Influence of space charge on the computed statistical properties of stored ions cooled by a buffer gas in a quadrupole rf trap

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We use an improvement of a three-dimensional model [Phys. Rev. A 27, 2321 (1983)], taking space charge into account for ions confined in a quadrupole rf trap. This model allows us to calculate the spatial and energy probability densities of strongly confined ions in the presence of a light buffer gas and shows the evolution of these properties as the total number n of ions increases. The ions are Ce^+ in the presence of dilute helium. For the chosen working point, the distributions remain Gaussian when n does not exceed $(2-3)\times 10^5$ ions at room temperature. The influence of the temperature on the importance of space-charge phenomenon is also studied.

I. INTRODUCTION

Ion storage techniques by rf quadrupole field trapping allow one to maintain an ionic population for long periods of time in the absence of collisions, wall effects, and magnetic field. In order to reduce parasitic effects caused by temperature, the ionic population can be cooled.[1] We have shown theoretically that elastic collisions with a light buffer gas in thermodynamic equilibrium induce behavior close to the state of thermodynamic equilibrium in the ions.[2] After a reorganization time the velocities and the positions of the ions follow a Gaussian distribution which oscillates at the driving frequency.[3] Nevertheless, it may be necessary to confine such a large number of ions that space-charge effects perturb the trap characteristics and it is imperative to know the limitations which can be produced. Our purpose here is merely the study of these perturbations and we will not treat strong space-charge effects but limit ourselves to threshold effects in what we shall call the "weak space-charge limit."

The first models[4–6] taking this phenomenon into account were established with the hypothesis that the parasitic field corresponds to a uniform spatial distribution of the ions in the trap.

These models predict that the values of the fundamental frequencies of the motion decrease when the total number of confined ions n increases. Recent experiments and theory[7,8] (in absence of collisions) have indicated the limitations of these models. Previously we have introduced the influence of space charge, taking into account collisions with a light buffer gas.[9,10] Assuming a Gaussian distribution, this model permitted us to show that the secular frequencies decrease when n is greater than 10^4 ions. Moreover because of the existence of a nonlinear term in the equation of motion, the secular frequencies depend on the amplitude of the movement and therefore on its energetic state. With the view to giving an account of the shift and the spreading of these frequencies we defined an "effective" fundamental frequency. This approach also allowed us to show that for some values of n the characteristic ellipses of the trajectories in phase space were almost conserved.

In this paper we will use the previously described formalism[3] to compute the energetic distributions of the ionic population subjected to the space charge. Lastly, we will discuss the critical influence of the temperature of the buffer gas on the appearance of modifications caused by space charge. The work is concerned with similar conditions as previously given. (The driving frequency is $\Omega/2\pi = 10^6/2\pi$ rad s^{-1}.) These conditions allow us to neglect wall effects and are always satisfied in the case of collisionally cooled populations. The partial pressure of the buffer gas does not exceed 10^{-4} Torr, so that ion-atom collisions occur approximately every hundred periods of the rf field. At $T=300$ K the volume occupied by the ions is close to 10^{-6} mm^3 so that ion-ion collisions are supposed to be improbable and are neglected. The threshold lies around an ion number n equal to 10^4 ions and the limit of the model's validity is reached when n is approximately equal to 2×10^5 ions.

II. METHOD

The generative equations have been previously presented in detail.[3] Let f_0 be the main component of the distribution of the position and the velocity of an ion at the instant t_0. In the Gaussian case, it can be chosen as

$$\frac{K}{8\pi^3\sigma_{sx0}^2\sigma_{sz0}\sigma_{vx0}^2\sigma_{vz0}}\exp\left[-\frac{1}{2}\left(\frac{x_0^2+y_0^2}{\sigma_{sx0}^2}+\frac{\dot{x}_0^2+\dot{y}_0^2}{\sigma_{vx0}^2}+\frac{z_0^2}{\sigma_{sz0}^2}+\frac{\dot{z}_0^2}{\sigma_{vz0}^2}\right)\right], \quad (1)$$

