ISBN: 978-3-95450-257-8 ISSN: 2222-5692 doi:10.18429/JACoW-ECRIS2024-MOP15

STUDY OF NOBLE GAS MEMORY EFFECT OF ECR3 AT ATLAS*

R. Scott†, J. McLain, R. C. Vondrasek, Argonne National Laboratory, Lemont, IL, USA
S. Bhattacharya, M. Paul, Racah Institute of Physics, Hebrew University, Jerusalem, Israel

Abstract

Over the past three decades a portion of the accelerated beam time at the Argonne Tandem Linac Accelerator System (ATLAS) has been reserved for ultra-sensitive detection of argon radioisotopes. A unique noble-gas accelerator mass spectrometry (NOGAMS) technique at ATLAS combines electron cyclotron resonance ion source (ECRIS) positive ion production, acceleration up to ~6 MeV/u and detection methods for separating isobars and other m/q contaminants. The ECR3 ion source was recently chosen for such experiments due to the limited scope of material introduced into the plasma chamber, inferring a lower background production compared to ECR2. A recent ^{39,42}Ar NOGAMS experiment has highlighted a need to understand the beam production of material that is no longer being actively introduced into the ECRIS, known as memory effect. A quantitative study of source memory was performed to determine the decay characteristics of argon in the ECR3 ion source. Results of this study as well as details of setup and operation of ECR3 for NOGAMS experiments are presented.

INTRODUCTION

The ATLAS facility at Argonne National Laboratory has provided heavy ion beams for nuclear physics experiments for over 40 years. ATLAS runs experiments 24 hours a day, 7 days a week with a typical ion beam species change once per week and maintenance time dispersed throughout. The ultra-sensitive noble-gas accelerator mass spectrometry NOGAMS [1,2] technique has been developed and improved at ATLAS over the past 30 years. The technique differs from conventional accelerator mass spectrometry (AMS) which, based on negative ion injection, cannot analyze noble-gas ions. Positive ions are produced from an electron cyclotron resonance ion source (ECRIS) followed by mass/charge (m/q) selection with a dipole magnet, acceleration to ~6 MeV/u using linear accelerator sections and delivery to the gas-filled Enge split-pole spectrograph [3] for ion detection, as well as isobaric and background separation (see Fig. 1).

The first detection of low concentrations of ^{81}Kr and ^{39}Ar at ATLAS occurred in 1992 [4] using the now retired ECR1 [5]. In 2002, ECR2 [6] was used to provide ion beams for ^{39}Ar detection in ocean samples for the study of ocean circulation, allowing smaller sample sizes than those required for low level counting (LLC) [7]. ECR2 was used again in 2015-16 to provide ion beams of low concentration ^{37,39}Ar for a measurement of ^{36}Ar(n,γ)^{37}Ar and ^{38}Ar(n,γ)^{39}Ar neutron-capture cross sections [8]. ECR3 [9] was used for the most recent NOGAMS detection of ^{39,42}Ar in a study of neutron induced reactions at the National Ignition Facility (NIF) [1,10,11]. Parameters of ECR3 operation are discussed later. Table 1 provides a summary of a few of the ion source operating parameters of the mentioned NOGAMS experiments.

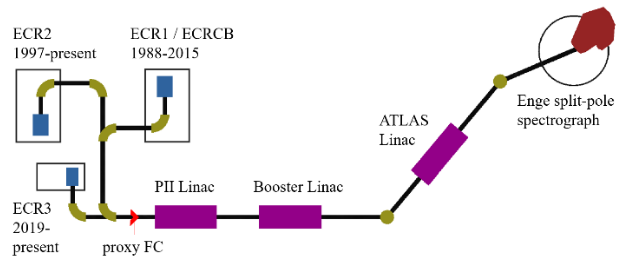

Figure 1: ATLAS accelerator NOGAMS layout.

Part of the recent ECR3 NOGAMS experiment was devoted to detection of ^{42}Ar in a NIF shot gas sample. Prior to running the NIF sample, a sample with a high concentration of ^{42}Ar [12] was used in ECR3 for identification and calibration with the detection system. Unexpected, but verified ^{42}Ar counts were observed with the NIF sample following the calibration run. Ion source memory of the calibration gas was suspected to produce those counts. A study of the ion source memory was performed, replicating the same ion source operating conditions. These results are provided within.

