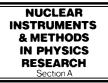


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Buffer gas cooling of ion beams

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Abstract

The cooling action of a buffer gas on ions contained within it can be used to cool an ion beam, thereby greatly improving its emittance and energy spread. It can also be used to greatly enhance the collection of an ion beam in an electromagnetic trap. The basic principles will be introduced in the context of a prototype system for such a beam cooler. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Ion guide; Ion trap; Electromagnetic traps; Buffer gas cooling; Radionuclides; Mass spectrometry

1. Introduction

The buffer gas cooling of ion beams is a subject of considerable recent interest. This is because of the realization by Kim and coworkers [1] that a low-pressure gas, usually referred to as a "buffer gas," can be used for cooling ion beams to close to the temperature of the gas itself.

The manipulation of ions in low-pressure gases is a very old subject, dating back to the 19th century. However, manipulations that made use of the cooling properties of the gas, i.e. the tendency of the ion temperature to approach that of the colliding gas molecules, were hampered by the diffusion associated with such cooling. Without containment of the ions they simply become lost within the gas. Useful damping (i.e. "cooling") of ion motion by buffer gas was therefore only possible once ion confinement by electromagnetic fields became feasible in a gas. Such containment was first proven to be feasible by Paul and coworkers [2].

Buffer gas damping of ion motion in a trap was first demonstrated by Hugget and Menasian in 1965 for Hg⁺ ions in a Paul trap into which a small amount of helium was introduced. An introduction to the basic principles was presented by Dehmelt in 1967 [3]. A more thorough summary of the early work is given in the review book by Dawson [4]. Damping time constants achieved in such devices range typically from about 1–100 ms.

A similar phenomenon has recently been demonstrated by Savard et al. [5] for the cooling of ion motion in a Penning trap. In this particular case, as observed by Penning, simple cooling of the magnetron motion would result in an expansion of the magnetron orbit, due to the fact that the overall energy of magnetron motion decreases with radius. However, in that work, the magnetron orbit was collapsed by using an azimuthal RF

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quadrupole field to transform its motion into cyclotron motion which does reduce in radius with reduced energy. This technique has now become very important in the preparation of ion collections for high-precision mass spectroscopy in Penning traps. Damping time constants achieved by this technique are typically about 100 ms.

The application of RF quadrupole confinement to ion beams in a background of buffer gas was first demonstrated by Teloy and Gerlich at Freiberg in 1974 [6].

Subsequently Okuno [7] applied the technique to studies in physics. This was followed by the study of Douglas and French [8] of the effect of background gas on the transverse motion of ions in a radiofrequency quadrupole rod structure such as used in a quadrupole mass filter for chemical analysis. Indeed it was shown that background gas at moderate pressures, up to about 1 Pa, did result in a significant increase in the transmission of ions through a 1-mm diameter orifice following the quadrupole rods.

These favors were returned to physics in the work of Xu and coworkers [9] on the transport of radioactive nuclei into cells for nuclear studies.

These results led to a study of the beam cooling process itself by Kim, cited above [1], which showed that a transverse radiofrequency quadrupole field could be used to confine ions injected at up to 100 eV and subsequently cool them to close to room temperature by a buffer gas at pressures up to 100 Pa. That work led to the work reported here.

The benefits of beam cooling derive from the concentration in phase space (the six-dimensional space made up of the momentum and position coordinates) that cooling of a collection of particles implies. Essentially, the smaller the volume occupied by the particles and the smaller the range of their momenta, the easier it is to detect them and to carry out precise measurements on them. In high-energy experiments, the cooling is achieved by immersing the beam in a bath of cold electrons running along with the beam at the average velocity of its particles. However, because of the relatively low velocities of the ion beams used in mass spectrometry, this technique is not applicable. There, the possibility of immersing the ion beam in a bath of cold gas molecules, usually referred to as a "buffer gas," is of great interest and workers at many institutions are now considering this possibility. This paper introduces the principles of buffer gas cooling in the context of a prototype system for such a cooler.

Unlike electrons, gas molecules cannot easily be made to run along with an ion beam and so they will thermalize any ions among them to their own relatively slow flow velocities. For a cooled beam, the cooled ions must therefore be dragged through the gas and reaccelerated. Furthermore, for the cooled ions to form a beam of the greatest phasespace density, they must be radially constrained into the smallest possible transverse area while they are being dragged through the gas.

