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# Depletion of the excited state population in negative ions using laser photodetachment in a gas filled RF quadrupole ion guide

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**Abstract.** The depopulation of excited states in beams of negatively charged carbon and silicon ions was demonstrated using collisional detachment and laser photodetachment in a radio frequency quadrupole ion guide filled with helium. The high lying, loosely bound <sup>2</sup>D excited state in C<sup>-</sup> was completely depleted through collisional detachment alone, which was quantitatively determined within 6%. For Si<sup>-</sup> the combined signal from the population in the <sup>2</sup>P and <sup>2</sup>D excited states was only partly depleted through collisions in the cooler. The loosely bound <sup>2</sup>P state was likely to be completely depopulated and the more tightly bound <sup>2</sup>D state was partly depopulated through collisions. 98(2)% of the remaining <sup>2</sup>D population was removed by photodetachment in the cooler using less than 2 W laser power. The total reduction of the excited population in Si<sup>-</sup>, including collisional detachment and photodetachment, was estimated to be 99(1)%. Employing this novel technique to produce a pure ground state negative ion beam offers possibilities to enhance selectivity, as well as accuracy, in high precision experiments on atomic as well as molecular negative ions.

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## 1. Introduction

Negative ions are quantum systems where electron correlation effects play an important role in both structure and dynamics. The addition of an extra electron to an otherwise neutral system changes the fundamental properties of the quantum system [1]. An atomic negative ion only supports a few bound states, and its binding energy is typically an order of magnitude smaller than the ionization potential of the neutral atom. The most striking property of an atomic negative ion is its lack of optically allowed transitions. Presently, there is only one case, namely the  $\text{Os}^-$  ion, where it has been unambiguously proven that an excited state with a parity opposite to the ground state exists [2, 3]. The lack of bound-bound transitions means that essentially all investigations of the properties of negative ions involve a bound-free transition, in which the extra electron in a negative ion is ejected due to the impact of a photon, electron, atom, molecule, or ion. Negative ions are usually produced in sputter ion sources or plasma ion sources which results in internally hot ion beams where all bound excited states are populated. The distribution of these various states strongly varies with the ion source conditions and is thus usually unknown.

In experimental investigations of the structure of negative ions, laser based methods, such as laser photodetachment threshold spectroscopy (LPTS) [4], photo electron spectroscopy [5], and photodetachment microscopy [6], are commonly used. Electron collision experiments [7, 8], on the other hand, are mainly used to investigate the dynamics of negative ions. While photo electron spectroscopy and photodetachment microscopy do not necessarily rely on a pure ground state beam, a high population of excited states may compromise LPTS studies. If the excited states with a lower binding energy than the state under investigation are populated, they produce a sub-threshold photodetachment signal that can severely limit both the precision and the accuracy of a threshold determination. This can be seen in the study of  $\text{P}^-$  by Andersson *et al.* [9], where the determination of the ground state threshold has an uncertainty that is more than twice that of the lower lying thresholds. The reduced precision comes mainly from the noise in the base line due to the presence of the sub threshold signal. The accuracy on the other hand is mainly affected by the unknown shape of the background. In collision experiments, the involvement of multiple initial states in a reaction renders the interpretation of experimental results difficult. To date no ion sources exist that can efficiently produce an internally cold ion beam, consisting of only ground state negative ions. Thus the development of a corresponding cleaning procedure to remove the unwanted excited components is highly desirable.

The problem of excited state populations in fundamental studies of negative ions is similar to the problem of isobaric interference in Accelerator Mass Spectrometry (AMS). AMS, currently the most sensitive method for ultra trace level element analysis, is utilized in numerous fields of applied research [10, 11]. In AMS, the ions are produced from a negative ion source. After pre-acceleration and mass selection the ions are accelerated towards the positive high voltage terminal of a tandem accelerator. Using

