

## EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

### Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

## Measurement of shifts in the electron affinities along isotopic chains

Dag Hanstorp<sup>1</sup>, Jakob Welander<sup>1</sup>, David Leimbach<sup>1</sup>, Annie Ringvall-Moberg<sup>1,2</sup>, Michel Godefroid<sup>3</sup>,  
Per Jönsson<sup>4</sup>, Jörgen Ekman<sup>4</sup>, Tomas Brage<sup>5</sup>, Klaus Wendt<sup>6</sup>, Reinhard Heinke<sup>6</sup>, Oliver Forstner<sup>7</sup>,  
Yuan Liu<sup>8</sup>, Sebastian Rothe<sup>2</sup>, Tim Giles<sup>2</sup>, Katerina Chrysalidis<sup>2,6</sup>, Pierre Larmonier<sup>2</sup>, Valentin Fedosseev<sup>2</sup>  
and Bruce Marsh<sup>2</sup>.

<sup>1</sup> Department of Physics, University of Gothenburg, SE 412 96 Gothenburg, Sweden

<sup>2</sup> CERN, CH-1211 Geneva 23, Switzerland

<sup>3</sup> Université libre de Bruxelles, B 1050 Brussels, Belgium

<sup>4</sup> Materials Science and Applied Mathematics, Malmö University, 205 06 Malmö, Sweden

<sup>5</sup> Division of Mathematical Physics, Department of Physics, Lund University, Box 118, SE-221 00 Lund,

<sup>6</sup> Institut für Physik, Johannes-Gutenberg Universität, Mainz, Germany

<sup>7</sup> Friedrich Schiller Universität, Jena, Germany

<sup>8</sup> Physics Division, Oak Ridge National Laboratory (ORNL), Oak Ridge, Tennessee, USA

Spokesperson: D. Hanstorp (dag.hanstorp@gu.se)

Co-spokesperson: J. Welander (jakob.welander@gu.se)

Contact persons: S. Rothe (sebastian.rothe@cern.ch)

### ABSTRACT

We propose to conduct a series of measurements of the isotope shift in the electron affinity (EA) for different elements. The specific mass shift is sensitive to electron correlation that is particularly pronounced in negative ions and neutral atoms. Hence, a study of isotopes shifts in electron affinities is an excellent method to get benchmark data for theoretical models that go beyond the independent-particle model. The treatment and interpretation of the experimental data will be supported by multiconfiguration Dirac-Hartree-Fock (MCDHF) calculations performed by leading specialists in the field of theoretical atomic physics.

In the scope of this LOI we envisage to determine the EA isotope shift between two chlorine isotopes on the neutron rich and neutron deficient side. This experiment will allow us to establish the limits of our technique in terms of our detection method and the laser system. The collinear laser photodetachment spectroscopy will be conducted using the GANDALPH experimental beam line at GLM using the RILIS laser system in narrow linewidth configuration.

**Requested shifts:** 8 shifts

## 1. INTRODUCTION AND MOTIVATION

Negative ions are unique quantum systems. The lack of a long-range Coulomb force acting on the valence electron gives them binding energies that are about an order of magnitude smaller than the ionization potentials for neutral atoms. Furthermore, the short-range binding force can only support few, if any, bound excited states and essentially all transitions within negative ions are optically forbidden. So far, electric dipole transitions in negative ions have only been observed in the cases of  $\text{Os}^-$ , [1]  $\text{Ce}^-$  [2] and  $\text{La}^-$  [3]. Hence, traditional spectroscopic methods, which are used to identify and to determine the structure of atoms and positive ions, can normally not be applied to negative ions. Nevertheless, these lightly bound systems are readily available in many environments such as the solar atmosphere [4], the interstellar medium [5] and plasmas [6]. Furthermore, the unique properties of negative ions find use in many different applications, such as accelerator-based mass spectrometry (AMS) [7] and fusion research [8].

From the fundamental point of view, the lack of a long-range Coulomb force causes the inter-electronic interaction to become relatively more important. As a consequence, the independent particle model, which adequately describes the atomic structure under normal conditions, breaks down. Experimental investigations of negative ions can therefore serve as a useful probe of electron correlation and hence be used to test theoretical models that go beyond the independent particle approximation [9].