Another necessity is for the original ion beam to be decelerated before entry into the cooling gas. This is because of the difficulty of bringing the ions to rest in the gas itself while still being able to drag them out of the gas to re-form them into a beam. Essentially, the large amount of gas required would flood the vacuum systems of the incoming and extracted beams.

Finally, while there is a benefit in the reduction of the energy spread of the extracted beam that results from cooling, there could be a further benefit from collecting the ions into bunches before extraction. In fact, for ion trap experiments, which are based on the manipulation and observation of well-defined ion bunches, this feature is essential. Examples for such experiments are ISOLTRAP at CERN [10], CPT at Argonne National Laboratory [11], JYFLTRAP at the University of Jyväskylä [12], and SHIPTRAP at GSI [13]. Ion bunches can be easily formed with buffer gas cooling since the cooled ions are nearly at rest and therefore amenable to collection in an electromagnetic trap. Also, in general, a given collection of ions can be observed more clearly in the presence of background noise when these ions are grouped in bunches. (This is a corollary of the general principle introduced above of the importance of reducing phase space volume for precise observations of particles.) Thus the option of including a trap should be considered for any ion beam cooler system.

A buffer gas cooling system for an ion beam will therefore have the overall configuration shown

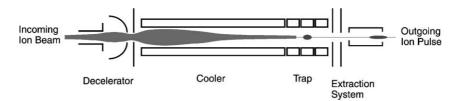


Fig. 1. Schematic of the basic components of a buffer gas system for cooling and bunching a continuous ion beam.

schematically in Fig. 1. The decelerator, the cooler, and the extraction system are placed on a highvoltage platform whose positive potential ("deceleration HV") is equal or close to the beam energy divided by the charge that each ion carries. The precise potentials on the various electrodes are obtained by applying small negative potential offsets relative to the high-voltage platform.

The central item of this system is the ion cooler. Its acceptance, i.e. the momentum spread and spatial extent of the incoming beam that it can accommodate, sets the requirements on the decelerator, and the nature of the ion collection it produces sets the requirements on the trap and/or the extraction system. The operating principles of the cooler will therefore be presented in more detail.

2. The cooling ion guide

The radial constraint of a beam of ions that are being cooled by a buffer gas is most easily achieved by an axiperiodic oscillating electric quadrupole field. The confining properties of such a field were first presented in 1958 by Paul [2] and are now widely used in what are commonly called "quadrupole mass filters." In their application to ion beam confinement, the devices are run in a non-mass-selective mode and are referred to as "ion guides." The basic principles of such containment are available in many publications (see, for example Ref. [4]) and will not be duplicated here. Only the pertinent features of the resulting motion within such a containment device will be presented.

Representative motions of an ion in an axiperiodic oscillating quadrupole electric field with buffer gas are shown in the computer simulations in Fig. 2, where the action of the buffer gas is treated as a simple viscous drag. At low pressures, the motion is seen to be basically oscillatory, as if the particles were contained within a simple harmonic potential well, with a small ripple at the frequency of the driving electric field. The effect of increased gas pressure is an increased damping of the oscillatory motion.

At a pressure of about 25 Pa, the oscillatory motion is seen to be critically damped, leaving only the ripple caused by the driving field. At this point, the decay time of the excursion of an ion from the axis is minimal. In the case shown, where the gas is nitrogen, it is about $10 \,\mu$ s. For helium, the pressure at which this occurs will be about 10 times higher. As the gas pressure is further increased, the decay time itself increases due to the increased viscosity.

Thus it is seen that buffer gas cooling of an ion beam can be very quick, of the order of several tens of microseconds. However, this high cooling rate comes at the expense of relatively high buffer gas pressures that could flood the vacuum systems of the incoming and exiting beams. The design of a practical system for using buffer gas cooling for an ion beam therefore requires a detailed study of the behavior of ions in an ion guide at various buffer gas pressures.

In order to conduct such a study, a radiofrequency quadrupole (RFQ) ion guide system was built for $^{133}Cs^+$ ions in nitrogen. This combination was selected because of the relatively high buffer gas pressures that can be accommodated with nitrogen and because of the well-known interaction potentials for collisions between a cesium ion and a nitrogen molecule [14]. Thus the behavior of ions in the guide can be simulated by Monte-Carlo calculations, facilitating the interpretation of experimental results.

