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A SEARCH FOR DOUBLY-CHARGED NEGATIVE IONS VIA ACCELERATOR MASS SPECTROMETRY

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Abstract

The Argonne FN tandem accelerator in conjunction with an Enge split-pole magnetic spectrograph has been used as a highly sensitive mass spectrometer to search for doubly-charged negative ions of ^{11}B , ^{12}C and ^{16}O . No evidence for the formation of these ions in an inverted sputter source and the subsequent acceleration in the tandem has been found. The following limits for the ratio of doubly-charged to singly-charged ions were measured:
 $X^{--}/X^{-} < 1 \times 10^{-15}$, ^{11}B ; $< 2 \times 10^{-15}$, ^{12}C ; $< 2 \times 10^{-14}$, ^{16}O . A relatively abundant formation of the short lived, metastable He^{-} ion in the sputter source has been observed.

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1. Introduction

It has long been an intriguing possibility to produce beams of doubly-charged negative ions for tandem accelerators. Since the energy is doubled in the first stage of the acceleration higher charge states at the terminal and a substantial overall increase in energy could be achieved. This would greatly extend the use of medium-sized tandem accelerators for heavy ion physics.

Reports on the observation of double-charged negative ions in the literature [1-7] are sporadic and as a whole inconclusive. The most encouraging results were reported in the early seventies [3] where beams of O^{--} , Te^{--} , Bi^{--} , F^{--} , Cl^{--} , Br^{--} and I^{--} with intensities ranging from 0.2 to 2 nA were observed on an ion source test bench. However, subsequent experiments by other groups yielded negative results, even when the experimental conditions were very similar. In particular, the I^{--} currents observed in ref. 3 were not confirmed [5] when the source material was spiked with radioactive iodine. Limits of $I^{--}/I^- < 10^{-5}$ could be set in this experiment. In another attempt [6] no evidence was found for O^{--} and F^{--} at a level of 10^{-6} as compared to singly-charged negative ions.

Since, at present, the situation is far from being settled it has been thought worthwhile to use the highly-sensitive technique of Accelerator Mass Spectrometry (AMS) for a search of doubly-charged negative ions. With this method one can reach detection limits which are many orders of magnitude lower than those of previous experiment. In the present experiment positive evidence for the production of a doubly-charged negative ions is obtained when the ion is observed in a heavy-ion detection system after acceleration in the tandem. This means that only the product of formation probability and survival probability (during the flight from the ion source to the terminal) can be measured. However, since one is probably dealing with very loosely

bound ions both contributions may be complex functions of a number of parameters and a disentangling might be very difficult under any circumstances. Our approach to this problem was simply to use conditions for routine operation of singly-charged negative ions at our FN tandem. The system was then set up to accelerate doubly-charged negative ions of ^{11}B , ^{12}C and ^{16}O and the ions were detected after acceleration with a sensitivity of 10^{-14} to 10^{-15} as compared to singly-charged negative ions.

2. Experimental procedure

The experiments were performed with the Argonne FN tandem accelerator. A schematic lay-out of the system used for AMS measurements is shown in fig. 1. This figure has been reproduced for convenience from ref. 8 where details of the AMS experiments at Argonne are described. In short, the main components are an inverted sputter source of the Chapman type [9], a 40° injection magnet, a 9 MV FN tandem with NEC accelerator and corona tubes and a pelletron charging system, a 90° analyzing magnet, and an Enge split-pole magnetic spectrograph equipped with an ionization-type focal plane detector.

With a tandem an unambiguous signature of doubly-charged negative ions can be obtained if one chooses to work only with ions fully stripped at the terminal. For a given terminal voltage, TV , the total energy gain for singly-charged negative ions is $(1 + Z) TV$, whereas it is $(2 + Z) TV$ for doubly-charged negative ions. With a redundant particle identification system such as the split-pole spectrograph which measures $B\rho$, ΔE and E_{total} one can very cleanly separate the desired particles from any background ions. In these measurements the spectrograph is positioned at zero degree and a thin stripper foil in the target chamber serves to further reduce background ions which happens to match the magnetic rigidity of the high-energy beam transport system.

