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Beam purification by photodetachment (invited)[a)]

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Ion beam purity is of crucial importance to many basic and applied studies in nuclear science. Selective photodetachment has been proposed to suppress unwanted species in negative ion beams while preserving the intensity of the species of interest. A highly efficient technique based on photodetachment in a gas-filled radio frequency quadrupole ion cooler has been demonstrated. In off-line experiments with stable ions, up to 10^4 times suppression of the isobar contaminants in a number of interesting radioactive negative ion beams has been demonstrated. For selected species, this technique promises new experimental possibilities in studies on exotic nuclei, accelerator mass spectrometry, and fundamental properties of negative atomic and molecular ions. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.3671747]

I. INTRODUCTION

Negative ions are in demand for a wide variety of fundamental and applied research topics in nuclear science. For studies of short-lived exotic nuclei in nuclear physics and nuclear astrophysics, negative ion beams are required for radioactive ion beam facilities such as the Holifield Radioactive Ion Beam Facility (HRIBF) at Oak Ridge National Laboratory and the Exotics with Cyclotron and Tandem facility at INFN-LNS. In the field of accelerated mass spectrometry (AMS), accelerated negative ion beams are used for the detection of long-lived rare isotopes for a variety of studies such as archeological dating and climate change. Negative ions also play significant roles in many physical and chemical processes involving weakly ionized gases and plasmas. For many studies, ion beams free of contaminants must be provided in order to unambiguously interpret the experimental data. Isobar suppression is one of the main challenges for these studies. Selective removal of isobaric contaminants from negative ion beams by photo-detachment has been suggested for AMS applications[1,2] as well as for purification of radioactive ion beams (RIBs).[3] The efficiency of photodetachment of a relatively fast ion beam is limited by the photon fluence interacting with the anions. A technique based on selective photodetachment in a radio frequency quadrupole (RFQ) ion cooler has shown to be able to substantially increase the efficiency of the photodetachment process and thus the degree of isobar suppression.[4] In off-line experiments with stable ions, a factor of up to 10^4 times suppression of the isobar contaminants in a number of interesting radioactive negative ion beams has been demonstrated. Such highly efficient removal of unwanted contaminant species opens new experimental possibilities in nuclear research with negatively charged RIBs and in AMS applications as well as in fundamental studies of negative atomic and molecular ions. In this paper, different photodetachment techniques for beam purification will be described.

II. SELECTIVE PHOTODETACHMENT

Berkovitz *et al.*[1,2] first proposed to suppress the isobars of neighboring elements with lower electron affinities (EA) using fixed frequency laser light. The principle is simple: if the electron affinity of the isobaric contaminant, EA_1, is lower than the electron affinity of the radioactive ions of interest, EA_2, the contaminant anions can be selectively neutralized by photodetachment with photons of energy $EA_1 < h\nu < EA_2$. They showed a strong depletion of the $^{32}S^-$ (EA = 2.077 eV) ions, while the more strongly bound $^{36}Cl^-$ (EA = 3.62 eV) ions were not affected, when illuminated with a Nd:YAG laser at 532 nm ($h\nu = 2.33$ eV).[1] In a subsequent ^{59}Ni (EA = 1.156 eV) experiment,[2] a suppression factor of 125 for the ^{59}Co (EA = 0.661 eV) isobaric background was obtained with a 1064 nm laser ($h\nu = 1.165$ eV). However, in their demonstration experiments, the laser interacted directly with fast moving negative ions accelerated to 100 keV. Consequently, the laser-ion interaction time was limited to a few microseconds and powerful pulse lasers were required for sufficient photodetachment. Such pulsed lasers typically have low repetition rates (10–30 Hz) and short pulse widths (nanoseconds). Thus, the overall degree of isobar suppression was very small due to a low duty cycle.