where K is a normalization factor and $\sigma_{sx0},\sigma_{sz0},\sigma_{vx0},\sigma_{vz0}$ are the components of the spatial and velocity dispersions. In the case of the weak space charge, we cannot suppose that the dispersions remain simply dependent on the coefficients of the motion. So, it is necessary to find two classes of parameters to know the distributions and therefore to use two equations:

$$\int [g(\vec{x}_0,\dot{\vec{x}}_0)-g(\vec{x}_k,\dot{\vec{x}}_k)]f_0(\vec{x}_0,\dot{\vec{x}}_0)d\vec{x}_0 d\dot{\vec{x}}_0=0 \,, \tag{2}$$

$$\int_{t_0}^{t_0+T_m} \int f_0(\vec{x}_0,\dot{\vec{x}}_0) \int f_{V_2}(\vec{v}_2)\,|\vec{v}_2-\dot{\vec{x}}(t)|\,[f_C(\vec{c})g(\vec{u}'_k)d\vec{c}-g(\vec{u}_k)]d\vec{x}_0 d\dot{\vec{x}}_0 d\vec{v}_2 dt=0 \,. \tag{3}$$

g is a function of the position and the velocity, $\vec{v}_2$ represents the velocity of the buffer gas, $\vec{c}$ the impact parameter (collisions are assumed to follow hard-sphere model). The subscript k denotes the values at the instant $t_k=kT_m$, where T_m is the period of the driving frequency. The solution of these equations gives the values of the dispersions. $\vec{\sigma}_{v0}$ and $\vec{b}=\vec{\sigma}_{v0}/\vec{\sigma}_{s0}$ are well suited to the research of the solution. It is necessary to add in the system of the motion equation a coupling term equal to

$$(e/m)\vec{\nabla}V(t)$$

with

$$V(t)=\frac{ne}{4\pi\epsilon_0}f_{X(t)}(\vec{x}(t))\otimes\frac{1}{|\vec{x}(t)|} \,,$$

where e/m is the charge to mass ratio of the ion. The potential $V(t)$ represents the field created by the ionic charge due to the Coulomb interactions.

At any time, $\vec{\sigma}_s(t)$ and $\vec{\sigma}_v(t)$ can be computed from the integration of the trajectories corresponding to given initial conditions and ionic density. The confirmation of the Gaussian model is obtained by using relations (2) and (3) for different functions g; the results, in each case, must be independent of g.

When a weak space-charge field is present the trajectories in the phase space are not elliptic but can be locally approximated by such curves. It is thus possible to compute the individual fundamental frequencies ω_i for a given time and initial conditions. The effective frequencies are finally obtained from the average values of the samples. (Note that our first definition of this parameter was only valid in the adiabatic approximation.[10])

For instance,

$$\omega_{ix}=\frac{1}{t_m}\arccos\frac{Bx_0x_1+\dot{x}_0\dot{x}_1}{Bx_0^2+\dot{x}_0^2}\,,\quad B=\frac{\dot{x}_1-\dot{x}_0}{x_1x_0}$$

and

$$\omega_x^*=E(\omega_{ix}) \,.$$

III. RESULTS

Numerically the two equations cannot be treated simultaneously since solving for one parameter requires a knowledge of the other and moreover the knowledge of $\vec{\sigma}_s(t)$. We use, therefore, an iterative method. The initial values introduced in the equations being those found for a number of ions $n'<n$, the parameters being initialized with $n=0$, i.e., without space-charge effect.

We have explained[3] that the distribution is not Gaussian if the value of the parameters depend on the chosen function g and we have decided to take for the limit of the Gaussian representation the condition that the different values of b_x, b_z do not differ from each other by more than 10%.

The search for b_x and b_z is made with 500 samples, thanks to a variance reduction method. A greater number is needed to obtain a precise value for σ_{vx0} and σ_{vz0}. Finally, we compute the time dispersions with 1000 samples.

The method has been used for values of n lying between 10^3 and 6×10^5 ions. The spatial dispersions are close to 2 (for z) or 4 (for x and y) mm, which gives an idea of the volume occupied by the ions.

