Electrostatic bottle for long-time storage of fast ion beams

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A technique for storage of fast-ion beams (keV) using only electrostatic fields is presented. The fast-ion trap is designed like an optical resonator, whose electrode configuration allows for a very large field-free region, easy access into the trap by various probes, a simple ion loading technique, and a broad acceptance range for the initial kinetic energies of the ions. Such a fast-ion storage device opens up many experimental possibilities, a few of which are presented. [S1050-2947(97)50803-1]

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Ion trap techniques have been applied to a variety of experiments in the fields of physics and chemistry; in particular, high precision laser spectroscopy, mass measurements, and studies of particle dynamics [1]. In most cases, the ions are stored with very low kinetic energy (between 0 and a few eV), in a combination of electrostatic and magnetostatic or time-dependent fields. The ions are confined in a relatively small region of space, and their trajectories are usually a complex function of the fields and of the geometry of the electrodes.

In the past few years, heavy-ion storage rings have provided another means for trapping ions [1,2]. In these large-scale devices, ions are stored with a high kinetic energy (MeV's) using a combination of magnetic steering and focusing fields. These rings have mainly been used for the study of interactions between ions and electrons (using the so-called electron cooler), laser cooling, and beam dynamics, as well as spectroscopy. For molecular ions, the storage ring technique allows for complete relaxation of the energy initially stored in the vibrational and rotational degrees of freedom [3,4]. Such relaxation is important in studies of low-energy molecular ion reactions with electrons, atoms, and molecules, as well as in laser spectroscopy, because in these cases the reaction rates are known to depend strongly on the initial population of the rotational and vibrational states. The energy stored in these degrees of freedom is radiatively released while the beam is stored (for dipole-allowed transitions, the typical time scale is milliseconds to seconds for vibrational relaxation, and seconds to minutes for rotational relaxation). Among the advantages of the storage ring is the fact that the beam has a well-defined kinetic energy and direction in space, allowing for highly efficient detection techniques of reaction products (for example, molecular fragment imaging as in molecular ion recombination studies [5]). Such a detection scheme is usually unavailable with the ion trap technique. However, storage rings are large-scale (and expensive) machines that require high-energy accelerators for producing fast beams, and access time to users is limited.

In the following, we present a fast-ion storage device based purely on electrostatic fields. This electrostatic trap combines the advantages of the ion traps (inexpensive, small, and simple to operate) with the advantages of the storage rings (beams with well-defined direction and energy and the possibility of single-particle counting). The present device was designed with the initial goal of performing laser-molecular ion beam experiments [6] on fully relaxed molecular ions, i.e., molecular ions in their ground state. Furthermore, time-dependent ion spectroscopy over a time scale (seconds to minutes) that is out of reach in standard experimental setups (where the interaction time is limited by the length of the experimental apparatus, i.e., a few μs) will be feasible. In such experiments, the molecular ions are continuously photodissociated, and transitions leading to the breakup of the molecular ions are monitored as a function of storage time, i.e., as a function of the internal population of the rovibrational states. Such measurements are of importance for the field of interstellar molecular cloud modeling [7]. A few examples of experiments feasible with this fast-ion storage device will be discussed below.

Figure 1 shows a general schematic view of the electrostatic bottle ion trap. The ions are created in an external ion source (not shown), accelerated up to an energy of a few keV, and mass selected. After focusing and collimation, the beam is directed into the ion trap along its axis. The trap is made of two cylindrically symmetric ''electrostatic mirrors,'' each of which is made up of a stack of cylindrical electrodes which both trap the beam in the longitudinal direction and focus the beam in the lateral direction. Upon injection, the entrance set of electrodes is grounded so that the beam can reach the exit set of electrodes. The potentials of these electrodes are set so that the beam is stopped, reflected, and focused. When the reflected beam reaches the entrance electrodes, the potentials of these are rapidly switched on (about 100 ns rise time) to the same values as for the exit electrodes. For a proper choice of voltages, the ions are trapped between the two mirrors, and they bounce back and forth, the lifetime being limited by the residual gas pressure present in the trap.

The electrostatic cylindrical mirrors are made of five electrodes each (labeled $E1$ to $E5$ in Fig. 1) that produce the retarding field. Electrode $E1$ is grounded, while electrode $E5$ is at a potential V_s, which is high enough to stop the ions.