a foil or gas stripper, the negative ions are converted to positively charged ions with a distribution of charge states before they undergo another step of mass selection and final detection. While molecular isobars are generally destroyed in the charge exchange process, some atomic isobars are able to form stable negative ions and restrict the achievable detection limit for the isotopes of interest. Berkovits *et al.* [12] proposed the use of selective photodetachment to address this problem. Using a pulsed Nd:YAG laser at the fundamental wavelength 1064 nm for the  $^{59}\text{Ni}$ - $^{59}\text{Co}$  isobaric pair, Berkovits *et al.* achieved a depletion of two orders of magnitude for the  $^{59}\text{Co}$  negative ions compared to  $^{58}\text{Ni}^-$  *within the laser pulse*. However, the *overall* suppression of the  $^{59}\text{Co}$  isobaric background was very small due to the pulsed laser duty cycle of only  $10^{-4}$  [12]. This technique was advanced by Liu *et al.* [13] by performing the selective photodetachment in a buffer gas filled radio frequency (RF) quadrupole ion guide (henceforth referred to as the cooler). The significantly longer interaction time, generated by interaction of a continuous wave (cw) laser beam with the slow negative ions of only few eV kinetic energy inside the cooler, yielded more than two orders of magnitude overall depletion of  $^{59}\text{Co}$  in a continuous ion beam. In a recent study [14] it has been shown that selective depletion of Co in the cooler could be as high as a factor  $10^4$ , while the intensity of the desired Ni beam was only reduced by about 20% under the same conditions.

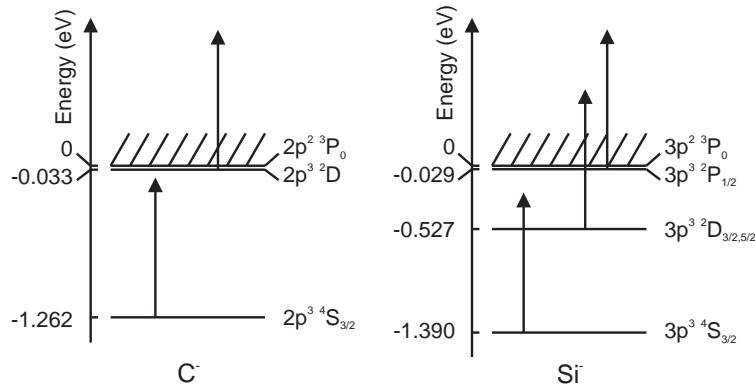
In this work we have investigated how the method developed by Liu *et al.* [13], can be applied to produce pure ground state beams of negative ions for fundamental studies. The method was tested on negative ions of Si and C. However, it is universal and can be applied to any atomic or molecular negative ion that possesses bound excited states.

## 2. Experimental method

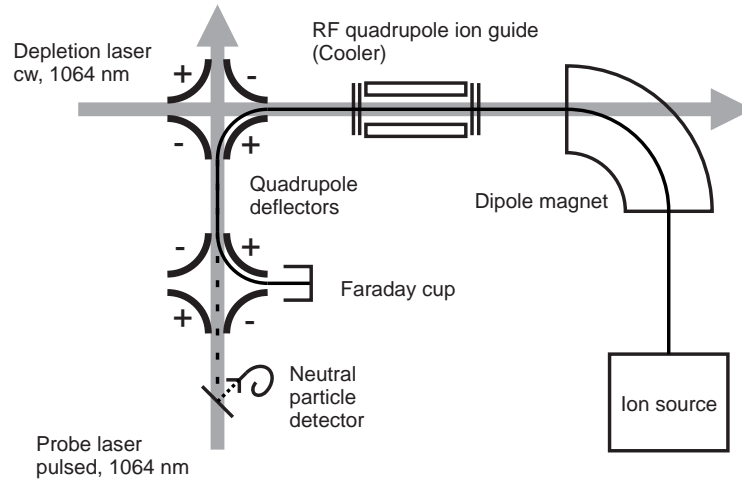
The energy levels of  $\text{C}^-$  and  $\text{Si}^-$  are shown in figure 1. These two negative ions were selected for this study because their excited states can be photodetached by the fundamental radiation from a Nd:YAG laser at 1064 nm (1.165 eV), whereas the ground states are unaffected by the laser radiation. However, there is a significant difference between the two ions.  $\text{C}^-$  exhibits only one high-lying excited state, the  $^2\text{D}$  state very loosely bound at 0.033 eV below the continuum.  $\text{Si}^-$ , on the other hand, has two excited states of differing character: a similarly high-lying loosely bound  $^2\text{P}$  state at -0.029 eV and a much more strongly bound  $^2\text{D}$  state at -0.527 eV. Differences in depleting the excited populations in the two ions are thus expected.