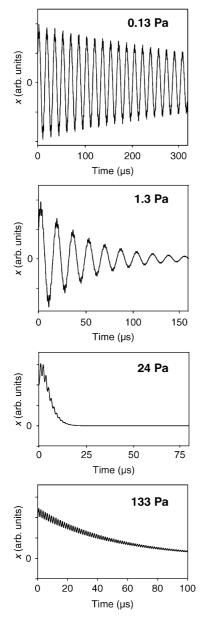


Fig. 2. Representative motions of an ion in an axiperiodic oscillating electric quadrupole field with nitrogen as a buffer gas. The frequency of the field is 700 kHz. The ion is $^{133}Cs^+$ and the strength of the field is such that the macro-oscillation frequency, as can be seen in the figure, is about 50 kHz.

The ion guide for this system was made of four rods of diameter 16 mm and length 180 mm, arranged as a quadrupole separated by 14 mm between opposite rods. The rods were housed in a container, itself inside a high-capacity (20001/s) pumping system. Orifices of 1 mm diameter were provided at each end of the container to allow the entrance and extraction of ion beams. With this system, buffer gas pressures beyond 100 Pa could be maintained while still allowing the injection of a 100-eV beam of $^{133}Cs^+$ ions into the gas and their extraction after cooling. By segmenting the rods into 5-mm disks and connecting the segments to a resistor chain, a relatively uniform DC electric field could be maintained along the axis of the structure. This allowed control of the axial as well as the radial motion of the ions.

A representative set of transmission curves obtained with this apparatus is shown in Fig. 3. It is seen that at a low buffer gas pressure, the ion transmission is only about 10%. It is also seen that this transmission peaks at a particular value of the RF potential. This is because of the stronger confinement that is achieved with the increased RF potential up to a certain point and because of the onset of instability that results from increasing the potential beyond that point.

At 30 Pa the transmission is seen to have risen to about 40%. It is also observed that the stability region has broadened a little due to the increased viscosity of the gas. Furthermore, at this higher pressure, it is necessary to have an axial field to drag the ions through the gas. At 50 Pa the transmission is seen to have reached a peak of about 60%. The stability region has now considerably broadened and the need for an axial field is even more pronounced. This trend continues in the measurements at 100 Pa.

The peak transmission of 60% indicates that the diameter of the cooled beam is of the order of 1 mm. A system was therefore built to fully extract a beam of this size and to study its profile. This was a miniature electric quadrupole made of vanes that approximated the shape of the equipotential surfaces within the ion guide and which were activated using the RF supply to the guide divided down to the level of these equipotentials. The length of the vanes was 40 mm and the separation of opposite vanes was 3.5 mm, so the RF potential amplitude of the vanes was set to 1/16th that of the main rods. Such a transition region allows the ions to escape from the buffer gas region into the

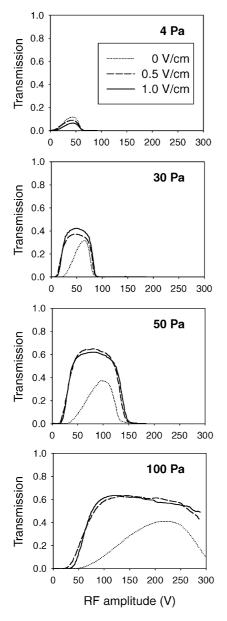


Fig. 3. A representative set of transmission curves for $^{133}Cs^+$ ions in the quadrupole ion guide at various buffer gas pressures. The figures in the enclosed legend are for the various DC axial electric fields that were applied. The radio frequency was 250 kHz and the amplitude 300 V peak-to-peak. The ion energy at injection was 100 eV.

extraction region without experiencing any change in the electric field that confines them. After exiting the vane structure, the ions were accelerated to a collection plate.

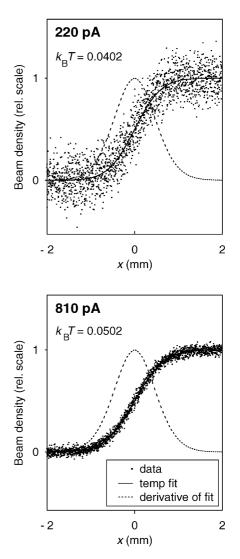


Fig. 4. Typical beam profiles obtained by the rotating beam profiler.

The profile of the extracted beam was obtained by making the collection plate from a pair of copper semicircles mounted on an insulating disk and rotated so that the gap between the plates crossed the extracted beam. Monitoring the current on the plates gave an integral of the beam profile. Fig. 4 shows that the profiles obtained by this method were approximately Gaussian, as would be expected for the distribution of an ion collection that is in thermal equilibrium while it is being restrained within a simple harmonic potential well. The solid lines shown in Fig. 4 were obtained by fitting such distributions to the data, with the temperature as the only free parameter.