When n does not exceed 10^4 ions the parameters are not greatly perturbed. Figure 1 shows the evolution of b_x and b_z with n and for different g functions. When n is approximately in the range $(2-3)\times10^5$ ions, the values obtained for each g function are different by more than

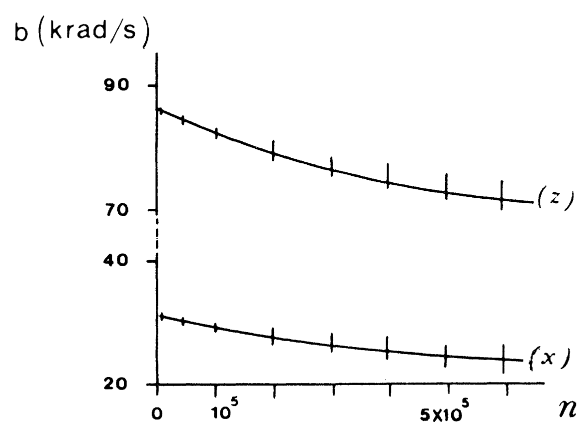

FIG. 1. Evolution of the parameters $b: b_x=\sigma_{vx0}/\sigma_{sx0}$ and $b_z=\sigma_{vz0}/\sigma_{sz0}$ with n. For $n>2\times10^5$ the method no longer allows one to obtain $\vec{\sigma}_{s0}$ from $\vec{b}$ then; the computed values depends on the g functions. (Functions used: $|x|, |z|, |\dot{x}|, |\dot{z}|, x^2, z^2, \dot{x}^2, \dot{z}^2, |x^3|, |z^3|, |\dot{x}^3|, |\dot{z}^3|$.)

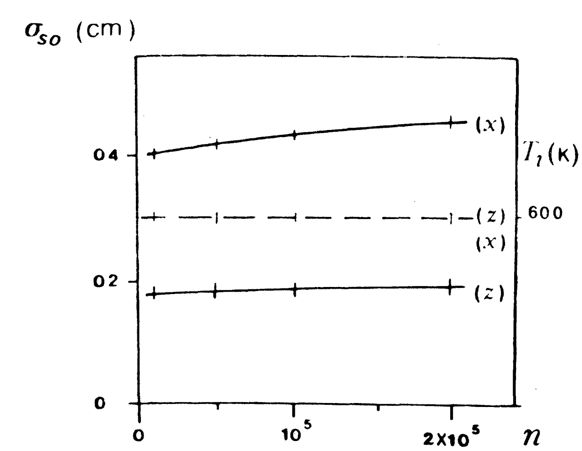

FIG. 2. Evolution of $\sigma_{sx0}, \sigma_{sz0}$ (T_{x0}, T_{z0}, respectively) in the area of validity of the model. σ_{sx0} and σ_{sz0} increase with n, whereas the ionic temperatures T_{ix} and T_{iz} remain constant.

10%. On the other hand, for the same values of n, σ_{vx0} and σ_{vz0} remain constant, so that it is possible to represent the spatial disperions σ_{sx0} and σ_{sz0} as functions of n (Fig. 2). Since the model is based on a perturbation method it allows one to demonstrate the advent of the strong space-charge regime from the determination of the value of n at which the model breaks down. In fact, when n increases, the shape of the spatial distribution evolves and begins to evidence repulsive behavior, i.e., the distribution tends to spread.

When n is less than 2×10^5 ions, the shape of the ionic cloud remains approximately constant, because, firstly, the ratio $\sigma_{sx0}/\sigma_{sz0}$ does not depend on n, and secondly, temporal variations of the energetic and spatial distributions are not changed and stay periodic at the frequency of the driving field. As in the space-charge-free case, the relative amplitude of the variations with time is always of the order of q for the spatial dispersions and 0.7 for the velocity dispersion,[3] the influence of the outer source is not modified here.