In the following the sequence of measurements for the detection of $^{12}\text{C}^{--}$ ions is described ($^{11}\text{B}^{--}$ and $^{16}\text{O}^{--}$ were investigated accordingly). First $^{12}\text{C}^{--}$ were injected into the tandem set to a terminal voltage of 8 MV. 56 MeV $^{12}\text{C}^{6+}$ ions were selected with the high-energy transport system and the current measured in a biased Faraday cup in the target chamber of the spectrograph. The cup was then exchanged with a $100\text{ }\mu\text{g}/\text{cm}^2$ Au target and the Coulomb-scattered particles measured at 20° in the spectrograph. (A ΔE vs E_{total} spectrum from this measurement is shown in figure 2a.) The terminal voltage was then lowered in such a way that $^{12}\text{C}^{6+}$ ions originating from $^{12}\text{C}^{--}$ injection would gain exactly the same energy in the tandem:

$$TV(^{12}\text{C}^{--}) = TV(^{12}\text{C}^-) \times (1 + 6)/(2 + 6) = 7\text{ MV}.$$

At this condition $^{12}\text{C}^{6+}$ ions originating from $^{12}\text{C}^-$ injection can only reach 49 MeV. A spectrum measured with the high-energy beam transport system re-tuned for these ions is shown in fig. 2b. For the $^{12}\text{C}^{--}$ measurements, however, the transport system was not changed once it was set up with the 56 MeV $^{12}\text{C}^{6+}$ ions. In order to inject possible $^{12}\text{C}^{--}$ ions the 40° magnet field had to be lowered to $B(^{12}\text{C}^{--}) = B(^{12}\text{C}^-)/\sqrt{2}$. Besides $^{12}\text{C}^{--}$ ions this setting accepts $^{12}\text{C}^-$ ions from $^{12}\text{C}_2^-$ break-ups between preacceleration and magnet. This kind of molecular dissociation was, in fact, one of the main obstacles in searches for doubly-charged negative ions in previous, low-energy experiments [3,5,6]. For our particular setting of $^{12}\text{C}^{--}$ we also accept $^6\text{Li}^-$ ions. At the low-energy Faraday cup after the 40° magnet we observed a "mass-6" current of 50 pA, most of it presumably from the $^{12}\text{C}_2^-$ break-up. A ΔE vs E spectrum measured in the focal plane detector with the spectrograph at 0° is shown in fig. 2c. The only prominent peaks in the spectrum are from 39 MeV $^{12}\text{C}^{5+}$ ions produced by the C_2^- break-up process described above and from 28 MeV $^6\text{Li}^{3+}$ ions. Multiple charge exchange processes in the tandem produce a

"white" background of ^{12}C ions which is filtered with the 90° magnet. $^6\text{Li}^{3+}$ ions can be considered as a real beam since they are degenerate with a $^{12}\text{C}^{--}$ $\rightarrow ^{12}\text{C}^{6+}$ acceleration. The lower injection energy (132 keV as compared to 264 keV for $^{12}\text{C}^{--}$) is partly compensated by the lower energy loss of ^6Li in the terminal stripper foil (10 and 30 keV for ^6Li and ^{12}C , respectively). Therefore $^6\text{Li}^{3+}$ was used to fine-tune the terminal voltage. It turned out that it had to be lowered by 20 kV from the calculated value to get maximum count rate of $^6\text{Li}^{3+}$ ions ($\sim 200 \text{ sec}^{-1}$) in the spectrograph. The result of a 69 min run with this condition is shown in fig. 3. The power of background discrimination can clearly be seen since, despite a huge $^6\text{Li}^{3+}$ component, the interesting region where $^{12}\text{C}^{6+}$ ions from $^{12}\text{C}^{--}$ injection should show up is perfectly clean.

The set-up of $^{11}\text{B}^{--}$ and $^{16}\text{O}^{--}$ detection was done in the same way as for $^{12}\text{C}^{--}$. However, since there was no degenerate beam available for fine-tuning of the terminal voltage the correction for the calculated values as measured with the $^6\text{Li}^{3+}$ ions was used. The interesting regions in the ΔE vs E spectra were also background free, similar to the conditions shown in fig. 3.

III. Results and discussion

Table 1 summarizes the results of the relevant measurements. It is apparent that not a single count of doubly-charged negative ions was observed in any of the measurements. Although great care has been taken to set-up the system correctly for the detection of doubly-charged negative ions, there remain a few uncertainties. In particular, it is not clear how the transmission through the tandem is effected for these ions. Due to the higher energy at the terminal the ions are stiffer at the entrance of the high-energy tube, resulting in a reduced focussing strength. However, it seems unlikely

that this effect could reduce the transmission by more than an order of magnitude as compared to singly-charged negative ions. The ratio in the last column of the table was calculated from the counting rate of fully-stripped ions in the spectrograph and the current measured in the spectrograph Faraday cup when singly-charged negative ions were injected.