Sandström *et al.* have recently demonstrated a technique, laser photodetachment mass spectrometry (LPMS),[5,6] that can selectively suppress a specific isotope of an element, while leaving the other isotopes of the same element unaffected. The LPMS technique utilizes the mass-dependent Doppler shift of the laser frequency experienced by fast moving ions in collinear geometry with the laser beam to achieve isotope selective suppression and detection. For ions acceler-

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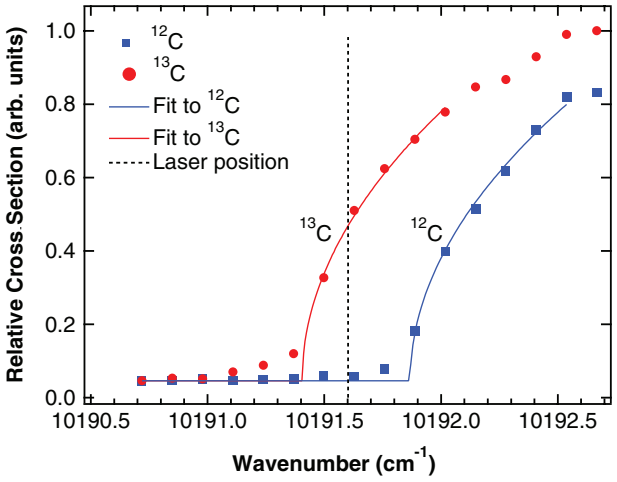

FIG. 1. (Color online) Doppler shifted photodetachment threshold for ^{13}C and ^{12}C carbon isotopes. Circles and squares are measured data and the lines are Wigner law fits to the data. [Reprinted with permission from P. Andersson *et al.*, Nucl. Instrum. Methods B **266**, 3667 (2008). Copyright 2008, Elsevier.]

ated to the same kinetic energy, different isotopes with different masses will have different final velocities and thus experience different Doppler effects. As a result, the effective photodetachment thresholds for neighboring isotopes become slightly separated. A narrow bandwidth tunable laser is then used to select a specific isotope to be neutralized. Using co-propagation of the laser and the ion beams, one can selectively detach the heavier isotopes of an element, and *vice versa*. However, this method is limited to negative ions in which the detached electron is a p-orbital electron. This is because the cross section for photodetachment of a p-electron increases sharply as an inverse square root function with photon energy in the region just above the threshold. This sharp-onset makes it possible to detach the selected isotopic ions with sufficient efficiency and thus sufficient discrimination against the other isotopes. Figure 1 shows an example of Doppler-shifted photodetachment thresholds for ^{13}C and ^{12}C. It is noted that the condition of detaching a p-electron can be satisfied by elements in the right half of the periodic table (group 13–17) which all have p-orbital valence electrons.

The LPMS technique has been demonstrated to improve the detection sensitivity of a specific isotope of sulfur and carbon by as much as a factor of 50 at ion beam energies as low as 5 keV.[5,6] It could be a great tool to enhance the sensitivity of analytical mass spectroscopy. However, for many applications, such as beam purification for nuclear research with RIBs and AMS, it is the overall degree of suppression of unwanted ions that matters. The applicability of LPMS is then limited due to the very low overall efficiencies caused by the short interaction time (typically a few microseconds) between the laser and the fast-moving ions.

The technique of photodetachment in a RFQ ion cooler, developed at HRIBF,[4] can significantly increase the laser-ion interaction times and can provide good spatial overlap between the laser beam and the negative ions as well. Thus, very high overall photodetachment efficiency can be obtained. This technique is very promising for applications such as purifying isobaric contaminants in RIBs for nuclear research at HRIBF or other ISOL facilities, or removing unwanted stable isobars in radioisotope ion beams for AMS.

III. PHOTODETACHMENT IN A RFQ ION COOLER

A RFQ ion cooler is a quadrupole mass filter operating in rf-only mode and filled with a buffer gas.[7] In such a cooler, ions suffer multiple collisions with the buffer gas molecules. If the buffer gas is sufficiently lighter than the ions, the ions will lose kinetic energy in the collisions until they are thermalized with the buffer gas. Such buffer gas cooling has been used to produce atomic and molecular beams with reduced kinetic energy spread and bunching for high-precision measurements.[8,9]

A RFQ ion cooler has been developed at HRIBF for improving the emittance of negatively charged RIBs such as ^{17,18}F$^-$.[7] The quadrupole consists of four parallel cylindrical rods of 8-mm diameter and 40-cm length, equally spaced with an inscribed circle of 3.5 mm radius. It operates at frequencies around 2.75 MHz with RF voltages up to 500 V. The cooling process involves decelerating energetic ion beams to low energies during injection into the cooler and then re-accelerating the cooled ions from the cooler. For negative ions, it is necessary to decelerate the ions to less than 40 eV in order to reduce losses due to collisional detachment. A number of negative ions of interest, produced in a Cs-sputter negative ion source, have been cooled to ~2 eV FWHM in the RFQ ion cooler and more than 50% transmission efficiency has been demonstrated for some negative ions.[10]