A sketch of the experimental setup is shown in figure 2. Negative ions were produced in a cesium sputter ion source from a sample containing 70% SiC and 30% C. The ions were accelerated to about 5 keV kinetic energy and mass selected in a  $90^\circ$  sector magnet with a radius of 0.61 m. A mass resolution of  $m/\Delta m = 200$  ensured appropriate selection of the desired element.

Using a series of electrostatic electrodes, the ions were decelerated to energies below 20 eV directly before injection into the cooler. The center-of-mass collision energies for 20 eV  $\text{Si}^-$  and  $\text{C}^-$  ions colliding with stationary He atoms are 2.5 and 5 eV, respectively.



**Figure 1.** Energy levels of C and Si. The arrows indicate the photon energy (1.165 eV) of the fundamental radiation from a Nd:YAG laser that was used in the experiment. The photon energy is less than the binding energy of the ground state in both Si<sup>-</sup> and C<sup>-</sup>.



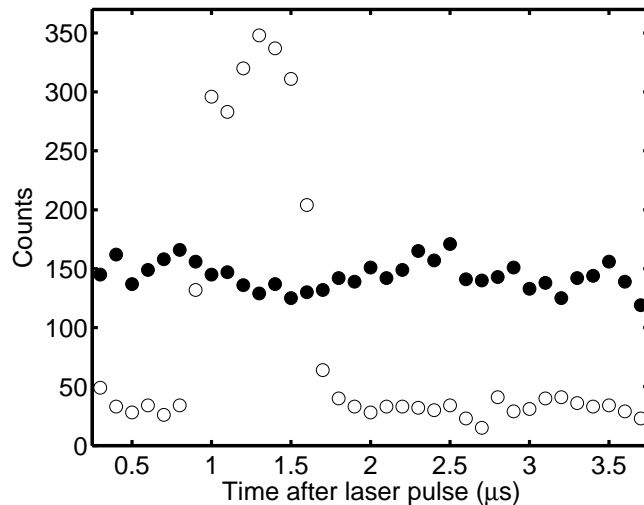
**Figure 2.** The experimental set-up showing the depletion laser passing through the ion cooler and the probe laser applied in the probe region between the two quadrupoles. Both laser powers are measured external to the vacuum system.

The ion guide, operated with helium gas as a cooling medium, was formed by four rod-electrodes of 40 cm length to which a radio frequency field of 2.86 MHz was applied. Four additional electrodes were used to create a weak gradient DC field that pushed the ions through the ion guide. A 3 mm diameter aperture was placed at the entrance to the cooler and a 2 mm diameter aperture at the exit. At the exit of the ion guide, the ions were re-accelerated to their initial kinetic energy. A complete description of the cooler is given elsewhere [15]. The radiation from a cw Nd:YAG laser operating at the fundamental wavelength of 1064 nm was overlapped with the ion beam inside the cooler to provide photodetachment. This laser is denoted as the depletion laser. The interaction time between the ions and the depletion laser was on the order of milliseconds [13]. This ion retention time inside the cooler was achieved at helium buffer gas pressures

of  $10^{-2}$  to  $10^{-1}$  mbar.

The population of the excited states in the ion beam after the cooler was probed using a second laser in pulsed operation and a subsequent time-resolved detection of the neutral particles produced. The probing photodetachment interaction took place between two quadrupole deflectors arranged behind the cooler as shown in figure 2. The first deflector bent the ion beam by  $90^\circ$  out of the path of the depletion laser beam, and the second quadrupole deflector guided the ions into a Faraday cup, where the transmitted ion beam current could be measured. In the 19 cm long drift region between the two quadrupole deflectors the ion beam was overlapped with the laser beam from a 20 Hz pulsed 1064 nm Nd:YAG laser. This laser is denoted as the probe laser. Apertures of 3 mm diameter were mounted on the quadrupole deflectors to define the overlap between the probe laser and the ion beam. The neutrals created by photodetachment in the probe region continued on their trajectories through the second deflector and then impacted onto a neutral particle detector. The detector was composed of a glass plate with a conductive coating, placed in the path of the neutrals. Neutrals impinging on the glass plate produced secondary electrons that were accelerated to and detected by channel electron multiplier (CEM). The detector assembly is described in detail elsewhere [16]. The pulses from the CEM were amplified and converted to TTL pulses. These were recorded by a multichannel scaler (FAST ComTec Model P7882) in order to obtain the time structure of the signals. A photo diode was used to trigger the data collection on the pulses from the 20 Hz probe laser. As illustrated in figure 1, the photon energy of the 1064 nm laser radiation was sufficiently large to photodetach the excited states but small enough to leave the ground state negative ions unaffected for both  $\text{Si}^-$  and  $\text{C}^-$ . Thus, this detection method is state selective. The excited states in the carbon and silicon negative ions could be detected and the relative depletion of the excited populations could be measured.

