is limited by the length of the free flight region and the dispersion due to the velocity spread. The ion trap can be viewed as an extremely long (13 km, for a 4.2 keV Ar<sup>+</sup> ion beam stored for 100 ms) TOFMS, without debunching. It can also be compared to the more sophisticated Fourier transform ion cyclotron resonance mass spectrometer (FTICR MS) where very high mass resolutions are obtained due to the long observation time of the motion of ions in a uniform magnetic field, and the independence of the measured frequency on the initial velocity distribution of the ions. In a sense, the linear ion trap could transform a standard TOFMS into a system with mass resolution equivalent to that of a FTICR MS. A simple estimate of the resolution of our system can be obtained by curve fitting the peaks shown in Figs. 2(e) and 2(f). The average position of a peak, after  $t = 100$  ms of trapping, was fit with a standard deviation of  $\Delta t = 10$  ns. This leads to a mass resolution of  $\Delta m/m = 2\Delta t/t = 2 \times 10^{-7}$  for  $m = 40$ . Because the resolution increases with the trapping time, and trapping times of the order of tens of seconds are possible [23], we can expect very high frequency and thus

mass resolution. However, more work is needed to fully understand the synchronization effect before a definite statement about the mass resolution can be made.

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