Our results indicate that no doubly-charged negative ions of B, C and O do exist. However, it is possible that a sputter source is not the best way to produce these ions. If metastable electron configurations play an important role in the formation of doubly-charged negative ions then it is likely that a charge exchange source would give higher yields. It is well known that He^- ions are metastable and can most efficiently be produced by a charge exchange reaction of He^+ ions in Li vapor. In order to check to what extent $^4\text{He}^-$ is formed in the sputter source we have set up our system to detect $^4\text{He}^{2+}$ ions in the spectrograph. Counting rates exceeding $10^3/\text{sec}$ were easily obtained. Since we have not observed any doubly-charged negative ions in one-hour running periods we conclude that the probability to form these ions is at least a factor of 10^6 smaller than for He^- ions. As mentioned in the introduction our measurement can only test the product of production and survival probability. The observation of abundant $^4\text{He}^{2+}$ ions together with the half-life of He^- ($T_{1/2} \approx 15 \mu\text{sec}$) sets an upper limit for the half-life of $^{11}\text{B}^-$, $^{12}\text{C}^-$ and O^- ions. However, it is also possible that more subtle effects such as for example the ionization of very weakly bound electrons ($E_{\text{Binding}} < 1 \text{ meV}$) in the electric field of the accelerator contribute to the destruction.

The present experiment has shown that very clean conditions can be established for a search of doubly-charged negative ions using the technique of accelerator mass spectrometry. Considering the impact of a positive result

on heavy ion tandem physics it might be worthwhile to extend this search to heavier elements. Since the fully stripping technique leads to the cleanest conditions one would like to do these searches with one of the new large tandem accelerators.

Table 1

Summary of measurements for the detection of doubly-charged negative ions.

Source Material	Singly-charged negative ions		Doubly-charged negative ions		X^{--}/X^- b)	
	Injector (μA)	Spectrograph (nA)	Injector ^{a)} (pA)	Time (min)	Spectrogr. (counts)	(10^{-15})
Boron	$^{11}B^-$, 0.36	$^{11}B^{5+}$, 105	$^{11}B^{--}$, <10	62	$^{11}B^{5+}$, 0	< 1
Graphite	$^{12}C^-$, 1.3	$^{12}C^{6+}$, 41	$^{12}C^{--}$, 50	67	$^{12}C^{6+}$, 0	< 2
Copper	$^{16}O^-$, 2.4	$^{16}O^{8+}$, 5.5	$^{16}O^{--}$, 10	40	$^{16}O^{8+}$, 0	<20

a) Negative ion current measured with the 40° magnet field set to $1/\sqrt{2}$ times the value for singly-charged negative ions. For the origin of this current see text.

b) The ratio is calculated from the fully stripped ions and is corrected for the increase in stripping efficiency at the terminal for doubly-charged negative ions.

References

- [1] W. K. Stuckey and R. W. Kiser, *Nature* 211 (1966) 963.
- [2] J. H. Fremlin, *Nature* 212 (1966) 1453.
- [3] H. Baumann, E. Heinicke, H. J. Kaiser and K. Bethge, *Nucl. Instr. and Meth.* 95 (1971) 389.
- [4] M. Anbar and G. A. St. John, *J. Am. Chem. Soc.* 97 (1975) 97.
- [5] L. Frees, E. Heinicke and W. S. Koski, *Nucl. Instr. and Meth.* 159 (1979) 105.
- [6] W. Goldstein, Ph.D. Thesis, University of Munich, 1979, unpublished.
- [7] R. W. Kiser, in *Topics in Curr. Chem., Instr. Inorg. Chem.* (Springer, Berlin-Heidelberg-New York, 1979) Vol. 85, p. 89.
- [8] W. Henning,, W. Kutschera, M. Paul, R. K. Smither, E. J. Stephenson and J. L. Yntema, *Nucl. Instr. and Meth.* 184 (1981) 247.
- [9] P. J. Billquist and J. L. Yntema, *Nucl. Instrum. Methods Phys. Res.* 178 (1980) 9.

Figure Captions

- Figure 1. Schematic layout of the Argonne FN tandem accelerator and the experimental system used in the AMS measurements.
- Figure 2. Isometric plot of the energy loss, ΔE , versus total energy signals, E_{total} , measured in the focal plane detector of the split-pole spectrograph. All three dimensions are linear. a) and b) are ^{12}C ions elastically scattered to 20° from a $100\text{ }\mu\text{g}/\text{cm}^2$ thick Au target. c) 10 min spectrum measured at 0° , when the system was set up to detect $^{12}\text{C}^{--}$ ions. The choice of the vertical scale allows for single counts to be visible.
- Figure 3. Two-dimensional density plot of the energy loss, ΔE , versus total energy, E_{total} , measured with the focal plane detector of the split-pole spectrograph for the detection of $^{12}\text{C}^{--}$ ions. The system was optimized for $^6\text{Li}^{3+}$ counting rate ($\sim 200\text{ sec}^{-1}$). The spectrum is accumulated for 69 minutes and the density threshold lowered to show single counts. Although the intense $^6\text{Li}^{3+}$ peak generates wide-spread background (pick-up, pulse height loss) the interesting area is perfectly clean.

Fig. 1

Fig. 2

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Fig. 3

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