As a consequence of the lack of bound excited states, the only atomic parameter that can be determined with high accuracy for a negative ion is the electron affinity (EA), which is defined as the energy gain by attaching an extra electron to a neutral atom. The EA of most elements that form stable negative ions [10, 11] have been determined experimentally. The most precise measurements today are made using the Laser Photodetachment Threshold (LPT) method [12] or the photodetachment microscope [13]. The first EA measurement of a radioactive isotope was conducted in 2016 at ISOLDE using the LPT method to determine the EA of radiogenic  $^{128}\text{I}$ . This measurement together with obtaining the first photodetachment signal of astatine was a mile-stone towards the measurement the EA of astatine, one of the few elements that form negative ions where the EA is unknown.

We now propose to extend the electron affinity program at ISOLDE to study the isotope shift in the electron affinity of isotopic chains of other elements. The isotope shift between the stable isotopes  $^{35}\text{Cl}^-$  and  $^{37}\text{Cl}^-$  was the first isotope shift in a negative ion to be investigated both experimentally and theoretically by Berzinsh *et al.* [12] in 1995. In their work a discrepancy between the experimental and theoretical result was observed. In 2013, this discrepancy was solved through state-of-the-art *ab initio* calculations by Carette and Godefroid [14] using non-relativistic many-body theory. It will be difficult to simply transpose these correlation models from the non-relativistic to the full relativistic multi-configuration schemes, but there is some hope to take advantage of the partitioned correlation function interaction approach [15]. In this context, an extended study of a chain the isotopes of chlorine, with a corresponding larger isotope shift would be very valuable. The isotope shift observed by Berzinsh *et al.* [12] was less than a tenth of the

laser bandwidth, and the hyperfine structure in the odd isotopes investigated partly obscured the observation of the isotope shift.

Within the scope of this letter of intent, we envisage to test and refine the LPT technique by studying selected radioactive chlorine isotopes. Of special interest will be the even isotopes where the lack of hyperfine structures will give sharper thresholds. Experimentally, chlorine is a favorable choice: Firstly, it is prolifically produced directly as negative ions in a surface ionizing process, and secondly the RILIS laser system can provide light with a bandwidth that is one third of that in used in the experiment of Berzinsh et al.[12].

## **2. ISOTOPE SHIFT THEORY**

The isotope shift is the small difference that is observed in the line frequencies of spectral lines appearing for different isotopes of the same element. This shift is composed of a volume shift and a mass shift component.

The volume shift is caused by the difference in the nuclear charge distributions defining the field in which the electrons move. This effect has been extensively used to extract and study the change of the effective nuclear charge radii, from which useful information relevant in nuclear physics can be deduced, such as nuclear deformations or subshells closure signatures [16].

The mass shift, on the other hand, is caused by the finite mass of the nucleus. In a one-electron system the electron will not circulate around the nucleus, but instead the nucleus and the electron will move around their center-of-mass. The size of this effect can easily be calculated by replacing the electron mass in the Schrödinger equation by its reduced mass. This is called the normal mass shift. In many-electron systems the situation becomes much more complicated. If the electron motion is correlated, the mass shift can either be enhanced or reduced. The deviation from what would be expected from the normal mass shift is called the specific mass shift. The latter is a pure atomic effect caused by a correlated motion of the electrons. Therefore, negative ions, where the electron correlation is particularly pronounced, are exceptionally suitable systems to perform investigations of the specific mass shift.

The mass shift dominates for lighter masses, whereas the volume shift is the largest for heavier masses. In order to determine the volume shift, which can be used to extract the mean square charge radii of nuclei, the mass shift is required to be determined and subtracted from the total isotope shift. Hence, a detailed understanding of the specific mass shift is of importance directly in atomic physics, as a means of investigating the electron correlation effect, and indirectly in nuclear physics, where it is required in order to extract an accurate value of the volume shift, and hence the determine nuclear charge radii, from an isotope shift measurement.

The isotope shift in the electron affinity is defined as the difference in the energy gain when adding an electron to the two different isotopes under investigation. Hence, this property will be determined by the structure of the ground states of both the negative ion and the neutral atom. An examination of the literature shows that the electron affinities of atoms, with the exception of the hydrogen atom, have been experimentally determined with higher precision than the calculated

values. However, the calculated value of the IS of chlorine by Carette and Godefroid [14] is more precise than the observed value by Berzinsh et al. [12]. Consequently, a precise determination of the isotope shift in the EA of chlorine would be very valuable to test the validity of the many body particle calculations performed by Carette and Godefroid.

