Negative Mass Instability for Interacting Particles in a 1D Box: Theory and Application

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We demonstrate that the synchronization effect observed [Pedersen *et al.*, Phys. Rev. Lett. **87**, 055001 (2001)], when a bunch of ions oscillates between two mirrors in an electrostatic ion beam trap, can be explained as a negative mass instability. We derive simple necessary conditions for the existence of a regime in which this dispersionless behavior occurs and demonstrate that in this regime, the ion trap can be used as a high resolution mass spectrometer.

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The classical problem of interacting particles in a box has always been an interesting one as it serves as a model for various physical problems in the real world. Gases, liquids, and solids are examples of systems that contain many interacting particles. Particle collision experiments also have a long history; they have been the source of much of our understanding of the fundamental nature of particles and their interactions. These two areas are quite ideally combined when charged particles are trapped by electromagnetic fields.

In a recent experiment, Pedersen *et al.* [1,2] have demonstrated that, when a fast (keV) bunch of interacting particles oscillates between two electrostatic mirrors which form a stable trap [3–5], the bunch size can be kept constant, or self-bunching can occur, even though the particles do not have exactly the same velocity and in spite of the non-negligible Coulomb repulsion between them that should have rapidly spread the bunch. This counterintuitive behavior (called synchronization by Pedersen *et al.* [1,2], but which will be termed as self-bunching in this Letter) was tentatively explained as due to the interaction between the particles and the special kinematics properties of the potential well formed by the two electrostatic mirrors of the trap. Several empirical conditions needed for this behavior were given and were shown to be in agreement with the experimental observations. However, no formal treatment was given.

In this work, we present a formal criterion for the self-bunching to occur, and we show that the phenomenon belongs to a much larger class of behavior, called negative mass instability, which can be expected whenever classical interacting particles are bound by a soft wall potential. We also demonstrate that this effect can be exploited for high resolution mass spectrometry. A different, more microscopic view of the self-bunching is described in Ref. [6].

In order to solve the equation of motion for the particles in the trap, we choose, as suggested by Pedersen *et al.* [2], to represent the electrostatic trap as a one-dimensional potential well with sloping (soft) walls (see Fig. 1):

$$V(X) = \begin{cases} 0, & \text{if } |X| \leq L/2, \\ F(|X| - L/2), & \text{if } |X| > L/2, \end{cases} \tag{1}$$

where L is the length of the field-free region between the mirrors, and F is the electric field generated by the potential wall, representing the field produced by the mirror electrodes in the real trap. The problem to be solved is the motion of N charged particles of identical mass m and charge q. We represent the interaction between the various particles in the bunch using the mean field approximation [2], and we analyze the dynamics of a single (test) particle with mass m and charge q, relative to a homogeneously charged sphere of radius R_0 and of mass Nm, with a charge density ρ so that its total charge is $Q = Nq$, where $N \gg 1$.

The Hamiltonian for the system is given by

$$H = \frac{p_1^2}{2Nm} + \frac{p_2^2}{2m} + QV(x_1) + qV(x_2) + qU(x_1 - x_2), \tag{2}$$

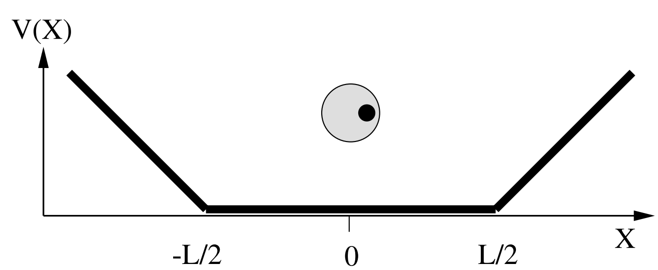

FIG. 1. The one-dimensional potential used in the model. The charged particle (black dot) is located inside the homogeneously charged (same sign as for the particle) sphere, and both move in the potential well.

where x_1 and x_2 are the coordinates of the sphere and test particles, respectively, p_1 and p_2 are their momenta, $U(x) = -\frac{\rho x^2}{6\epsilon_0}$ is the potential inside the sphere, at a distance x from its center, and ϵ_0 is the vacuum permittivity.

The equation of motion for the relative coordinates (x, p) between the sphere and the test particle can be obtained after separation of the center of mass coordinates:

$$\dot{x} = \frac{p}{\mu} \simeq \frac{p}{m}, \tag{3}$$

$$\dot{p} = -\frac{N}{N+1} xqV''(X) - kx \simeq -xqV''(X) - kx, \tag{4}$$

where $\mu = Nm/(N+1) \simeq m$ is the reduced mass, X is the center-of-mass coordinate, and $k = -\frac{\rho q}{3\epsilon_0}$.