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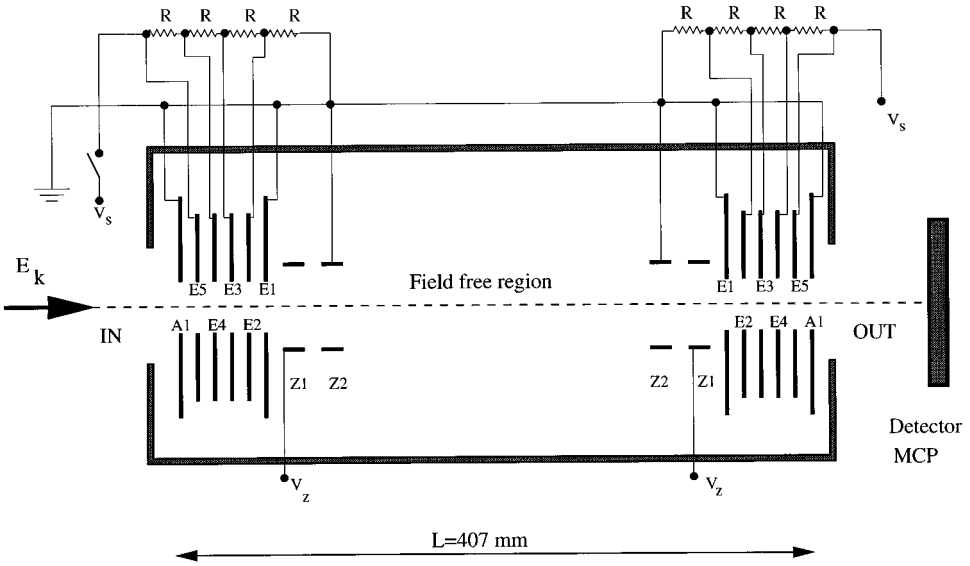

FIG. 1. Schematic view of the ion trap. The ion beam is injected from the left, when the entrance electrodes are grounded. The three electrodes $E5$, $Z1$, and $Z2$ form an asymmetric Einzel lens, which is used for focusing the ions. Neutral particles escaping the trap are monitored by a microchannel plate detector downstream. The drawing is not to scale.

The potential V_s is divided linearly across the electrodes. An additional grounded electrode ($A1$) closes the stack of electrodes to reduce the electric fields outside the trap. Two additional electrodes (labeled $Z1$ and $Z2$) act as focusing electrodes, where $Z1$ is at a potential V_z and $Z2$ is grounded. As such, these two electrodes, together with electrode $E5$, act as an asymmetric Einzel lens. Thus, for a given geometry, the ion trap is characterized by only two parameters, which are the stopping and focusing potentials, V_s and V_z, respectively. More technical details will be given in a forthcoming publication [8].

The main question is to determine the values of V_s and V_z for which the ions are trapped between the two mirrors. It is well known that many principles of geometric optics can be applied to ion optics. The above system is very much based on the same principle as for an optical resonator made of two equivalent mirrors. For an optical resonator working with a Gaussian beam, the stability criterion is related to the focusing properties of the mirrors [9]:

$$0 \leqslant \left(1 - \frac{L}{2f}\right)^2 \leqslant 1, \tag{1}$$

where f is the focal length of the two mirrors and L is the effective distance between them. Equation (1) is equivalent to

$$\frac{L}{4} \leqslant f \leqslant \infty. \tag{2}$$

Thus, the stability condition (for a beam close to the axis of symmetry) requires the focal length of the mirrors to be larger than some critical portion of the trap length, a property that is easy to fulfill with the above design. Also, the value of the stopping potential has to be high enough to confine the ions longitudinally, i.e., the condition $qeV_s > E_k$ has to be fulfilled, where q is the charge of the ions and E_k is their kinetic energy. The focal length of the electrostatic mirrors of the trap was found as a function of the focusing potential by computing particle trajectories using SIMION (Ver. 6) [10]. As an example, for a beam of 4-keV singly charged ions and $V_s = 6.5$ kV, the range of values for which Eq. (2) is valid was found to be $3.13 < V_z < 3.5$ kV for a beam diameter up to 3 mm (more details will be given in a subsequent publication [8]). A few aspects of the above design merit particular attention.

First, the trap is completely electrostatic, a feature that seems *prima facie* in contradiction with the so-called ''nontrapping'' theorem (the Earnshaw theorem [11]), which forbids trapping of charged ions using purely static fields. However, this is valid only if the kinetic energy of the ions is zero. In the present case, the field is changing in the frame of reference of the ions due to their kinetic energy.