It is seen that the extracted beam is indeed about 2 mm wide, in keeping with the transmission of about 60% through a 1-mm hole. (The actual diameter inside the ion guide is not exactly the same as at the collector due to the focusing effects of the extraction field. For details of how this is taken into account in the fits shown in Fig. 4, see Ref. [1].) It is seen from the fits that the ions in the beam are cooled to approximately twice the temperature of the buffer gas molecules. This is in keeping with the results of Lunney [15] for the temperature of ions cooled by buffer gas in a Paul trap. It is also seen that there is evidence that at higher beam currents, the ions tend to be a little hotter, also in keeping with the results of Lunney.

While the experimental results showed that 100-eV ions injected into an ion guide were indeed cooled to about twice the temperature of the buffer gas itself, there seems to be no feasible method for investigating the dynamics by which this process takes place. This is because of the impossibility of inserting probes into the guide without destroying the electric fields within the guide. Monte-Carlo simulations were therefore carried out to investigate these dynamics. (Details of how these calculations were performed are given in Ref. [1].)

The results of the Monte-Carlo simulation for a typical operating condition of the ion guide are shown in Fig. 5. It is seen that a 60-eV ion beam injected at 1 mm diameter, and with no angular spread, will expand to about 8 mm diameter after penetrating 20 mm into the gas. It then quickly cools until at z = 180 mm it is only about 2 mm across.

It can be seen that the profile, particularly of the cooled beam, is not exactly circular but diamond-shaped. This is particularly apparent in the emittance plots where the shape becomes even that of a star. This is characteristic of RF quadrupole confinement, as explained in Ref. [4].

The accuracy of the Monte-Carlo simulation was checked by comparing its results for the time involved in the cooling process with experimental observations. These are shown in Fig. 6.

It is seen that the experimental observations agree very well with the Monte-Carlo simulations. The slight deviation can be attributed to the smallangle collision cutoff that has to be imposed to make the Monte-Carlo simulation feasible.

Thus it has been proven that a small amount of buffer gas can very effectively cool a 100-eV beam of ions to about twice the temperature of the gas

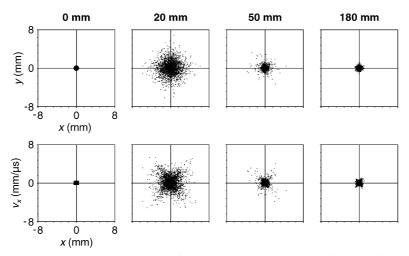


Fig. 5. The beam profile (upper) and emittance plots (lower) from Monte-Carlo simulations of a beam of 60-eV 133 Cs⁺ ions injected into the beam cooler with nitrogen at 8 Pa as the buffer gas. The numbers above the diagrams indicate the distance along the axis for which the various plots are made. The initial diameter of the beam, at z=0 mm, is 1 mm. The beam diameter is maximal at z=20 mm.

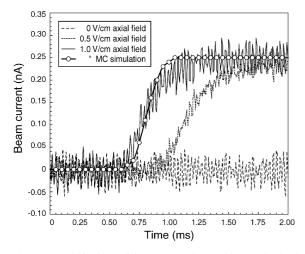


Fig. 6. The drift time of ions in the ion guide as obtained experimentally and from a Monte-Carlo simulation for 1.0 V/mm axial field.

and that RF quadrupole confinement during this cooling can produce a beam of very small emittance. It has also been proven that such cooled ions can be extracted from the cooler to form a very well-defined beam. What remains is to develop a decelerator to reduce the kinetic energy of a typical mass-separator beam to about 100 eV for injection into such a cooler.

3. The deceleration system

The decelerator for a buffer gas ion beam cooler using an ion guide must be designed around the acceptance of the ion guide (i.e. the detailed shape of the momentum-displacement diagrams of the ions that can be contained within the ion guide). A procedure by which this can be done is presented in Ref. [16]. The resulting design of a decelerator and beam cooler is shown in Fig. 7. The quadrupole rods have a diameter of 18 mm and the centers of opposing rods are 30 mm apart. The major differences with the test ion guide described above are that the longitudinal segments are only 20 mm long and that the rod assembly is designed to form a closed cylindrical system. This forces the buffer gas, which is injected at the far end of the system, to escape through the front entrance

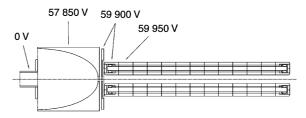


Fig. 7. A cross-section of the decelerator-cooler system that was built as a test bed for the system being developed for the ISOLTRAP experiment. The potentials shown are for the deceleration and capture of a 60-keV beam.