Figure 2 shows a simulation of ion trajectories in the HRIBF RFQ ion cooler filled with 30 mTorr He. The simulations show that during transit through the RFQ, the kinetic energy of the ions can be effectively dissipated and reduced to approximately the thermal energy of the buffer gas and the ion trajectories can be confined to a very small region near the longitudinal axis of the RFQ. Once thermalized, the ions also lose their forward momentum and their motion along the axis of the RFQ is essentially governed by diffusion. It is thus necessary to apply a small longitudinal electrostatic field to

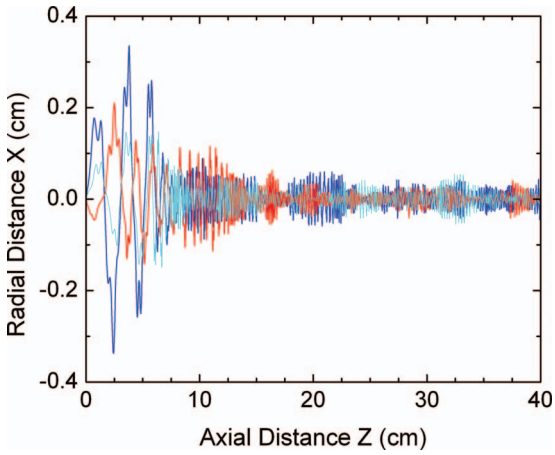

FIG. 2. (Color online) Monte Carlo simulation of trajectories of ^{19}F negative ions in the *XZ* plane of the HRIBF RFQ ion cooler with 30 mTorr He, rf frequency $f = 2.75$ MHz, rf voltage $V_{rf} = 80$ V, and the longitudinal electric field $E_z = 5$ V/m. The ions have an initial energy of 40 eV.

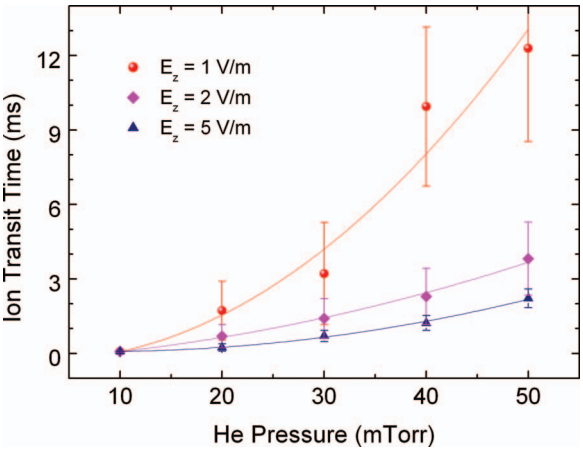

FIG. 3. (Color online) Calculated average transit time of ^{56}Co negative ions in the HRIBF RFQ ion cooler at different He buffer gas pressures and longitudinal electric fields.

push them toward the exit. Total ion transit time on the order of milliseconds can easily be achieved.[8,11,12] For the RFQ ion cooler at HRIBF, it is predicted that the ion transit time can be varied from less than 1 ms to more than 10 ms by changing the applied longitudinal field and buffer gas pressure, as illustrated in Figure 3. This combination of a small transverse ion beam dimension and extended ion transit time inside the RFQ ion cooler provides ideal conditions for laser ion interaction.

High efficiency photodetachment in a RFQ ion cooler has been demonstrated at HRIBF. The experimental setup has been reported in detail in Refs. 4 and 12. Figure 4 shows a measurement of $^{32}S^-$ negative ion current extracted from the HRIBF RFQ ion cooler. The $^{32}S^-$ ions were produced in a Cs-sputter negative ion source, accelerated from the ion source to about 5 keV energy, mass analyzed through a 90° dipole magnet, and then decelerated to less than 40 eV energy and focused into the ion cooler filled with He gas. Upon exiting the ion cooler, the $^{32}S^-$ ions were re-accelerated to energy of 5 keV and deflected by an electrostatic deflector to an off-axis Faraday cup (FC). A Nd:YAG laser beam with