Second, because the trap is electrostatic, the trapping is only energy (E_k) and charge (q) dependent (in fact, E_k/q). This is different from the high-energy storage ring devices, where the magnetic rigidity of the dipole magnets limits the maximum mass of the ions that can be stored at a given energy. Furthermore, one can store simultaneously different ions with the same E_k/q ratio, enabling the studies of ion-ion collisions in the trap, or its use as a part of a mass spectrometer.

Third, the central part of the ion trap, which can be made as short or long as needed [see Eq. (2)], is field free: Because the electrodes Z_2 are grounded, this region is shielded from the electrostatic fields of the mirrors and Einzel lenses. In this region, the ions travel in straight lines with their injection energy. This is different from the Kingdon trap [12], which is also an electrostatic trap, but where ions orbit around a charged wire, always in the presence of an electrostatic field. The fact that the ions travel in straight lines in the central region is very useful for experiments where merged- or crossed-beams configurations are required.

The lifetimes of various stored ions were measured using a microchannel plate detector located beyond the exit electrodes of the ion trap (see Fig. 1). At a few keV, the most

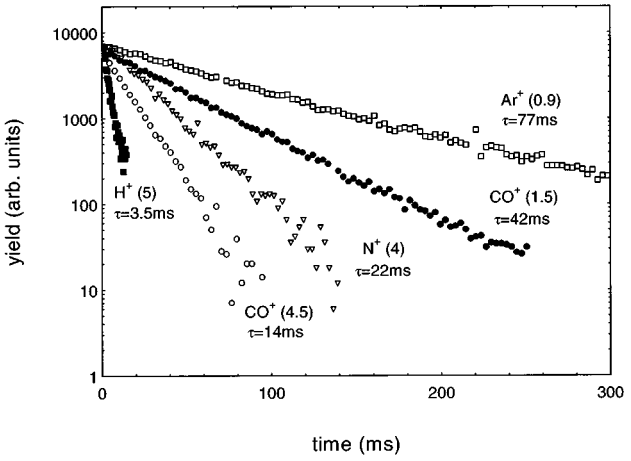

FIG. 2. Count rate as a function of storage time as measured by the microchannel plate detector for 4-keV Ar^+, CO^+, N^+, and H^+. The lifetime obtained by fitting the data using a single exponential decay for each of these species is indicated as well as, in parentheses, the residual pressure in units of 10^{-8} Torr.

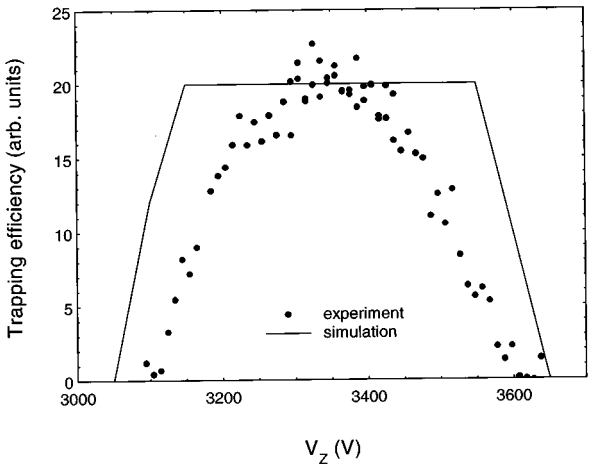

FIG. 3. Trapping efficiency, i.e., the number of 4-keV Ar^+ ions stored as a function of the Einzel lens potential V_z. The lifetime was found to be independent of V_z. The solid line is the result of a classical trajectories calculation.

important ion loss process in the trap is due to electron capture by the charged ions from the residual gas. After capturing an electron, the neutral particles are no longer trapped: They exit the trap through the central aperture in one of the two mirrors, and those traveling downstream hit the detector. By measuring the number of particles leaving the trap as a function of time, one can deduce directly the ion lifetime as well as the total number of particles stored in the trap. Figure 2 shows such measurements for the storage of 4-keV Ar^+, CO^+, N^+, and H^+ at different residual pressures. The total number of stored ions per injection was between 500 and 5000, depending on the injection current: The number of stored ions N_t is a function of the injection particle current I and the oscillation period T, and is given by $N_t = IT$. For example, a 4-keV Ar^+ beam has an oscillation period of 6.1 μs. It is unclear at this point what the limit is on the number of ions that can be stored due to space charge effects that affect the stability condition. The lifetimes were found to be proportional to the trap pressure, and a simple extrapolation leads to a lifetime of about a few tens of seconds at a pressure of 10^{-11} Torr. The lifetimes measured here can be used to deduce the cross section σ for electron capture. Using the relation $\tau = 1/(\sigma n v)$, where n is the density of the residual gas and v is the mean velocity of the ions, a cross section σ of the order of 10^{-15} cm^2 was obtained for all the ions displayed in Fig. 2, which is in reasonable agreement with the known values for electron capture cross sections in this energy range [13]. The doubly charged Ar^{2+} ion was also stored at 8 keV, with a lifetime of 50 ms at a pressure of 2×10^{-8} Torr. It is important to point out that the main parameter affecting the storage lifetime is the velocity of the ions because the cross sections for electron capture are more or less in the same range of values for most of the singly charged ions. Thus, the lifetimes of the heavy ions are expected to scale as $\sqrt{m}$, where m is the mass of the ion.