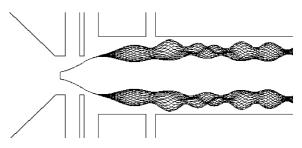


Fig. 8. The calculated beam profiles for a typical ISOLDE ion beam injected into the RFQ ion guide for cooling. The different profiles are for the same ion beam entering at different RF phases. The start of the profile maps is at a beam energy of 1900 eV after deceleration from 60 keV. The initial emittance of the 60-keV beam is taken to be an ellipse of 30π mm mrad. The separation of the rods forming the ion guide is 12 mm. The profiles are shown for a depth of 60 mm into the guide.

opening. This creates a longitudinal pressure gradient and thus reduces the pumping requirements for maintaining an adequate vacuum in the high-electric-field deceleration region and a sufficiently high buffer gas pressure in the ion guide.

The system was designed to operate with nitrogen at 100 mPa as the buffer gas, the pressure in the deceleration region then being about 10 mPa. Fig. 8 shows the calculated beam profile within this system for a number of RF phases immediately after injection into the cooler and before the buffer gas has had a significant effect on the ion motion.

Because of the approximations that must be introduced in the computer simulations in order to make the design work feasible, it was regarded as necessary to build a prototype to test the design. The prototype system was tested with a non-radioactive cesium ion source that provides a 100-pA 133 Cs⁺ ion beam at 40 kV.

Fig. 9 shows the ion beam transmission through the entire deceleration and cooling system as a function of the potential of the ion guide highvoltage platform (cage potential) for a nitrogen buffer gas pressure of about 80 mPa. When the deceleration HV is at ground potential, the highly energetic ion beam travels through the system practically without being decelerated because the relatively small DC and RF potentials it encounters are not sufficient to lead to a deviation of the beam's path. As the deceleration HV potential is raised, the ions preserve less energy as they enter the deceleration region and the ion guide. The DC potentials on the deceleration electrodes now manage to decelerate the beam substantially. In the process, some of the longitudinal momentum of the ions is transferred to transverse momentum. When the deceleration HV potential is now further raised to a potential of only a few volts below the potential that corresponds to the ion energy, the transverse phace space of the ions matches the acceptance of the ion guide. A substantial fraction of the ions is captured, cooled, and transported to the far end of the system. Fig. 9 thus shows that ion beams of 40 kV can be captured and cooled in the prototype system. The transmission for a deceleration potential of 0 V was estimated to be about 40-50% from geometrical considerations. This leads to an estimate of the capture and cooling efficiency of about 30% of the original ion beam.

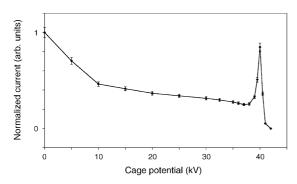


Fig. 9. The transmission of an ion beam through the decelerator-cooler system as a function of the cage potential. The energy of the incoming ion beam is 40 keV.

4. General considerations and outlook

The results reported above have made us quite optimistic about the development of efficient collector-cooler systems for the ISOLTRAP experiment in particular and for ion beams in general. From our experience, the successful development of such systems requires several technical aspects to be considered.

The first, and most important, are those concerning the implementation of high voltage. Here there are three distinct regimes to consider: atmospheric pressure, high vacuum and typical buffer gas pressures. In all three regimes, the general requirement of clean, rounded, and polished surfaces applies for all electrodes, but there are specific considerations for each region. For the atmospheric part of the system, the size of the cages to be kept at high voltage requires safe shielding and safeguards against surge currents from the inevitable occasional spark-overs. Inside the vacuum system, the arc problems are ameliorated somewhat, particularly for electrodes that are at a positive potential relative to their surroundings so that the emission of electrons is prohibited. Also, for such electrodes, a slight gas pressure (about 10 mPa seems to be optimal) can actually increase the breakdown potential, possibly due to the cleaning of the electrode surface by sputtering [17]. However, it appears that potential gradients should always be kept to less than about 5000 V/mm. For the buffer gas region of the system, one is often in the regime where the breakdown potential in the gas is minimal [18]. This is of concern when trying to achieve the highest possible electric quadrupole potential on the rods of the ion guide. Here it is important to avoid long paths for electrons to follow in moving between electrodes since it is such long paths that allow the electron to encounter a gas molecule and ionize it.