We also find that the energetic properties stay constant while the model is valid. For these values of n, the shape of the velocity distribution remains rigorously Gaussian.

The associated temperature is independent of n which means that the kinetic energy of each ion is constant. This property should be kept for larger values of n. The effective frequencies have also been computed and they are represented in Fig. 3. The indicated dispersions show that each trajectory corresponds to a different value of the individual frequency. The decrease of the values indicates that space charge moderates the motion. Calculations of some trajectories, in phase space, using the results found above show the evolution of the individual properties of the ions (Fig. 4). The trajectories practically retain, except for some small perturbations, their elliptic character.

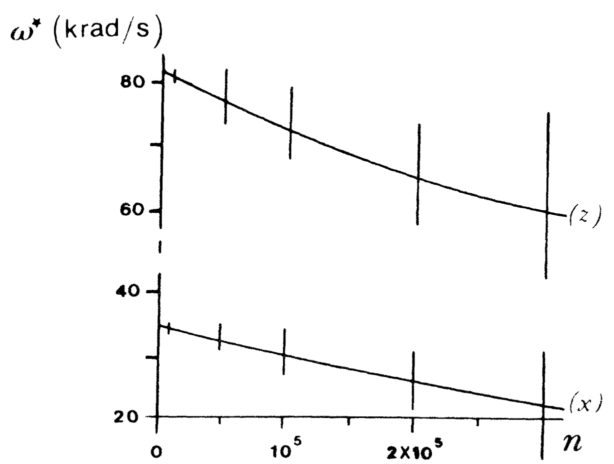

FIG. 3. Evolution of the effective fundamental frequencies with the total number of ions n. Above the validity threshold it is not possible to compute them because we do not know how to obtain a sample which represents the population. Near the threshold these frequencies are strongly reduced by the effects of space charge.

IV. INFLUENCE OF SPACE CHARGE ON COLLISIONAL COOLING

We know[3] that in the absence of space charge, the mean ionic temperature becomes of the order of twice the one of a light buffer gas introduced into the trap once the reorganization time has elapsed.

In this section we present the n value of the threshold of the Gaussian model for different values of the temperature T. When T decrease, the variation of b vs n is translated and the threshold of the strong space-charge regime appears at smaller and smaller values of n. The computed values show that the ionic temperature and the temporal variations of the parameters always remain constant of the properties of the ionic population. In Fig. 5 the variations of $\vec{\sigma}_{s0}$ and ω^* with T are given. Figure 6 shows the magnitude of the evolution of b with n against T. The shape of the phenomenon is governed by the fact that the kinetic energy of the ions is less when the ionic temperature decreases and so each ion is more sensitive to the repulsive potential exerted by the others.

V. CONCLUSION

We have presented a method which shows how the statistical properties of the ions are modified when space charge is not negligible ($n>10^4$ ions for $T=300$ K). For

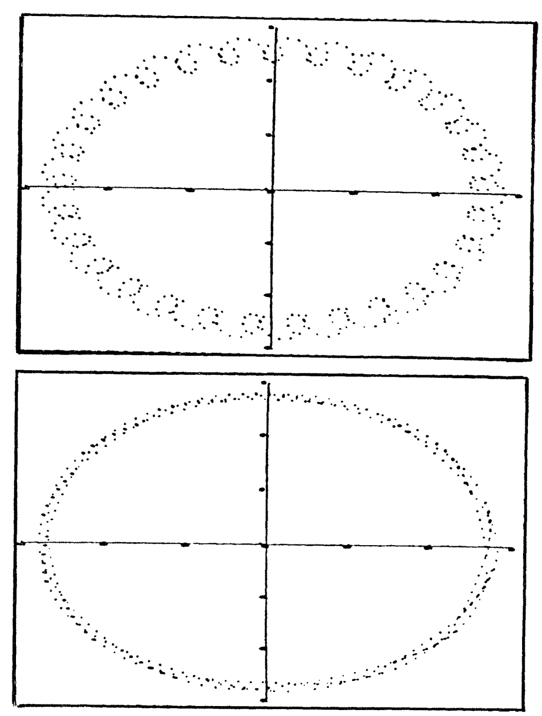

FIG. 4. Representation of the ionic motion in phase space at multiple instants of T_m in the case of perturbed trajectories (very large amplitude of the motion, event very unprobable). Duration of the integration: $400T_m$. $n=10^5$ ions. In the less perturbed case the spreading of the curves are more narrow; the same perturbations are observed for greater ion numbers and for more probable initial conditions. Top to bottom: movement along x, along z.

