International Journal of Mass Spectrometry 364 (2014) 16–20

Contents lists available at ScienceDirect

International Journal of Mass Spectrometry

journal homepage: www.elsevier.com/locate/ijms

Space charge and collective oscillation of ion cloud in a linear Paul trap

P. Mandal*, S. Das[1], D. De Munshi[1], T. Dutta[1], M. Mukherjee[1]

Raman Center for Atomic, Molecular and Optical Sciences, Indian Association for the Cultivation of Science, 2A&2B Raja S.C. Mullick Road, Kolkata 700032, India

ARTICLE INFO

Article history:
Received 18 December 2013
Received in revised form 4 March 2014
Accepted 18 March 2014
Available online 29 March 2014

Keywords:
Linear Paul trap
Nonlinear resonance
Space charge
Collective oscillation

ABSTRACT

Charged particles confined in an ion trap is still the simplest system to study the effects like anharmonicity, parametric oscillation, bi-stability and in addition space charge induced modification to all the above effects. Some of these effects like space charge induced frequency shift and emergence of collective oscillation has been observed in a linear Paul trap. These results though similar to earlier observations made on a hyperbolic Paul trap, they differ in some finer details which has been addressed in this article. A simplistic model, limited to the adiabatic approximation, has been utilized to support the observation made in the experiment.

1. Introduction

Ion trap finds numerous applications in physics, chemistry, biology and engineering. In some cases a few ions are used to carry out high precision experiments [1–4] while in other cases high density of ions are required to store in the trap [5]. A single ion in an ideal trap exhibits in-phase harmonic oscillations with characteristic secular frequency. However, the Coulomb repulsion between the trapped ions, even due to the presence of a second ion, alters the trap potential. Thus the physics of space charge, relevant to applications of trapped ions, deserves detail understanding. The problem has been studied mostly empirically by Paul et al. [6], and Fischer [7]. In spite of numerous studies on space charge and its effect on the trapped ions [8–13], some of the observed phenomena are yet to be understood from ab-initio theory. This includes collective oscillation that has been observed both in Penning [14] as well as in Paul trap of hyperbolic structure [15] which, from now onwards, we will call by 3D-QIT meaning 3D quadrupole ion trap in order to differentiate it from linear quadrupole ion trap (LQIT). In this article we report on space charge in a linear Paul trap and its effect on the ion dynamics in two different manifestations. First, it modifies the individual ion oscillation frequency due to the modification of the trapping potential similar to that observed in 3D-QIT [16]; second, under certain experimental condition of high charge density the ions behave as a single collective body showing motional frequency independent of the number of ions. Unlike the observation made in 3D-QIT [16], we for the first time observe vanishing signature of the nonlinear resonances (NLR) of individual ion oscillation with increasing ion density. In respect to the observed collective oscillation of the radial motion, our result shows similar behaviour as that was observed earlier in the collective oscillation of the axial mode of Ref. [15]. In addition to the earlier observed criticality of the excitation amplitude, we have observed criticality of the collective oscillation on ion number which needs to be explained theoretically. It is pertinent to compare the experiment made on a 3D-QIT with that of a LQIT due to the presence of less anharmonicities in later. As we will show here that such effect indeed modifies certain results observed earlier in a hyperbolic trap. We have utilized a simplistic model based on uniform spatial distribution of trapped ion to account for the space charge effect and show that it, within the adiabatic approximation, explains well the observed shifts of the individual ion oscillation frequency as consistent with the literature [17–19].

In Section 2, the relevant ion trap theory will be briefly reviewed followed by Section 3. Section 4 contains the experimental data and analysis with the space charge model. The observation of collective oscillation of the radial modes in a linear Paul trap and its criticality with respect to the ion number, have been shown in this section.

2. Theory

A linear Paul trap uses a radio frequency (rf) potential in addition to a dc potential for providing dynamical trapping of charged

* Corresponding author. Present address: Department of Physics, St. Paul's Cathedral Mission College, 33/1 Raja Rammohan Roy Sarani, Kolkata 700009, India. Tel.: +91 8902442561.
E-mail address: pintuphys@gmail.com (P. Mandal).
[1] Present address: Centre for Quantum Technologies, National University of Singapore, Singapore 117543, Singapore.

http://dx.doi.org/10.1016/j.ijms.2014.03.010

particles in the radial plane and a dc potential for axial confinement. The radial potential in terms of the radial coordinate (r, ϕ), can be expressed by [20]

$$\Phi(r,\phi,t) = \sum_k a_k \left(\frac{r}{r_0}\right)^k \cos k\phi (U - V_0 \cos \Omega t), \tag{1}$$

where r_0 is the distance of an electrode from the trap center, U is the dc potential, V_0 and $\Omega/2\pi$ are respectively the amplitude and frequency of the rf. Here a_k signifies the weight factor for kth order multipole. The equation of motion of a trapped ion corresponding to the quadrupole part ($k=2$) of the potential in Eq. (1) is given by the Matheiu's differential equation as,

$$\frac{d^2u}{d\zeta^2} + (a_u - 2q_u \cos 2\zeta)u = 0, \tag{2}$$

with $u = x, y$, where

$$\begin{aligned} a_x &= -a_y = \frac{4eU}{mr_0^2\Omega^2}, \\ q_x &= -q_y = \frac{2eV_0}{mr_0^2\Omega^2}, \end{aligned} \tag{3}$$

and $\zeta = \Omega t/2$. Solution of Eq. (2) shows that the ion oscillates with two different frequencies [21]: one equal to the frequency of the applied rf and is called micromotion while the other with a frequency $\omega_{0u} = \beta_u \Omega/2$ and is called the secular motion. Here $\beta_u \approx \sqrt{a_u + q_u^2/2}$ within the adiabatic approximation, i.e. for small a_u and q_u [21].