During the experiment the ion current was continuously monitored using the Faraday cup at the end of the second quadrupole deflector (see figure 2). The  $\text{Si}^-$  ion current was typically 60 pA with the cooler turned off and 35 pA with the cooler turned on. For  $\text{C}^-$ , the corresponding values were 170 and 35 pA, respectively. The power of both lasers was monitored when the laser beams exited the vacuum chambers through high transmission windows. The depletion laser power was also frequently measured at the output of the laser. Powers between 0 - 2 W out of the laser were used in the experiment, controlled by adjusting the pump power and by using neutral density filters. The actual power injected into the cooler was about 2 to 2.5 times higher than that measured after the magnet, determined by investigating the losses on windows and apertures along the laser path. The average power of the pulsed probe laser was about 16 mW, measured after the beam had passed the vacuum chamber. This corresponds to 0.8 mJ pulse energy at 20 Hz repetition rate.



**Figure 3.** Neutral detector counts as a function of time, measured for  $C^-$  beams without ( $\circ$ ) and with ( $\bullet$ ) the cooler in operation. Each curve corresponds to the sum of counts generated by 10 000 probe laser pulses.

### 3. Results and Discussion

#### 3.1. Carbon

First, the depletion of the ions in the excited  $^2D$  state in the  $C^-$  beam emitted from the ion source was investigated. The beam was transmitted through the cooler while not in operation, i.e. no buffer gas and all the DC and RF voltages turned off. By applying the probe laser, the excited  $^2D$  ions in the  $C^-$  beam were photodetached between the two quadrupole deflectors. The number of neutrals detected as a function of time is shown in figure 3 ( $\circ$ ). The peak in the signal between 0.9 and 1.7  $\mu s$  results from photodetachment induced by the probe laser between the two quadrupole deflectors. This time interval corresponds to the time of flight for an ion from the region between the quadrupole deflectors to the neutral particle detector. With uniform ion and laser beams a flat-topped distribution would be expected. The deviation from this shape is ascribed to a non uniform spatial overlap between the ion and the probe laser beams. The continuous background signal, i.e. the signal before 0.9 and after 1.7  $\mu s$ , is due to collisional detachment of both ground state and excited  $C^-$  ions by the residual gas in the probe region. Since only the  $C^-$  ions in the excited  $^2D$  state were photodetached by the probe laser, the number of counts above this background between 0.9 and 1.7  $\mu s$  serves as a measure of the excited population.

When the cooler was put into operation by inserting the buffer gas and switching on the RF and DC voltages, a substantial reduction of the population in the  $^2D$  excited state of  $C^-$  was observed. Figure 3 ( $\bullet$ ) shows the data obtained with about  $2 \times 10^{-2}$  mbar He pressure and 14 V on the DC electrodes in the cooler. As can be seen, there is no visible increase in the signal between 0.9 and 1.7  $\mu s$ . However, the background level