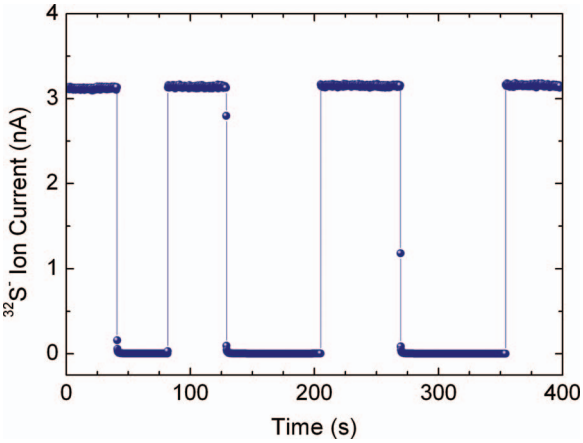

FIG. 4. (Color online) Measured ion current of $^{32}S^-$ ions accelerated from the RFQ ion cooler operated at a He pressure of ~6 Pa and $f = 2.75$ MHz. When the 532 nm laser beam was on, the $^{32}S^-$ ions were almost completely neutralized.

frequency-doubled output at 532 nm was directed into the ion cooler from the exit end. The laser beam propagated through the cooler in the opposite direction of the ion beam. The energy of the 532 nm photon is 2.33 eV, which is larger than the EA of S (2.077 eV). The laser was pulsed at 10 kHz repetition rate and had an average power of ~2.5 W. The data in Figure 4 correspond to a sequence of blocking (laser-off) and unblocking (laser-on) the 532 nm laser beam. As seen, when the laser beam was on more than 3 nA of $^{32}S^-$ ions were almost completely depleted. The photodetachment process was instantaneous and reproducible. The measurable residual $^{32}S^-$ ion current was ~2 pA, limited by the FC dark current. This corresponded to neutralization of more than 99.9% of the $^{32}S^-$ beam. Such high detachment efficiency offers promising applications in research with negative ion beams. The feasibility of using this technique for isobaric contaminant removal and elimination of excited state populations has been investigated for a number of applications. The results of recent developments are presented below.

IV. BEAM PURIFICATION BY SELECTIVE PHOTODETACHEMNT

A. Removal of isobar contaminants in negative radioactive ion beams

The HRIBF at Oak Ridge National Laboratory provides accelerated RIBs for experimental research on exotic nuclei. The purity and intensity of the RIBs are crucial to many experiments. A number of important RIBs needed for studies in nuclear physics and nuclear astrophysics at HRIBF are often dominated by isobaric contaminants. For example, ^{17,18}F, ^{33,34}Cl, and ^{56}Ni are dominated by $^{17,18}O^-$, $^{33,34}S^-$, and $^{56}Co^-$, respectively. As shown in Table I, the EA of the contaminant is smaller than that of the corresponding RIB. The efficiency of removing $^{17,18}O^-$ from $^{17,18}F^-$, $^{33,34}S^-$ from $^{33,34}Cl^-$, and $^{56}Co^-$ from $^{56}Ni^-$ beams by selective photodetachment in a RFQ ion cooler has been investigated using negative ion beams of stable isotopes. For the S^-/Cl^- and O^-/F^- pairs, a frequency doubled Nd:YAG laser at 532 nm ($h\nu = 2.33$ eV) or a frequency doubled Nd:YLF laser at 527 nm ($h\nu = 2.35$ eV) was used to remove the S^- and O^- ions, while no photodetachment of the Cl^- and F^- ions were expected. As shown above, more than 99.9% depletion of $^{32}S^-$ with 2.5 W of 532 nm laser radiation has been demonstrated. Under the same conditions for $^{32}S^-$, beams of $^{35}Cl^-$ negative ions were also injected into the ion cooler and interacted with the 532 nm laser, but no photodetachment of the $^{35}Cl^-$ ions was observed. Similar results have been obtained for suppression of O^-: photodetachment of 99.9% of $^{16}O^-$ ions in the

TABLE I. Electron affinities of RIBs and the isobars.