NOGAMS BEAM CONTAMINATION

Early NOGAMS experiments were predicated on a high sensitivity ^{39}Ar/Ar measurement, below naturally occurring Ar (8.1×10^{-16}), of multiple samples in ~ one week. A 100 eμA $^{40}Ar^{8+}$ beam, with a ^{39}Ar/^{40}Ar concentration of 1×10^{-17} at the ion source would yield 1 count/hr, assuming a typical 35% transmission from ECRIS Faraday cup (FC) to experimental station. At these low rates, m/q beam contaminants, such as the isobar ^{39}K, can limit the measurement due to pile up at the detector. The work done to reduce ^{39}Ar m/q contamination in ECR2, including quartz liners and cleaning methods, has been presented here [13]. In 2015, higher ^{39}Ar concentration samples $\geq 1 \times 10^{-13}$ were to be measured. Therefore, a lower beam intensity at the ion source could achieve sufficient detection. ECR2 was run at low RF power (22 W) without support gas. With a weaker plasma and less electron interaction with the plasma chamber surfaces, this operation resulted in significantly lower contamination rates of $^{39}K^{8+}$, without using any of the previous experiments mitigation methods (see Tables 1, 2).

Based on these results, ECR3 was chosen for the 2021 ^{39}Ar and ^{42}Ar series of experiments. ECR3, which was commissioned at ATLAS in 2019, has no history of solid

*This work was supported by the U.S. Department of Energy, Office of Nuclear Physics, under Contract No. DEAC02-06CH11357 and used resources of ANL's ATLAS facility, an Office of Science User Facility.
† rscott@anl.gov

ISBN: 978-3-95450-257-8 ISSN: 2222-5692 doi:10.18429/JACoW-ECRIS2024-MOP15

Table 1: NOGAMS Operational History at ATLAS

Year Measured	Ion Source	RF Power (W)	Support Gas	Ar Source Intensity (pnA)	Ar Cons. Rate (cc-STP/hr)	Isotopes Detected	Sample Detection Limit
1992	ECR1	105	oxygen	1312	1	^{39}Ar, ^{81}Kr	8×10^{-16}
2003	ECR2	300	nitrogen	13500	0.2	^{39}Ar	4×10^{-17}
2015	ECR2	22	none	250		^{39}Ar	9×10^{-16}
2016	ECR2	50	none	1500		^{37}Ar	
2021	ECR3	6	none	37.5	0.007	^{39}Ar,^{42}Ar	1×10^{-15}
2023	ECR3	7	none	29.4	0.007	^{39}Ar,^{42}Ar	

material introduction, and was expected to have similar or less $^{39}Ar^{8+}$ m/q contamination than ECR2. ECR3 was operated at 7 W in 2023 for the NIF ^{39}Ar and ^{42}Ar detection experiments. Even though ECR3 was run at lower RF power, $^{39}K^{8+}$ contamination rates were twice as high, and $^{34}S^{7+}$ contamination rates were over 100 times higher than when using ECR2 (see Table 2). In both 2015 and 2023, the full contaminant rates were measured with no blocking at the detector. In 2023, with further blocking put in place, 0 cps/µA for ^{39}K and 74 cps/µA were observed with no decrease in the ^{39}Ar rate. Perhaps the larger volume (1.37 l), diameter (7.6 cm) and radial pumping of the plasma chamber of ECR2 compared to ECR3 (0.53 l), (6.4 cm) with pumping only through the extraction hole, play a role in the lower raw potassium rates of ECR2. The larger ECR3 $^{34}S^{7+}$ ($M_0/\delta M$ = 1:270) contamination rates can mostly be explained by the inherently higher m/q discrimination of the ECR2 low energy beamline, when compared to that of ECR3. For ^{42}Ar detection, the isobar ^{42}Ca rate was easily separated at the detector, while $^{63}Cu^{12+}$ was considerably higher but still separated at the detector to provide a clean ^{42}Ar rate.

ECR3 NOGAMS OPERATION

As described for contamination reduction, the ECR3 ion source is run at low RF power, about 7 W, shifting the charge distribution lower than usual operation. The source peaks at 8+ with helium support and 62 W RF power, but peaks at 5+ at 7 W without support gas. Ideally, the charge state with the highest intensity would be used for faster sample concentration measurements. The first accelerating section of ATLAS has a m/q acceptance below 7/1 and the masses to be used for the NIF experiments are 38,39,40, and 42. The charge state $^{42}Ar^{6+}$ falls out of this acceptance, and $^{42}Ar^{7+}$ is inseparable in the accelerator from $^{12}C^{2+}$. This leaves 8+ as the lowest charge state without high intensity m/q contaminants.