Another important consideration is the cleanliness of the vacuum system. This is because of the many collisions with molecules that an ion will encounter during its cooling and hence the many opportunities for exchange of its charge and escape from the system. This is not so critical for a relatively benign ion such as ¹³³Cs⁺, but for more aggressive ions, particularly those of noble

gases, one encounter with a contaminant molecule can lead to instant loss. The vacuum system should therefore be fabricated using polished stainless steel or oxygen-free copper and non-organic insulators as much as possible and should be designed to be bakeable. Only ultra-pure sources of gas should be used as buffers.

Another design consideration that applies to any RF containment system is that all insulating surfaces should be kept very clear of the RF electric field. This is because of the possibility that stray electrons may be deposited on an insulator surface by the RF field and remain there as a DC patch of charge which can severely disturb the confinement properties of the RF field itself.

With proper attention to these considerations and with the experience gained using the prototype ion guide now in operation at McGill University as well as RFQ ion beam coolers in operation at radioactive-beam facilities, it should be possible to reach overall collection and cooling efficiencies of close to 100% for ISOLDE-type beams. The energy spread of such beams after cooling should approach that in the work described here: twice room temperature or about 50 meV (FWHM). The emittance that can be obtained with such cool beams will depend on the strength of confinement [19]. From the results presented here, it is expected that normalized emittances of the order of 0.5 eV us should be achievable (corresponding to about 1π mm mrad for a 60-keV beam).

As for the ion current that can be cooled, the results presented here show that currents of up to 1 nA display only a slight rise in beam temperature and hence a corresponding slight increase in emittance. This is no doubt due to the radial field of the space charge of the higher-current beam which pushes more ions out into a higher quadrupole field. This results in higher-energy collisions with the buffer gas molecules and hence in a higher temperature of the ion [15], an effect which is commonly called "RF heating." This effect will certainly get worse for larger currents but can be reduced somewhat by applying a greater axial field in the ion guide so that the beam is stretched. However, a high axial field can itself induce heating from an increase in collision energy due to the axial motion. The practical limit for the beam current that can be cooled will therefore depend on the emittance and energy spread that one is willing to tolerate. From general considerations, it is expected that it should be possible to cool currents of up to 1 μ A to an energy spread of less than 1 eV and normalized emittances of less than 2 eV μ s.

The problems of high beam current in an ion guide could be ameliorated somewhat by deliberately introducing negative ions that would neutralize the space charge of the positive beam. This is possible in RF quadrupole confinement because both positive and negative ions are confined simultaneously. Furthermore, the micromotions of the two are out of phase so that the possibility is considerably reduced of their being at rest relative to each other and therefore subject to charge exchange. However, with the axial fields necessary to pull the cooled ions through the buffer gas, it may not be possible to sustain enough negative ions to have an appreciable space charge neutralization.

In any case, the primary use of beam cooling is expected to be with weak beams that must be collected and cooled to enable their detection. As pointed out above, it is very important to collect such beams into bunches in a trap. Here the important considerations are not only the nature of the collected bunch but also the time it takes to prepare such a bunch. From the work reported here, it is seen that such a bunch will take about 1 ms to be formed. As with the cooled beam, the transverse emittance will depend on the temperature of the ions and the degree of radial confinement within the trap. Likewise, the longitudinal emittance of the bunch will depend on the ion temperature and the strength of the axial confinement.

However, such a trap will not be strong enough to contain many ions without excessive heating due to their space charge repulsion. It is expected that the limit will be of the order of several thousand and very modest beam currents will therefore overload the trap very quickly. This can be a problem if it is largely made up of contaminant isobaric ions from which the desired ions must be separated. For such cases it may well be appropriate to have the cooled ion beam

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collected in a large Penning trap immediately at the end of the cooler. This is possible because the magnetic field of the Penning trap would have a negligible effect on the RFQ ion guide itself. Although this would be considerably more expensive than a small linear trap incorporated in the RFQ ion beam cooler, it would allow the continuous stream of ions to be accumulated directly.

We hope that this contribution shows that 40 years after the development of the first quadrupole ion traps, buffer gas cooling finally offers the possibility of collecting any beam of rare ions and holding them in an electromagnetic trap for careful study. The authors are convinced that these advances will significantly enhance precision studies with radioactive nuclear beams all over the world.

Acknowledgements

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