These equations of motion can be solved exactly, but it is much more informative to develop first an approximate solution, from which a simple condition for self-bunching can be extracted. This solution (obtained under the impulse approximation, *vide infra*) is valid only when the force between the sphere and the test particle is repulsive ($k < 0$), which is always the case for an ion trap.

We solve the problem with the help of mapping matrices, which propagate the system in the (x, p) phase space, producing a Poincaré map at the center of the trap. The propagation of the particle-sphere system in the potential above can be divided into three sections: the field-free region, the mirror region, and the crossing point ($X = L/2$) between these two regions at each transit. The mapping matrices in these three regions are, respectively,

$$\mathbf{M_f} = \begin{pmatrix} 1 & \frac{t_f}{2m} \\ 0 & 1 \end{pmatrix}, \qquad \mathbf{M_s} = \begin{pmatrix} 1 & \frac{t_s}{m} \\ 0 & 1 \end{pmatrix}, \qquad \mathbf{M_k} = \begin{pmatrix} 1 & 0 \\ -\frac{2m}{t_s} & 1 \end{pmatrix}, \tag{5}$$

where t_f and t_s are the times spent in the field-free region and on the potential slope, respectively, so that the half period $T = t_f + t_s$.

A half cycle through the potential well (for particles without interaction) is expressed using the propagation sequence, starting from the center of the trap, moving above the wall, and back to the center, as:

$$\mathbf{M_T} = \mathbf{M_f} \cdot \mathbf{M_k} \cdot \mathbf{M_s} \cdot \mathbf{M_k} \cdot \mathbf{M_f} = \begin{pmatrix} -1 & \frac{t_s - t_f}{m} \\ 0 & -1 \end{pmatrix}. \tag{6}$$

Defining a slip factor

$$\eta \equiv \frac{t_s - t_f}{t_s + t_f} = \frac{|P_0|}{T} \frac{dT}{d|P_0|}, \tag{7}$$

where P_0 is the initial center-of-mass momentum, the propagation of n half oscillations is described by

$$\begin{pmatrix} x \\ p \end{pmatrix}_n = (\mathbf{M_T})^n \begin{pmatrix} x \\ p \end{pmatrix}_0 = (-1)^n \begin{pmatrix} 1 & \frac{T}{m^*} \\ 0 & 1 \end{pmatrix}^n \begin{pmatrix} x \\ p \end{pmatrix}_0, \tag{8}$$

where $m^* = -m/\eta$ is the effective mass.

From Eq. (8) it is clear that if the initial relative momentum p_0 is different from zero, the distance $|x_n|$ between the sphere and the test particle will increase with the number of iterations, unless the effective mass m^* is infinite, corresponding to $\eta = 0$. This condition is well known in time of flight mass spectrometers using Reflectrons (which are electrostatic mirrors) [7] and occurs when the additional time spent by the faster particles inside the reflector (as they penetrate deeper) is exactly compensated by the shorter time they spend in the field-free region.

The effect of the particle-sphere interaction can be included by integrating the relevant part of the equation of motion over a half-oscillation time T, assuming that the force is small and does not change much during this time (impulse approximation). This yields an additional change in the relative momentum $\Delta p = -kTx$ which can be written as a propagation matrix:

$$\mathbf{M_i} = \begin{pmatrix} 1 & 0 \\ -kT & 1 \end{pmatrix}. \tag{9}$$

This approximation is valid only when the particle-sphere force is repulsive ($k < 0$), as for an attractive force, an oscillatory motion is obtained, and the assumption that the force is approximately constant is incorrect. Multiplying Eqs. (6) and (9), we obtain the effective propagation matrix including the interaction (the multiplication order does not change the final condition for self-bunching),

$$\mathbf{M} = \mathbf{M_T} \cdot \mathbf{M_i} = -\begin{pmatrix} 1 - \frac{kT^2}{m^*} & \frac{T}{m^*} \\ -kT & 1 \end{pmatrix}. \tag{10}$$

Note that the determinant of this matrix $\det(\mathbf{M}) = 1$ (area preserving map [8]). The oscillations of the particle-sphere system in the potential well can be expressed by n multiplications of this matrix. Of interest for the present case are the conditions for which the relative motion in the phase space (x, p) is bound for any value of n, and the maximum distance between the center of the sphere and the test particle is smaller than the radius of the sphere.