Figure 2 shows the simple gas handling system used to feed a sample of interest into the ECR3 ion source. The gas of interest is fed directly into the plasma chamber through the injection side of ECR3 with an Agilent model 9515106 [14] variable leak valve with fine motor control. Gas sample sizes as low as 5 cc-STP can be expected with some samples containing a mixture of known proportions of ^{38}Ar and ^{40}Ar. The smallest sample size can be run multiple hours without a significant enough pressure decrease in the gas line to require a leak valve adjustment to maintain beam intensity, with a maximum consumption rate of 0.007 cc-STP/hr measured for Ar^{8+} at 300 enA.

The sample cylinder change sequence, intended to limit cross contamination of samples is as follows: leave RF on, stop extraction of beam, shut leak and sample cylinder valves, pump-out gas volume with independent pump to millitorr range, and stop pump-out. Then, while continuously purging the volume with boil-off nitrogen from the ATLAS cryogenic system, remove sample cylinder, attach new sample cylinder, stop continuous purge, perform pump-purge cycle on gas volume 10 times ending on pump-out, shut valve leading to pump and purge valves, open leak valve to ECR3 while maintaining a maximum ion source pressure of 2×10^{-7} Torr until leak valve is fully open. When source pressure is below 1.4×10^{-8} Torr (after about 15 min), shut leak valve, open new sample cylinder valve, resume beam extraction, and open leak valve to achieve desired total Ar^{8+} intensity.

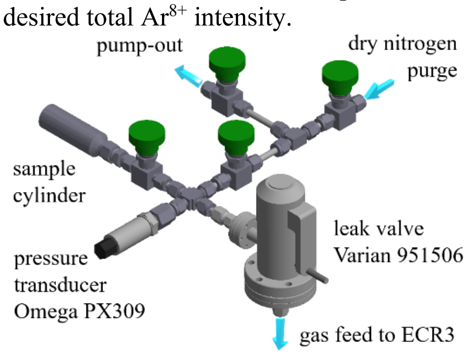

Figure 2: ECR3 NOGAMS gas handling system.

ECR3 ARGON GAS MEMORY EFFECT

ECR ion source memory is described as the continued ion beam production resulting from previous feeding of materials which are no longer being actively introduced into the plasma chamber. For argon gas, the beam intensity will follow a decay curve as residual material is consumed after the leak valve is shut. At the end of a recent NOGAMS experiment, two series of measurements were taken looking for the presence of ^{42}Ar in a sample resulting from neutron-induced reactions on ^{40}Ar seeds at the National Ignition Facility (NIF) [11]. In between these meas-

ISBN: 978-3-95450-257-8 ISSN: 2222-5692 doi:10.18429/JACoW-ECRIS2024-MOP15

Table 2: NOGAMS ^{39}K and ^{34}S Contamination at ATLAS

Ref	Date	Ion Source	ECR Source Ar8+ Intensity (eμA)	39K Detector Rate/Ar Intensity (cps/eμA)	34S Detector Rate/Ar Intensity (cps/eμA)	Configuration
[13]	Jun 2007	ECR2	83	50602		no treatment
[7,13]	Aug 2001	ECR2	75	17333		open quartz liner
[7,13]	May 2002	ECR2	76	129		closed quartz liner
[13]	Jun 2007	ECR2	98	459		ultrapure alum. thin liner
[13]	Apr 2008	ECR2	55	27273		ultrapure alum. coated PC + open quartz + cleaning
[8]	Oct 2015	ECR2	5.5	30	3	low rf power, no support gas
	Oct 2023	ECR3	0.16	66	355	low rf power, no support gas

-urements, a calibration sample with a concentration of 1.6 × 10^{-12} $^{42}Ar/Ar$ was fed into ECR3 for 4.9 hours to confirm the setting of the spectrograph magnetic field. While the first series detected no presence of ^{42}Ar, the second series detected 4 legitimate ^{42}Ar counts. Even though an added purge with UHP ^{nat}Ar was performed between the calibration and NIF sample change, the four counts can be suspected to be a result of memory effect from running the calibration sample. Investigating the origin of the ^{42}Ar counts was considered important since they would imply a fast two neutron capture process if they originated from the NIF sample.