Based on the inequality shown in Eq. (2), the trap operation should be very stable for a large range of the focusing parameters. This was tested by measuring the trapping efficiency, i.e., the number of stored ions at constant injection current, for different focusing conditions. The trapping efficiency for the 4-keV Ar^+ beam as a function of the Einzel lens potential V_z is shown in Fig. 3. A large region with a width of about 300 V where the trapping efficiency is more or less constant was found, demonstrating the exceptional stability of the trap against variation of the electric field. Also shown in this figure is the result of a simulation based on the computation of classical trajectories in the calculated electric-field configuration [10]. The region of stability shown in Fig. 3 is in good agreement with the simple condition given by Eq. (2), where the high value of V_z corresponds to a focal length of 90 mm, which is close to $L/4$, and the low value corresponds to a very long focal length. A measurement of the ion kinetic energy range for which stable trapping conditions exist was also carried out. The results show that stable storage conditions with good efficiency exist in the energy range $4.0 < E_k < 4.4$ keV for $V_z = 3.5$ kV. This can be advantageous in certain applications where the ions to be stored are produced by a process that does not lead to a well-defined beam energy, usually degrading the trapping efficiency significantly in other ion traps.

Experiments to be carried out using the electrostatic bottle described above will take advantage of its special characteristics enumerated above. The fact that the beam has a well-defined direction and energy in the central region of the ion trap, a region that is also field free, makes it possible to install particle detectors *inside* the trap without perturbing the motion of the ions. As pointed out above, the original goal for this ion trap was related to the study of the interaction between vibrationally and rotationally cooled molecular ions with electrons, photons, atomic or molecular (ion) beams. In such experiments, the molecular ions are dissociated by the interaction with the impinging particle and the fragments are measured by a detector located inside the trap. Therefore, it is possible to control the initial quantum state of the molecular ions, and to measure the kinetic energy release using fragment imaging techniques [5], thus following the complete reaction path of the dissociation process. Studies of this type have been carried out at the large storage ring facilities, mostly in the field of electron-molecular ion recom-

bination. In these cases, one takes advantage of the large energy of the molecular ions (MeV) and the presence of an intense electron beam that can be merged with the ion beam to measure cross sections of the recombination processes with high resolution [14].

Measurements of lifetimes of metastable states, as well as vibrational and rotational relaxation times of molecular ions, can also be carried out. The final temperature of these systems is related to the blackbody radiation [15], which is itself due to the trap temperature. The present trap can be cooled relatively easily to cryogenic temperatures, an important advantage for studies of weakly bound systems such as certain negative ions [16]. This was found to be a limiting factor in some cases using the heavy-ion storage ring technique, where cooling of the whole ring is practically impossible.

Another possible use is related to the measurement of the lifetime of doubly charged molecular ions AB^{2+}. Some of these ions are known to decay by tunneling through the potential-energy barrier, thus making them excellent probes of the potential energy curves [17]. This barrier is due to avoided crossings between, for example, two adiabatic potential energy curves corresponding to $A^{2+}+B$ and $A^{+}+B^{+}$ [17]. The tunneling lifetimes span a very large range, from instantaneous to minutes, and even longer. Using photodissociation and measuring the production rate of the two singly charged fragments ($AB^{2+}+h\nu \rightarrow A^{+}+B^{+}$) as a function of time, it is possible to measure the lifetime of many rovibrational states over a large period of time (limited by the lifetime of the stored beam).

Clearly, the special configuration of this trap will also open new, exciting possibilities in other fields and it is expected that new types of experiments exploiting the characteristics of the trap will be suggested and carried out in the near future.

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