To date, the isotope shift in the electron affinity has been experimentally investigated in hydrogen [17], carbon [18], oxygen [19, 20], sulphur [21], chlorine [12] and lead [22]. Here only two, or in the case of oxygen three, stable isotopes were investigated. On the theoretical side, this property has been estimated for several lighter elements by Godefroid and collaborators using multiconfiguration Dirac-Hartree-Fock (MCDHF) methods and elaborate correlation models [14, 20,23-25].

The theory of isotope shift can be found in Bauche and Champeau's seminal review [26] and in King's textbook [27]. Reformulations of the mass and field shifts have been proposed in 1987 [28, 29]. In the relativistic framework, Shabaev [30] derived the relativistic corrections to the recoil operator. These corrections can be estimated using the codes RIS3 [31] and RIS4 [32] and relativistic atomic wave functions calculated with Grasp2Kdev [33]. With RIS4, nuclear charge deformation can be included with a reformulation of the field shift. Most of the theoretical works on IS in EAs mentioned above [14, 20, 23-25] were performed in the non-relativistic framework using ATSP2k [34]. Computational atomic structure definitely moves to the fully relativistic approach in which non-relativistic methodologies are integrated [35]. Robust relativistic isotope shift calculations can be done [36, 37] using large multireference multiconfiguration expansions, with unavoidable limitations in the inclusion of electron correlation for neutral atoms and negative ions.

### 3. EXPERIMENT

#### 3.1. The GANDALPH experimental beam line

The laser photodetachment threshold (LPT) method [12] is a standard technique for precision determination of electron affinities. The photodetachment process on the negative ion represents a transition from the ground-state of the negative ion to the ground-state of the neutral atom, thereby releasing a photoelectron. For such an LPT measurement, a beam of negative ions is overlapped over a preferably large interaction length with a laser beam, e.g. in a collinear geometry. Furthermore, the collinear geometry has the advantage that the resolution in the experiment is improved by the so called Doppler compression [39]. Neutral atoms generated in the detachment process are detected while the laser frequency is scanned across the threshold region. If the photon energy exceeds the electron affinity, the photodetachment cross-section increases dramatically, as described by the Wigner-law [40]. Very precise values of the EA can be extracted from such a threshold measurement for all elements where the outgoing electron is emitted as an s-wave. This is the case for elements where the valence electron in the negative ion is in a p-orbital: the B, C, N, O and F groups in the periodic table. The relative photodetachment cross-section is measured as the neutralization rate. We have successfully demonstrated the feasibility of the laser

photodetachment technique under ISOLDE operating conditions first with a stable negative iodine and later with a radioactive ( $^{128}\text{I}$ ) beam. A schematic layout of the setup is shown in Figure 1.

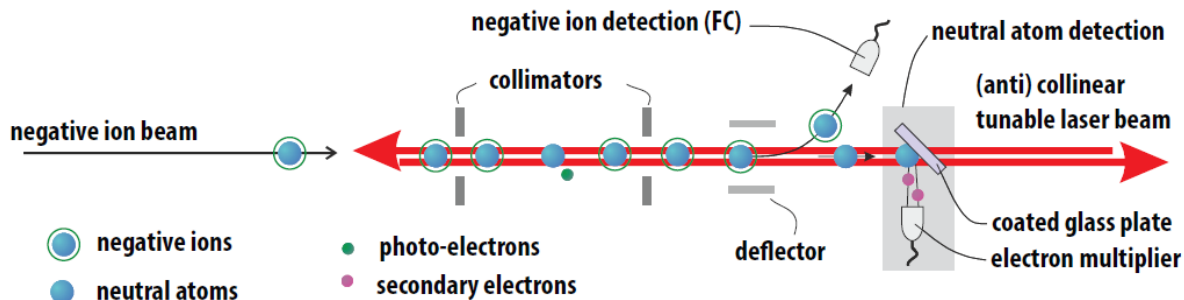


Figure 1: Schematic layout of the experimental setup. Negative ions, extracted from an ISOLDE target and ion source, are overlapped (anti)collinearly with the detachment laser beam. Undetached ions are deflected to a Faraday cup. Residual atoms from the photodetachment process impinge on a coated glass substrate and create secondary electrons which are detected by an electron multiplier.