RIB	EA (eV)	Isobar	EA (eV)
F	3.4	O	1.46
Cl	3.62	S	2.077
Ni	1.156	Co	0.661

RFQ cooler was obtained with about 2.5 W average power of a 527 nm laser, while the desired F^- ions were not affected.[12] For the Co/Ni pair, a continuous wave (CW) Nd:YAG laser at 1064 nm ($h\nu = 1.165$ eV) was used to selectively remove the Co^- ions. Again, 99.9% of the $^{59}Co^-$ ions were neutralized with less than 4 W laser power, while about 20% of the $^{58}Ni^-$ ions were depleted under the same conditions.[12] In all these studies, the depletion of the isobar negative ions could only be measured to the order of 99.9% due to the FC dark current of ~2 pA. The actual degree of depletion was believed to be higher.

In order to measure the actual degree of depletion it was necessary to use a detection method that is more sensitive than a Faraday cup. Therefore, a new detection method based on particle counting with a Channeltron detector was employed to measure the residual ions surviving the photodetachment process with much improved sensitivity and accuracy.[13] Using this new method, more than 99.99% photodetachment of $^{59}Co^-$ was observed with less than 3 W average power from a pulsed 1064 nm Nd:YAG laser at 10 kHz repetition rate, as shown in Figure 5 (upper). This corresponds to more than 10^4 times suppression for the Co^- isobar contaminants. Under identical conditions, less than 20% of the $^{58}Ni^-$ ions were detached [Figure 5 (lower)]. The small loss of $^{58}Ni^-$ ions is expected as the energy of the 1064 nm photons is slightly above the EA of Ni^-. This loss can be avoided by using lasers of longer wavelengths. We have used 1064 nm lasers in the studies because the lasers were conveniently available. Although the study was only conducted for Co/Ni negative ions, the same order of suppression (10^4) for the O and S contaminants is expected.

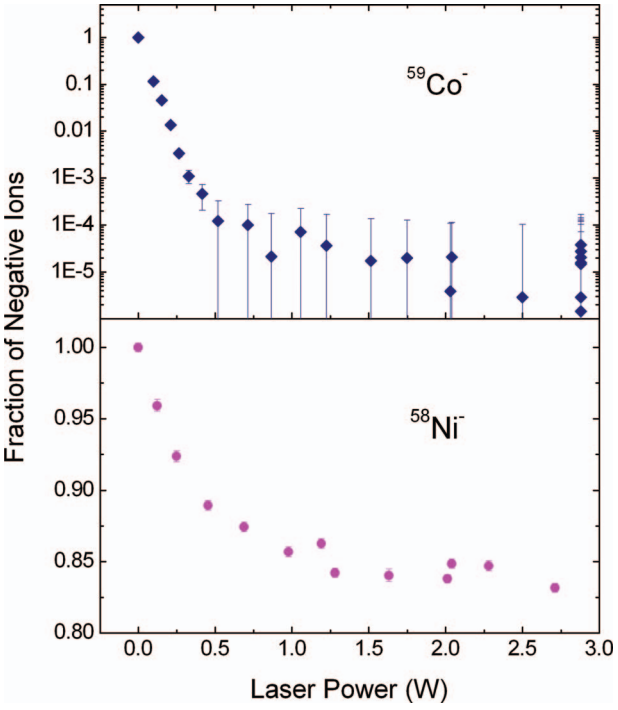

FIG. 5. (Color online) Fraction of $^{59}Co^-$ and $^{58}Ni^-$ ions not neutralized in the RFQ ion cooler by 1064 nm laser light. The cooler was operated at a He pressure of ~6 Pa, $f \sim 2.75$ MHz.

These results demonstrate that it is possible to achieve nearly complete suppression of the isobaric contaminants by photodetachment in a RFQ ion cooler for the selected RIBs. The laser power needed for this is readily available from existing commercial lasers. The RFQ ion cooler and the laser system are compact enough to implement at existing accelerator facilities. The transmission efficiency of the ion cooler ranges from about 35% for F^- to more than 50% for Cl^- and Ni^- ions,[10] which is sufficient for practical use. Therefore, this technique is very promising for real applications. It is being developed for online purification of the selected RIBs at HRIBF.