The trapped ion performs in-phase simple harmonic oscillation due to the quadrupole potential but gains energy from higher order multipoles that enhances their motional amplitude. The ion motion gets resonantly excited when the frequency of oscillation at a given operating parameter (a_u, q_u) satisfies the following condition [22–24]

$$n_x\omega_{0x} + n_y\omega_{0y} = \Omega, \tag{4}$$

where n_x, $n_y = 0, 1, 2, 3\ldots$ and $n_x + n_y = k$ and $k > 2$. If one of the trap parameters is varied, it will modify the β value and hence the secular frequency. Thus a nonlinear resonance appears at a definite value subjected to the condition defined by Eq. (4) and results in narrow instabilities within the broad stability diagram [25]. As the trap operating parameters get modified due to numerous factors like, the coupling between the axial and radial potentials [26] and space charge due to the trapped ions [16], the NLR over q-parameter-space shifts. The amplitude of such resonances is determined by the strength of the corresponding multipole which strongly depends on the geometry of the setup. The geometry dependence of higher order multipoles has been studied in detail in mass filters [27,28] and linear traps with cylindrical rods [29]. Here we discuss the effect of space charge on NLR of an ion cloud in LQIT of particular geometry described in Section 3.

A simple model has been employed to explain the observed shift in the nonlinear resonance as reported in this article as well as other experiment reported elsewhere [16]. In addition to the applied trapping potential, an ion experiences the space charge potential developed by all other trapped ions. Since the space charge potential is developed by the trapped ions themselves, the potential in the vicinity of an ion grows with the density of trapped ions (ρ). In this model, it is assumed that the space charge potential is proportional to the density of trapped ions and a quadratic function of the position coordinate as consistent with Poisson's equation. Thus the space charge potential in the radial plane can be represented by

$$\Phi_s = -\kappa\rho(x^2 + y^2), \tag{5}$$

where κ is a constant. Negative sign in Eq. (5) is justified by the positive type charge in space. Incorporating the space charge potential in the applied trapping potential (Eq. (1)), the equation of motion of an ion in the radial plane can be written following Eq. (2) as

$$\frac{d^2u}{d\zeta^2} + (a'_u - 2q_u \cos 2\zeta)u = 0, \tag{6}$$

where

$$a'_u = a_u - \frac{8\kappa\rho e}{m\Omega^2}. \tag{7}$$

The definition of the β parameter, within adiabatic approximation, is modified by a'_u and can be redefined by

$$\beta'_u = \sqrt{a'_u + \frac{q_u^2}{2}}. \tag{8}$$

Thus the secular frequency of the trapped ion decreases [30] as β parameter is effectively decreased due to the space charge effect. In order to match the secular frequency in presence of the space charge, the NLR shifts to higher value of q. If it shifts to a new position q' in presence of the space charge, it is given by

$$q' = \sqrt{2(a - a') + q_0^2}, \tag{9}$$

where $q' = q_0$ is the NLR center in absence of the space charge effect. From Eqs. (7) and (9)

$$q' = \sqrt{\frac{16\kappa\rho e}{m\Omega^2} + q_0^2}. \tag{10}$$

3. Experiment

The experimental setup consisting of an ionization setup, a linear Paul trap, extraction and detection setup is shown schematically in Fig. 1 and described in detail in Ref. [31]. The linear trap is assembled with four three-segmented cylindrical rods each of diameter 10 mm with a separation of 8 mm between two opposite electrodes ($2r_0$). The middle electrodes are taken 25 mm and end electrodes are 15 mm in length. The molecular nitrogen ions (N_2^+) are created from the background gas at pressure 10^{-7} mbar by electron impact ionization inside the trap. The accelerating potential is applied in the filament for time duration of few milliseconds to few tens of milliseconds, called the creation time (T_c) hereafter. The rf at $\Omega/2\pi = 1.415$ MHz is applied between diagonally connected electrode pairs in the middle while the end electrodes are set at dc potential of 20 V during trapping. However, the middle electrodes are kept at dc ground ($U = 0$) and hence $a_u = 0$. The ions are dynamically trapped for few hundreds milliseconds to seconds before they are extracted by lowering the axial potential in one direction. The extracted ions are guided by a cylindrical electrode floated at −200 V and accelerated towards the channel electron multiplier (CEM) detector, placed 12 cm away from the ion guiding cylinder and biased at −2.5 kV. As shown in the schematic (Fig. 1), the CEM output is processed successively by a preamplifier, amplifier cum discriminator and converted into TTL pulse before fed into a multichannel scalar (MCS) card that finally produces the time-of-flight (ToF) spectra. Typical TOF in our setup is 8 μs with full-width at half-maxima of 12 μs and this is well within the saturation limit for a typical CEM detector. The ions of different charge-to-mass ratio can be identified from the ToF [16].

The operating parameter q is varied by changing the rf amplitude while keeping the ion creation time, ion storage time and all other parameters constant. The experiment at a given q is repeated 100 times and the total number of trapped ions is counted. As expected, narrow NLRs appear within the stability diagram due to the presence of higher order multipoles of the potential in the trap. The observed NLRs in our setup correspond to the 6th, 7th and 8th order multipoles. It should be noted that the geometry of the trap setup