Within the frame of our astatine campaign we have constructed a transportable collinear laser spectroscopy setup, designed to be mounted at the GLM beam line. The GANDALPH (Gothenburg ANion Detector for Affinity measurements by Laser PHotodetachment) beamline is based on the design of the negative ion beam apparatus GUNILLA (Gothenburg University Negative Ion Laser Laboratory) [41] which has been successfully used to measure the EAs of several elements (e.g. tungsten, [42]). GANDALPH consists of the following components

- differential pumping section
- laser ion merging section
- laser/ion interaction region
- neutral detector
- tunable laser system
- data acquisition

The differential pumping system gives a background lower than  $10^{-9}$  mbar in the laser/ion interaction region while the apparatus is connected to the GLM beamline at  $10^{-6}$  mbar. Laser/ion overlap is achieved by a 10 degree electrostatic bend. The laser/ion interaction region is defined by two collimators. The neutralized fraction of the beam is detected in the neutral detector where secondary electrons are generated upon impact of the fast atomic fraction onto the detector plate that are then extracted and detected by a single channel electron detector. The collection of the secondary electrons can be gated in order to prevent saturation of the detector [24]. The photo-

electrons generated when the laser pulse strikes the detector plate are suppressed by a fast-switching detector plate. The laser system is part of the ISOLDE RILIS installation.

During the experimental campaigns for our LOI-I148 we have encountered features of our experimental setup that, in conjunction with beam steering issues of the GPS magnet, limited the experimental capabilities through contamination of our neutral detector. These issues will be addressed in the next version of the detector. One objective of the LOI is to ensure the correct functioning of the detector at low negative ion beam intensities.

### **3.2. RILIS laser requirements**

The threshold for photodetachment of chlorine is equivalent to a photon energy at a wavelength of 343 nm. Laser light with this wavelength is available at RILIS using Nd:YAG pumped Pyridine 1 dye with frequency-doubling. The experiments require that the laser is operated with minimum bandwidth using the thick intra-cavity etalon, giving a bandwidth of about 1.6 GHz. The optical beam path from RILIS to GLM has already been established and allows safe transport of the class 4 laser beams to GANDALPH.

### **3.3. Data Acquisition**

The data acquisition system is built upon the modular and well established RILIS DAQ commonly used for in-source laser spectroscopy that has been extended to integrate a time gated photon counter in a previous campaign.

The signal from the residual atoms of the photodetachment process will be obtained by integrating the events from the electron multiplier within a time window corresponding to the time of flight of the atoms. The background signal will be obtained simultaneously from the integral of the events in a delayed time window of the same duration. The ion beam intensity on the detector Faraday cup will be recorded for normalization.

### **3.4. Objective**

The scope for this LOI is to demonstrate the first EA isotope shift measurement on a radio isotope chain while providing information about the limits of the GANDALPH setup in terms of background generated by the decay of implanted beam onto the detector. We will also attempt the first anti-collinear detachment which is required for beam energy calibration.

#### 4. SHIFT REQUIREMENTS

Yield information on chlorine isotopes produced from an MK4 negative ion source and a Th/Ta target were distributed in the ISOLDE Users meeting 2016 (Presentation by M.Delonca). It is to be noted that the target thickness was reduced by a factor of 2 in the 2016 experiments for I-148.

Isotope	Half-life	Yield / Production rate [ ions/s/ $\mu\text{A } ^1\text{H}^+$ ]	
		<b>2005</b>	<b>2016</b>
<b><math>^{38}\text{Cl}</math></b>	37.2 m	$1.6 \cdot 10^5$	$1 \cdot 10^5$
<b><math>^{40}\text{Cl}</math></b>	81 s	$4.3 \cdot 10^4$	$9 \cdot 10^4$
<b><math>^{41}\text{Cl}</math></b>	38.4 s	$1.4 \cdot 10^4$	$3.5 \cdot 10^2$
<b><math>^{42}\text{Cl}</math></b>	6.9 s	$1.1 \cdot 10^3$	$1.4 \cdot 10^1$

The database yield for  $^{38}\text{Cl}$  using a Nb foil target lists  $1.5 \times 10^7$  ions/ $\mu\text{C}$ . Because of the spread of yields and depending which target material and ion source configuration is available for the experiment, we would request 2 shift for yield tests and optimization using simple direct ion detection in our beam line. Our approach will be to select two isotopes (neutron rich and neutron deficient) that are available with quantities in the order of  $2 \times 10^4$  ions/s in our beam line. Based on experience from  $^{128}\text{I}$  during the I-148 campaign, this ion rate is sufficient to perform a single scan within 3h (this includes optimizing the separator tune, our detector and the RILIS lasers and actual scanning).