B. Isobar suppression for AMS

This highly efficient beam purification technique could be a valuable tool for AMS measurements of certain ultra-trace radioisotopes. It could provide isobar suppression at low energies before the ions are injected into a tandem accelerator. A particular potential application is to increase the detection limit of ^{36}Cl. ^{36}Cl ($T_{1/2} = 301\,000$ year) is an important cosmogenic and anthropogenic tracer for geology, hydrology, and radioecology studies, as well as for nuclear waste management. The main limit in AMS analysis of ^{36}Cl is the presence of ^{36}S isobar interference. As presented above, S^- ions can be selectively and efficiently removed with laser light in a RFQ ion cooler. In a more dedicated study,[14] suppression of S^- by a factor of 3000 was obtained with a pulsed Nd:YLF at 527 nm ($h\nu = 2.35$ eV) and only about 2.5 W average laser power. Under the same conditions, no photodetachment of Cl^- was observed. Such large suppression of ^{36}S could significantly improve the sensitivity of ^{36}Cl measurements at an AMS facility. It could also make ^{36}Cl measurements possible with smaller AMS systems.

The HRIBF hosts the highest operating voltage (25-MV) electrostatic accelerator in the world. Using this, Galindo-Uribarri *et al.*[15] have successfully measured $^{36}Cl/Cl$ ratios as low as a few times 10^{-16} in sea water samples, a sensitivity required for oceanography applications. The feasibility of combining the technique of photodetachment in a RFQ ion cooler with the 25-MV accelerator is being studied to further push the detection limits for ^{36}Cl.

Photodetachment could also help to suppress the isobar interference for a number of other ultra-trace elements in AMS measurements: for example, ^{59}Co in ^{59}Ni beams, ^{92}Zr (EA = 0.426 eV) in ^{92}Nb (EA = 0.893 eV) beams, and ^{137}Ba (EA = 0.145 eV) in ^{137}Cs (EA = 0.472 eV) beams.

C. Production of pure ground-state negative ion beams

Negative ions are usually produced in sputter ion sources or plasma ion sources with significant populations of excited states. For atomic negative ions, virtually all of these bound excited states are metastable with lifetimes that can exceed seconds or even hours.[16] The presence of uncharacterized population of excited states often limits the precision and accuracy of fundamental studies of negative ions, such as

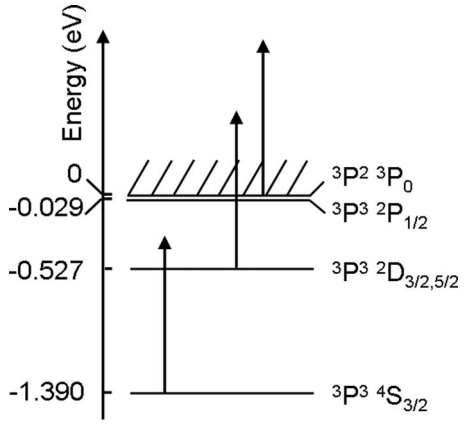

FIG. 6. Energy levels of Si negative ion. The arrows indicate the 1064 nm photon energy.

photodetachment threshold spectroscopy and studies of collisions between electrons and atomic or molecular anions. Since the excited states have smaller binding energies than the ground state, it is possible to selectively neutralize the excited anions with a laser of proper photon energy. The feasibility of depleting the excited populations in negative ion beams by photodetachment in a RFQ cooler has been recently investigated with Si negative ions.[17] Si^- has two bound excited states: a ^2P state of binding energy of 0.029 eV and a ^2D state that has a binding energy of 0.527 eV, as illustrated in Figure 6. The photon energy of a 1064 nm ($h\nu = 1.165$ eV) laser is large enough to remove the extra electron from the anions in the excited states but not sufficient to affect the anions in the ground state.

The experiment was conducted with the HRIBF RFQ ion cooler and a CW 1064 nm Nd:YAG laser. The experimental setup has been reported in Ref. 15. In the study, it was observed that the very loosely bound ^2P state may be depopulated through collisions in the ion cooler which was operated at a He pressure of ~6 Pa. About 98% of the remaining ^2D population in Si^- was removed by photodetachment inside the cooler, with only about 2 W 1064 nm laser power. The total reduction of the excited populations in Si^-, including collisional detachment and photodetachment, was estimated to be $99 \pm 1\%$.

These results show the possibility of obtaining pure ground-state atomic negative ion beams by state-selective photodetachment in a RFQ cooler. This technique can be applied to any atomic or molecular negative ions that possess bound excited states. The elimination of excited states will facilitate new or improved experiments on fundamental studies on negative ions such as enhancing the selectivity as well as accuracy of high-precision experiments on many atomic and molecular negative ions.

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