Two conditions can be obtained to satisfy these requirements. In general, a bound trajectory in the (x, p) phase space will be obtained when $|\mathrm{Trace}(\mathbf{M})| < 2$ [8]. Applying this to Eq. (10) yields the following general stability condition:

$$0 < \frac{kT^2}{m^*} < 4. \tag{11}$$

The requirement that the maximum displacement of the test particle has to be smaller than the sphere radius R_0 can be obtained from the largest deviation in the x

coordinate in the Poincaré section produced by the mapping given in Eq. (10). This yields the following confinement condition:

$$x_{\max}^2 = \frac{x_0^2 + \frac{\eta}{m}(x_0 p_0 T - \frac{p_0^2}{k})}{1 + \frac{\eta k T^2}{4m}} < R_0^2. \tag{12}$$

Assuming that k is small [relative to $4m/(\eta T^2)$], which is the case relevant for both the experiment performed by Pedersen *et al.* [1,2] and the impulse approximation, and letting $x_0 \approx R_0/2$, Eq. (12) can be written as

$$\rho > \frac{4\eta\epsilon_0 p_0^2}{mqR_0^2}, \tag{13}$$

where we have also used the fact that $R_0 \gg 2\eta p_0 T/3m$.

The results obtained in Eqs. (11) and (13) demonstrate that the relative motion between the test particle and the sphere can, under certain conditions, be bound (self-bunching) and that such an effect is due to the combination of the ion-ion interaction and the special kinematics which is the result of the shape of the external potential $V(X)$. It can be shown that the exact dependence of the force on the particle-sphere distance is not critical and that other dependencies would yield similar propagation matrices, which under the condition that their traces are less than 2 yield an analogous condition to the one in Eq. (11).

For the inequality of Eq. (11) to be true when $k < 0$ (repulsive interaction), the effective mass m^* must be negative. Such a requirement is equivalent to [see Eq. (7)]

$$\eta > 0, \quad \text{or} \quad \frac{dT}{d|P_0|} > 0. \tag{14}$$

This result is similar to the condition stated by Pedersen *et al.* [see Eq. (26) in Ref. [2]], which was deduced empirically and was shown to be in excellent agreement with both the experimental data and numerical simulations. As explained in Ref. [2], the physical reason behind the kinematical condition expressed in Eq. (14) is that in such a case, the overall effect of the repulsive Coulomb interaction is to speed up the slower particles and to slow down the faster ones, keeping the bunch length constant.

Equation (13) demonstrates that there is a minimum density required for stabilizing a bunch of particles of size $\sim 2R_0$. The dependence of the minimum charge density on p_0 is as expected, i.e., if the momentum spread of the particles is large, a higher charge density is required, in agreement with the simulation performed in Ref. [2]. A numerical evaluation of Eq. (13) is consistent with the ion densities used in the experiments [1,2].

The equations of motion can be solved exactly for both a particle-sphere attractive or repulsive interaction. In such a case, the mapping matrices are rather complicated (more details will be given in a future publication), but it is possible to arrive at a compact form for the stability condition:

$$\left| \cos(\omega T) - \frac{\sin(\omega T)}{(1+\eta)\omega T} + 4\frac{\cos(\omega T) - \cos(\eta\omega T)}{(1+\eta)^2(\omega T)^2} \right| < 1, \tag{15}$$

where $\omega = \sqrt{k/m}$.

Figure 2 shows the stability condition graphically for various values of k/m as a function of ηT^2 [see Eq. (11)]. This parametrization allows one to separate the quantities depending on the particle properties (k and m) from the properties of the external potential (η and T). The hatched area represents the region of stability obtained under the impulse approximation as given in Eq. (11) (for $k < 0$ only), while the shaded area represents the region of stability as given by Eq. (15). Figure 2 shows clearly that for $k < 0$, the stability condition $\eta > 0$ holds, as no stable configurations can be found in the lower left quarter of the plot. It is also interesting to point out that for $k > 0$, the system can be unbound, even though the force between the particle and the sphere is attractive.

The self-bunching effect described above (for $k < 0$) is related to the so-called negative mass instability which was first introduced by Nielsen, Sessler, and Symon in 1959 [9] for relativistic circular accelerators or storage rings and extensively considered in a number of both experimental and theoretical works [10]. In these machines, the negative mass instability occurs when, for example, the angular velocity decreases with increasing energy, a situation occurring above the so-called transition energy. This effect has been, to our knowledge, observed only in accelerators working at relativistic energies, and the theoretical treatment has been implemented using the Vlasov equation [10].