A quantitative study of source memory was conducted following the experiment, replicating its operating conditions and run sequence. The spectrograph and accelerator were unavailable for this study, so measurement was limited to the FC after m/q separation using the dipole magnet following extraction from ECR3. Highly enriched ^{38}Ar (99.96%) [15] was used as a surrogate for the ^{42}Ar calibration sample, ^{40}Ar (99.99%, ^{38}Ar 0.004%) [15] was used to replicate the NIF sample, and the FC was used as a proxy (see Fig. 1) for the detection system. If the ^{38}Ar enriched sample is run with same total Ar beam intensity, running conditions and period as the ^{42}Ar calibration sample, then their decay curves would be the same. The ratio of total $^{42}Ar/^{38}Ar$ memory ions over a specific period will equal the ratio of the total ions $^{42}cal/^{38}enr$ measured (normalized for time) prior to decay. This relationship can be expressed as

$$\frac{\Sigma 42_{mem}}{\Sigma 38_{mem}} = \frac{\Sigma 42_{cal}}{\Sigma 38_{enr}} * \frac{t_{38\,enr}}{t_{42cal}} ,$$

where $\Sigma 42_{cal}$ is the number of counts detected from the calibration sample over 4.9hours, and $\Sigma 42_{mem}$ is the expected detector counts from memory after running the calibration sample. Solving for $\Sigma 42_{mem}$ we get

$$\Sigma 42_{mem} = \Sigma 38_{mem} * \frac{\Sigma 42_{cal}}{\Sigma 38_{enr}} * \frac{t_{38\,enr}}{t_{42cal}} .$$

For the memory test, enriched $^{38}Ar^{8+}$ was run and monitored for 4 hours with a beam intensity of 235 enA, equalling the total Ar^{8+} intensity achieved during the experiment. The number of ^{38}Ar ions $\Sigma 38_{enr}$ was derived from the charge current on the FC and the run time. The cylinder sample change sequence described earlier was performed, installing next ^{40}Ar (99.99%) and opening the leak valve to achieve 235 enA of $^{40}Ar^{8+}$. The $^{38}Ar^{8+}$ intensity decay (see Fig. 3) and stable $^{40}Ar^{8+}$ intensity were monitored for the same period as the NIF sample. The number of ^{38}Ar memory ions $\Sigma 38_{mem}$ was derived from the integral sum of beam intensity over the decay period. Assuming no memory effect or background Ar contribution, the enriched ^{40}Ar would yield $^{38}Ar^{8+}$ intensity of 9.4 epA. For the FC measurement a Keithley 6485 digital picoammeter with zero correction was used [16].

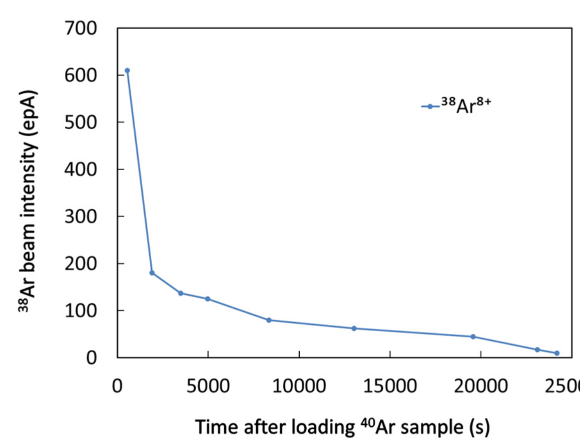

Figure 3: Decay curve of ^{38}Ar on the proxy FC following ECR3 operation with enriched 99.96% ^{38}Ar gas at 235 enA on the same FC.

The resulting value of expected memory counts $\Sigma 42_{mem}$ was 0.8. Based on this estimate (three valid ^{42}Ar counts and one memory count) the ^{42}Ar yield in the NIF sample could be higher than expected. With such low statistics, all 4 counts coming from memory cannot be strictly ruled out, but a proposal for a repeat experiment is justified.

CONCLUSION

NOGAMS experiments at ATLAS have seen a large reduction in beam contamination from background materials using lower RF power from the ECR ion sources with a reduced Ar beam intensity that does not affect measurement of ^{39,42}Ar samples with concentrations > 1 × 10^{-15}. ECR3 has recently been used for NOGAMS and has demonstrated higher ^{39}K and ^{34}S contamination rates per eμA of Ar^{8+} ions when compared to ECR2, but rates were still low enough for the ^{39}Ar detection levels needed. Live ^{42}Ar was detected for the first time using NOGAMS at ATLAS from a calibration sample with a ^{42}Ar/Ar concentration of 1.6 × 10^{-12}. A NIF sample installed in ECR3 following the calibration sample resulted in unexpected ^{42}Ar counts. A study was conducted to see if these counts were from source memory, with results indicating some could have come from a higher yield in the NIF sample. ATLAS beam time dedicated to a longer measurement of ^{42}Ar yield in the NIF sample has been requested and approved for November 2024.