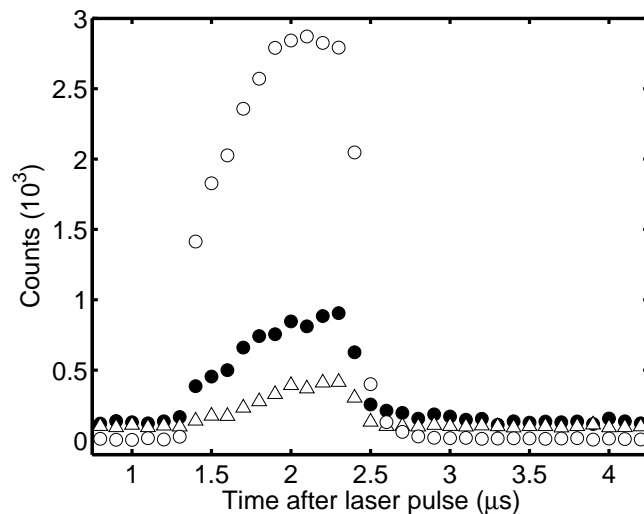
is substantially higher than in the case when the cooler was turned off. This was caused by increased collisional detachment, mainly from the ground state  $C^-$  ions, induced by the helium gas that leaked from the cooler into the probe region between the two quadrupole deflectors. As the vacuum system did not include a differential pumping section immediately after the cooler, the background pressure in the probe chamber increased from the  $10^{-8}$  mbar level to the  $10^{-6}$  mbar level with the cooler in operation. By comparing the measurements with the cooler on and off, a complete suppression of the population of the excited state could be deduced, which is quantitatively determined to within 6% (one sigma). The large uncertainty is due to the relatively large background signals from collisional detachment in the probe region. The total depletion of the loosely bound excited state was achieved by detachment inside the cooler through collisions with the buffer gas. This agrees with our expectation since the binding energy of the  $^2D$  excited state of  $C^-$  is only 33(1) meV. Photodetachment was therefore not needed to achieve a complete depletion of the excited state in  $C^-$ .

### 3.2. Silicon

The same experimental scheme was repeated for  $Si^-$  ions, with the addition of the depletion laser. Figure 4 shows three data sets recorded, without any normalization, with the cooler off ( $\circ$ ), cooler on ( $\bullet$ ), and cooler on combined with depletion laser ( $\Delta$ ). Because of the larger mass and thus smaller velocity, the neutral Si atoms produced by the probe laser were detected later than the C atoms, between 1.4 and 2.6  $\mu s$  after the probe laser pulse, as seen in figure 4. As in the experiment with  $C^-$  an increase of the background level can be observed in the data sets when the cooler is on. The increase is of the same size as in the  $C^-$  but not as obvious in the figure since the photodetachment signal from  $Si^-$  is an order of magnitude larger. It can also be noted that the background level decreases when the depletion laser is applied. This decrease could be explained if the excited ions have a larger cross section for collisional detachment than the ground state ions. With the cooler in operation, the number of  $Si^-$  ions in the excited states, measured by the probe laser, was reduced substantially compared to the situation with the cooler off. Interaction with the cw laser beam in the cooler further depleted the population in the excited states. The depletion laser power used for the data in figure 4 was 50 mW measured after the vacuum chamber.

The effect of the cooler without the depletion laser is discussed first. The cooler was put on with a He buffer gas pressure of about  $2 \times 10^{-2}$  mbar and a DC voltage of 14 V. The population in the  $^2P$  and  $^2D$  excited states of  $Si^-$ , and hence, the photodetachment signal, was then decreased due to collisional detachment in the cooler, similar to the case of  $C^-$ . The detection method can neither distinguish the two excited states nor provide information on the detection probability for each state. However, in the case of  $C^-$ , essentially all negative ions in the  $^2D$  excited state with a binding energy of 33(1) meV were removed by collisions in the cooler. Because the  $^2P$  state of  $Si^-$  has a similar binding energy of 29(5) meV, complete reduction of the  $Si^- (^2P)$  ions by collisional detachment in





**Figure 4.** Neutral detector counts as a function of time, measured for silicon with cooler off ( $\circ$ ), cooler on ( $\bullet$ ), and cooler on plus the depletion laser interacting with the ions inside the cooler ( $\triangle$ ). This data was recorded with 50 mW depletion laser power (measured after the magnet), a He pressure of  $2 \times 10^{-2}$  and 14 V on the DC electrodes in the cooler.