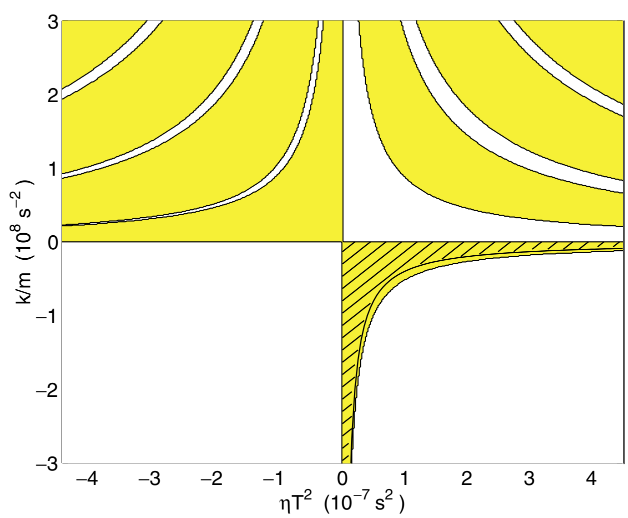

FIG. 2 (color online). Stability diagram for the system described in Fig. 1. The hatched area represents the stability condition as expressed in Eq. (11) (for $k < 0$ only); the shaded area is the stable (self-bunching) region, obtained from the exact solution, Eq. (15).

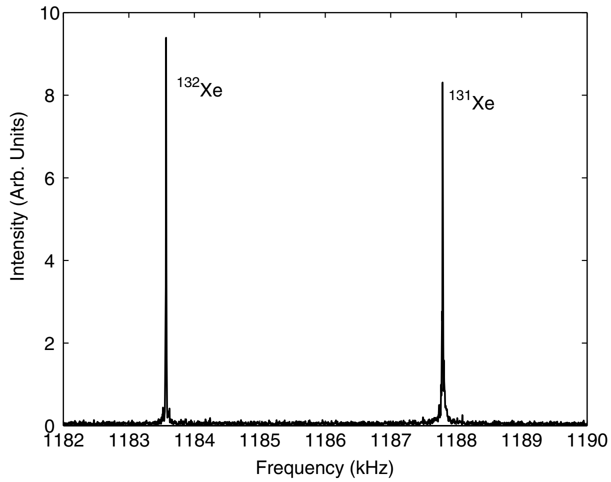

FIG. 3. Frequency spectrum obtained by fast Fourier transform of the pickup signal when a bunch comprising two isotopes of singly charged xenon ions is injected into the trap. Only the seventh harmonics are shown.

However, unlike the large accelerators where the negative mass instability is a nuisance, it leads to an interesting application in the electrostatic trap. The self-bunching effect, discovered by Pedersen *et al.* [1,2] and theoretically explained here, shows that it is possible to trap a bunch without its size changing with time. Since the oscillation frequency of the ions in the trap is proportional to $\sqrt{q/m}$ and the shape of the signal induced by the bunch on a capacitive pickup near the center of the trap is independent of time, a measurement of the oscillation frequency directly yields a mass spectrum of the stored ions, under the conditions given in Eqs. (11) and (13).

The application of our trap as a folded, time-of-flight (TOF) mass spectrometer, albeit without self-bunching, has been previously described [11]. A mass resolution of $\Delta m/m = 3.5 \times 10^{-4}$ for $m = 40$ was obtained. Figure 3 shows a frequency spectrum of data taken in the same instrument, in the self-bunching configuration. Two isotopes of xenon, $^{131}Xe^+$ and $^{132}Xe^+$, were injected into the trap with a kinetic energy of 4.2 keV. The signal from the pickup was recorded for 300 ms, and the Fourier transform in Fig. 3 shows the seventh harmonics. The width of the peaks (about 1 1/2 channels wide) is less than 4 Hz, which corresponds to $\Delta m/m = 2\Delta f/f \sim 7 \times 10^{-6}$, while the distance between the peaks is consistent with the mass difference between the two isotopes. As the detection efficiency of the capacitive pickup is mass independent, and trapping of very heavy species is possible in this electrostatic trap, such a system is well suited for heavy masses, where the detectors in standard TOF spectrometers are less efficient. In principle, the mass resolution is proportional to the measuring time, so that even higher resolution is feasible. In practice, however, there are several limiting factors. The most important one is the finite lifetime of the bunch in the trap (about 300 ms with the configuration described in Ref. [2]), which limits the overall measuring time to about 1 s at a background pressure of 5×10^{-10} T. In any case, the resolution obtained here is superior to the one obtained in standard TOF mass spectrometry [12] and approaches the values measured using the Fourier transform ion cyclotron resonance (FTICR) technique [13], which is considered to be the most precise technique of general use available today [14], but necessitates superconductor magnets to achieve high resolution. Additional studies are needed to understand the limit of the mass separation: Preliminary data show that when the mass difference between two species is very small, they tend to produce a single bunch under the self-bunching conditions, a phenomenon known in FTICR as well, and called peak coalescence [13].

As the theoretical model developed here is quite general, the negative mass instability is likely to be observed in other experimental setups where interacting particles are trapped and in motion in a nonharmonic potential. Moreover, the exact solution demonstrates that, even when the force between the particles is attractive, nontrivial behavior can be expected. The implications of this result to other fields, such as astronomy and planetary sciences, may be worth investigating.

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