We will attempt the following number of scans: 2 isotopes, 2 laser scan directions, 2 laser beam geometries (collinear and anti-collinear), with all scans performed 2 times (i.e 1 repetition) to determine effects on the detector. This sums up to 16 laser scans requiring 48h or minimum 6 shifts.

We request a total number of 8 shifts preceded by 4 shifts for the off-line setup and 1 shift for reference scans after the on-line shifts.

<b>Description</b>	<b>Element</b>	<b>Number of shifts (offline)</b>	<b>Protons</b>
Stable beam tuning	stable chlorine	(4)	no
Yield measurements	chlorine isotopes	2	yes
Threshold determinations	chlorine isotopes	6	yes
Reference stable isotope scans	stable chlorine	(1)	no

## 5. CONCLUSION

In this letter of intent we propose an experimental investigation of the isotope shifts in chlorine isotopes. Chlorine was chosen since this system has been treated theoretically, and since it is experimentally favorable.

We foresee that these first investigations will be followed by a full proposal for a sequence of isotopes of chlorine as well as studies of isotope shifts in electron affinities of other elements. The isoelectronic fluorine isotope would be very interesting since the smaller number of electrons make a more accurate theoretical treatment feasible. A systematic study along the halogen group will allow us to investigate the transition from light systems where the mass shift dominates to the heavy elements where the volume shift is the largest contributor to the isotope shift. Other interesting candidates for further study using this method are carbon, silicon, and oxygen. The relatively few electrons make them tractable for a theoretical treatment, at the same time as their electronic structure will, according to the Wigner law, give a relatively sharp threshold which in turn improves the precision of the measurements.

Experimentally, some of these elements will require production of doubly charge exchange of positive ion beams. For this GANDALPH can be eventually connected to the CRIS beam line.

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## 7. APPENDIX

### 7.1. DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises:

Part of the Choose an item.	Availability	Design and manufacturing
RILIS	<input checked="" type="checkbox"/> Existing	<input checked="" type="checkbox"/> To be used in non standard operation. Requires narrow bandwidth operation of dye lasers
GANDALPH	<input checked="" type="checkbox"/> Existing	<input type="checkbox"/> To be used without any modification <input checked="" type="checkbox"/> To be modified

### 7.2. HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

<i>Hazards</i>			
	<i>RILIS</i>	<i>GANDALPH</i>	<i>[Part 3 of the experiment/equipment]</i>
<b>Thermodynamic and fluidic</b>			
Pressure	[pressure][Bar], [volume][l]	10 <sup>-9</sup> mbar	
Vacuum			
Temperature	[temperature] [K]		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	[fluid], [pressure][Bar], [volume][l]		
<b>Electrical and electromagnetic</b>			
Electricity	[voltage] [V], [current][A]		
Static electricity		5000 V	
Magnetic field	[magnetic field] [T]		
Batteries	<input type="checkbox"/>		
Capacitors	<input type="checkbox"/>		
<b>Ionizing radiation</b>			
Target material	[material]		
Beam particle type (e, p, ions, etc)			
Beam intensity			
Beam energy			

Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:	<input type="checkbox"/>		
• Open source	<input type="checkbox"/>		
• Sealed source	<input type="checkbox"/> [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			
• Description	<input type="checkbox"/>		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
<b>Non-ionizing radiation</b>			
Laser	Class 4, 330-1000 nm	Class 4, 330-1000 nm	
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300MHz)			
<b>Chemical</b>			
Toxic	[chemical agent], [quantity]		
Harmful	[chemical agent], [quantity]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chemical agent], [quantity]		
Corrosive	[chemical agent], [quantity]		
Irritant	[chemical agent], [quantity]		
Flammable	[chemical agent], [quantity]		
Oxidizing	[chemical agent], [quantity]		
Explosiveness	[chemical agent], [quantity]		
Asphyxiant	[chemical agent], [quantity]		
Dangerous for the environment	[chemical agent], [quantity]		

Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): 5 kW