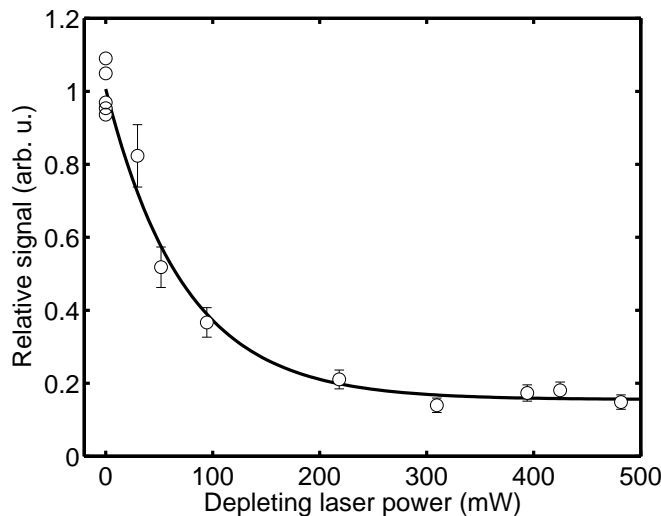
the cooler can also be expected. Thus, the observed reduction of the excited ions, when the cooler was turned on but without the depletion laser, was most likely a combination of two effects: complete depletion of the  $^2P$  state and partial depletion of the  $^2D$  state that has a much higher binding energy (0.527 eV). It seems as if a significant fraction of the ions in the  $^2D$  state have survived the collisions in the cooler.

When the depletion laser was added, the remaining population in the  $^2D$  excited state was further reduced (see figure 4). For a given laser beam flux  $\Phi$  (photons $\times$ cm $^{-2}$  $\times$ s $^{-1}$ ) and photodetachment cross section  $\sigma$  (cm $^2$ ), the fraction of negative ions not neutralized by the laser is given by

$$\frac{n}{n_0} = e^{-\sigma\Phi t} \quad (1)$$

where  $n_0$  is the initial number of negative ions,  $n$  is the number of remaining ions, and  $t$  is the laser-ion interaction time. Figure 5 shows the relative excited  $^2D$  state population as a function of the depletion laser power, normalized to the value without the depletion laser. The data were obtained when the cooler was operated at a He pressure of about  $2 \times 10^{-2}$  mbar.

Variations of the signal due to fluctuations in the probe laser power, ion-laser overlapping, and ion beam intensity was investigated by recording multiple data points with the depletion laser turned off. The datapoints were found to vary less than 10%; this error combined with the statistical error is indicated in figure 5. With the depletion laser power corresponding to 50 mW measured after the magnet, about half of the remaining  $^2D$  population was removed. This corresponds to the data shown as triangles in figure 4. If the dependence in figure 5 were purely exponential, increasing the laser

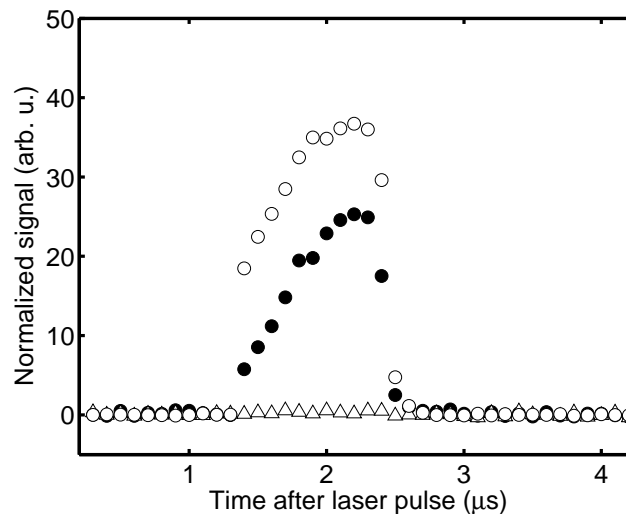


**Figure 5.** Relative population in the  $^2\text{D}$  excited state of  $\text{Si}^-$  as a function of the depletion laser power, obtained with a He pressure of  $2 \times 10^{-2}$  and 14 V on the DC electrodes in the cooler.

power by a factor of 10 would result in up to 99.9% depletion. However, the measured data indicate that the photodetachment in the cooler was saturated and the achievable depletion was limited to about 85%. A fit of an exponential plus a constant offset of 15% agrees with the data, as shown in figure 5.

The observed laser power dependence suggests that it is not possible to deplete excited ions by simply increasing the laser power. The offset component in figure 5 is thought to arise from the excited  $\text{Si}^-$  ions not sufficiently cooled and slowed down. Their transit time through the cooler and the effective interaction time with the laser radiation were thus too short. Consequently, this fraction of the ions could not be efficiently photodetached in the cooler. However, the photodetachment efficiency could be significantly improved by varying the operation parameters of the cooler. Increasing the buffer gas pressure results in more thorough cooling of the ions and thus more excited ions removed. In addition, reducing the longitudinal DC electric field that pushes the ions through the cooler can increase the ion residence time in the cooler and thus the photodetachment efficiency.