ACKNOWLEDGEMENTS

M. Paul gratefully acknowledges support from USA-Israel Binational Science Foundation (grant 2020136) and from Pazy Foundation (Israel) under grant ID231-2020.

REFERENCES

[1] M. Paul *et al.*, "^{40}Ar proposed as probe of neutron-induced reactions in a high-density stellar-like plasma at the National Ignition Facility," *EPJ Web of Conferences*, vol. 279, 13004, 2023. doi:10.1051/epjconf/202327913004

[2] M. Paul *et al.*, "Positive-ion accelerator mass spectrometry at ATLAS: Peaks and pits," *Nucl. Instrum. Methods B*, vol. 456, pp. 222-229, 2019. doi:10.1016/j.nimb.2019.04.003

[3] M. Paul *et al.*, "Heavy ion separation with a gas-filled magnetic spectrograph," *Nucl. Instrum. Methods A*, vol. 277, pp. 418-430, 1989. doi:10.1016/0168-9002(89)90771-7

[4] W. Kutschera *et al.*, "Long-lived noble gas radionuclides," *Nucl. Instrum. Methods B*, vol. 92, pp. 241-248, 1994. doi:10.1016/0168-583X(94)96013-5

[5] R. Pardo, "Operating experience of an ECR ion source on a high voltage platform," *Nucl. Instrum. Methods B*, vol. 40-41, Part 2, pp. 1014-1019, 1989. doi:10.1016/0168-583X(89)90529-6

[6] M. Schlapp *et al.*, "A new 14 GHz Electron-Cyclotron-Resonance Ion Source (ECRIS) for the Heavy Ion Accelerator Facility ATLAS" *Review of Scientific Instruments*, vol. 69, pp. 631-633, 1998. doi:10.1063/1.1148725

[7] P. Collon *et al.*, "Development of an AMS method to study oceanic circulation characteristics using cosmogenic ^{39}Ar," *Nucl. Instrum. Methods B*, vol. 223-224, pp. 428-434, 2004. doi:10.1016/j.nimb.2004.04.081

[8] M. Tessler *et al.*, "Stellar ^{36,38}Ar(n,γ)^{37,39}Ar Reactions and Their Effect on Light Neutron-Rich Nuclide Synthesis," *Phy. Rev. Lett.*, vol. 121, p. 112701, Sep. 2018. doi:10.1103/PhysRevLett.121.112701

[9] R. Scott, R. C. Vondrasek, "ECR3 commissioning and planning for C-14 ion beams at the Argonne Tandem Linac Accelerator System," in *Proc. of the 24th Int. Workshop on ECR Ion Sources (ECRIS2020)*, East Lansing, Michigan, USA, Sep. 2020, pp. 157-159. doi:10.18429/JACoW-ECRIS2020-WEZZO03

[10] L. Callahan *et al.*, "Initial tests of Accelerator Mass Spectrometry with the Argonne Gas-Filled Analyzer and the commissioning of the MONICA detector," *Nucl. Instrum. Methods B*, vol. 532, pp. 7-12, 2022. doi:10.1016/j.nimb.2022.09.009

[11] National Ignition Facility Discovery Program, 2019 Awarded Campaigns, https://lasers.llnl.gov/nif-users/prior-year-awards/2019-awarded-campaigns

[12] R. N. Sahoo *et al.*, "The ^{41}Ar(n,γ)^{42}Ar reaction," *EPJ Web of Conferences*, vol. 284, 01037, 2023. doi:10.1051/epjconf/202328401037

[13] P. Collon, *et al.*, "Reducing potassium contamination for AMS detection of ^{39}Ar with an electron-cyclotron-resonance ion source," *Nucl. Instrum. Methods B*, vol. 283, pp. 77-83, 2012. doi:10.1016/j.nimb.2012.04.020

[14] Agilent Technologies, Inc. Santa Clara, California, USA, http://www.agilent.com

[15] Isoflex USA Ltd., San Francisco, California, USA, http://www.isoflex.com

[16] Tektronix, Inc., Beaverton, Oregon, USA, http://www.tek.com/en/products/keithley