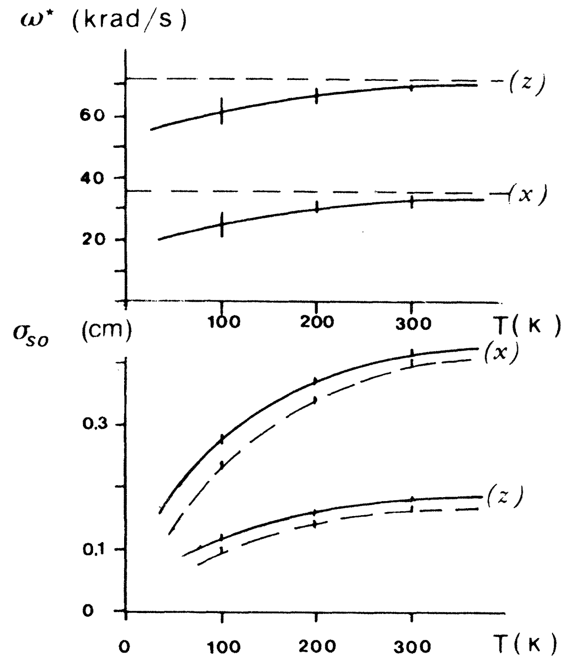

FIG. 5. Evolution of the parameters $\sigma_{sx0},\sigma_{sz0},\omega_x^*,\omega_z^*$ with T, for $n=5\times10^4$ ions [dashed lines correspond to the space-charge-free case ($n=0$)].

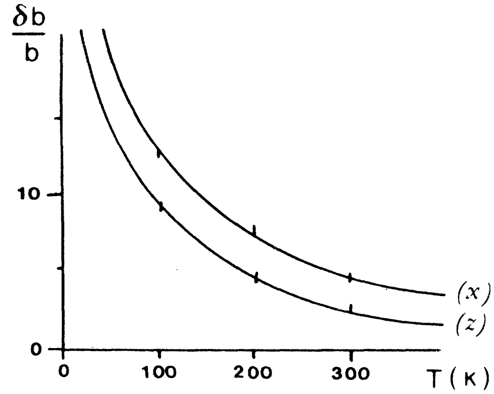

FIG. 6. n dependence of the parameters with T. Evolution of the relative variation of b: $\delta b/b=(b_{\text{“}n=0\text{”}}-b_{n=5\times10^4})/b_{\text{“}n=0\text{”}}$.

the first time we show how space charge modifies the individual mechanistic properties, the mean spatial dispersion of the ionic population, the shape of the spatial distribution, and the fundamental frequencies, oppositely to the energetic properties. The method is a perturbation method and therefore presents some limitations. However, it permits us to define a domain of n values ($n\le2\times10^5$ ions at $T=300$ K for which the properties are only quantitatively modified (weak space charge).

It is also possible to obtain a threshold, i.e., a value of n for which the properties are perturbed qualitatively (appearance of the strong space-charge regime). This threshold is a very acute function of the temperature of the ionic cloud. It will be interesting to apply this model to the study of the influence of the working conditions (a,q,Ω) on the n values in the weak space-charge regime. The method could be refined by the introduction of a spatial distribution which describes the spreading of the cloud in the presence of space charge. This could be done by defining a two-parameter distribution. This will make already rather cumbersome calculations notably heavier, but will allow one to see whether or not the character of the energetic distribution remains Gaussian in the strong space-charge regime. Finally, this technique could be equally applied to ions cooled by other methods such as "laser cooling."

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