Figure 6 shows the data obtained when the He pressure was increased to  $3 \times 10^{-2}$  mbar and with a voltage of about 10 V applied to the DC electrodes in the cooler. This is about 30% smaller than the voltage used for the data presented in figures 4 and 5. The three data sets shown were recorded with the cooler off ( $\circ$ ), cooler on only ( $\bullet$ ), and cooler on plus the depletion laser ( $\triangle$ ). The depletion laser power was about 600 mW measured after the dipole magnet, which corresponded to about 2 W at the output of the laser. The signals have been normalized with respect to the ion current. It can be seen that the excited  $\text{Si}^-$  ions not detached through collisions in the cooler (closed circles) were almost completely removed by the depletion laser (triangles). Quantitatively, the



**Figure 6.** Neutral detector counts as a function of time, measured for silicon with cooler off ( $\circ$ ), cooler on ( $\bullet$ ), and cooler on plus the depletion laser ( $\triangle$ ). The data have been normalized with respect to the ion current. Each curve corresponds to the integrated signal from 10 000 laser pulses. This data was recorded with 600 mW depletion laser power (measured after the magnet), a He pressure of  $3 \times 10^{-2}$  and 10 V on the DC electrodes in the cooler.

photodetachment efficiency was determined to be 98(2)%. It is important to note that the laser power needed to achieve this degree of depletion varies with the experimental arrangement. It depends on the transit time through the cooler, the spatial distribution of the ion and laser beams, and possible losses of laser power along the path between the laser, the cooler and the final power meter. By adding the depletion by collisional detachment without the depletion laser (i.e. the difference between cooler off ( $\circ$ ) and cooler on only ( $\bullet$ ) in figure 6), the total reduction of the ion populations in the excited states was 99(1)%.

#### 4. Summary and Outlook

The effects of both collisional detachment and photodetachment in a gas filled quadrupole ion guide have been investigated using  $C^-$  and  $Si^-$  ions with the aim to produce pure ground state negative ion beams. Selective photodetachment of the excited ions was obtained with 1064 nm photons from a Nd:YAG laser that could leave the ground state ions unaffected. The population in the excited states was detected using a novel detection scheme also based on photodetachment by 1064 nm photons. We have shown that the loosely bound  $^2D$  excited state in  $C^-$  can be very efficiently removed by collisions with helium molecules within the RF quadrupole ion guide. A complete reduction of the excited state population in the  $C^-$  beam through collisional detachment alone was observed and can be quantitatively determined to within 6% (one sigma). A more sensitive detection scheme will be necessary to detect the residual excited ions

with less uncertainty. In particular, the background signal from collisional detachment in the probe region could be substantially reduced by introducing a differential pumping system that prevents helium gas from leaking into the probe region.

$\text{Si}^-$  exhibits two significantly different bound excited states. We expect the  $^2\text{P}$  state, which has a very low binding energy similar to that of the  $\text{C}^-$  ( $^2\text{D}$ ) state, to be completely depleted by collisional detachment in the cooler. Accordingly, the photodetachment signal from  $\text{Si}^-$  was decreased by a factor of two when the cooler was turned on, an effect attributed to collisional detachment primarily of the ions in the  $^2\text{P}$  state and possibly some of the ions in the stronger bound  $^2\text{D}$  state. With a cw laser beam of less than 2 W the remaining population in the  $^2\text{D}$  state was decreased by 98(2)%. This indicates that the selective photodetachment in the cooler is extremely efficient. The total depletion of the excited populations in the  $\text{Si}^-$  beam, including collisional detachment and photodetachment, is determined to be 99(1)%.

A further advantage of the technique should not be overlooked: In this study the negative ion beam cooler was primarily used to increase the interaction time between the ions and the laser radiation. At the same time, the energy spread of the negative ion beam will be reduced through collisions with the helium gas. This intrinsic property of the cooler will be further advantageous in many high resolution experiments, as the thermal spread of the ion beam can also limit the resolution in collision or laser spectroscopic experiments.

To conclude, a state selective depletion of negative ions in a gas filled quadrupole ion guide has been demonstrated. This novel technique to produce a pure ground state negative ion beam offers possibilities to enhance selectivity as well as accuracy in high precision experiments on atomic as well as molecular negative ions.

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