EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of shifts in the electron affinities of chlorine isotopes

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ABSTRACT
We propose to conduct measurements of the isotope shift in the electron affinity (EA) for chlorine isotopes. The specific mass shift is sensitive to electron correlation that is particularly pronounced in negative ions and neutral atoms. Hence, a study of isotope shifts in electron affinities is an excellent method to obtain 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. The collinear laser photodetachement spectroscopy will be conducted using the GANDALPH experimental beam line at GLM using the RILIS laser system in narrow linewidth configuration.

This proposal is based on the letter of intent I-177

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. 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.

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 [1].

The only atomic parameter that can be determined with high accuracy for a negative ion is the electron affinity (EA) [2,3]. The most precise measurements today are made using the Laser Photodetachment Threshold (LPT) method [4] or the photodetachment microscope [5]. 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}I [6]. This measurement together with obtaining the first photodetachment signal of astatine was a mile-stone towards measuring 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}Cl$^-$ and ^{37}Cl$^-$ was the first isotope shift in a negative ion to be investigated both experimentally and theoretically by Berzinsh *et al.* [4]. 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 [7] using non-relativistic many-body theory. It will be difficult to 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 [8]. In this context, a study of chlorine isotopes with a large mass difference hence a larger isotope shift would be very valuable.

Within the scope of this proposal, we envisage to measure the isotope shift in the EA between two even isotopes where the lack of hyperfine structures will give sharper thresholds.

2. ISOTOPE SHIFT THEORY

The isotope 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 [9]. 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. 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 the size of the mass shift need to be known, so it can be subtracted from the total isotope shift. Hence, a detailed understanding of the specific mass shift is of importance both directly in atomic physics and indirectly in nuclear physics when the charge radii is determined from the volume shift.

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 isotope shift of chlorine by Carette and Godefroid [7] is more precise than the observed value by Berzinsh *et al.* [4]. 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 [7].

To date, the isotope shifts in the electron affinity have been experimentally investigated in H [10], C [11], O [12, 13], S [14], Cl [4] and Pb[15]. 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 [7, 13, 16-18].

The theory of isotope shift can be found in Bauche and Champeau's seminal review [19] and in King's textbook [20]. Reformulations of the mass and field shifts have been proposed in 1987 [21,22]. In the relativistic framework, Shabaev [23] derived the relativistic corrections to the recoil operator. These corrections can be estimated using the codes RIS3 [24] and RIS4 [25] and relativistic atomic wave functions calculated with Grasp2Kdev [26]. With RIS4, nuclear charge deformation can be included with a reformulation of the field shift. Most of the works on IS in EAs mentioned above [7, 13, 16-18] were performed in the non-relativistic framework using ATSP2k [27]. Computational atomic structure definitely moves to the fully relativistic approach in which non-relativistic methodologies are integrated [28]. Robust relativistic isotope shift calculations can be done [29, 30] using large multi-reference multiconfiguration expansions, with unavoidable limitations in the inclusion of electron correlation for neutral atoms and negative ions.

3. EXPERIMENT

Within the frame of our astatine campaign we have constructed a transportable beam line named GANDALPH (Gothenburg ANion Detector for Affinity measurements by Laser Photo-detachment). A beam of negative ions is merged with a laser beam over 50 cm (Fig. 1). Neutral

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atoms generated by the absorption of a photon 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 [31].

The neutral atoms are detected by allowing them to impinge on a glass plate where secondary electrons are generated which are detected with a channel electron detector. The photo-electrons generated when the laser pulse strikes the detector plate are suppressed by a fast-switching detector plate. 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. We have successfully demonstrated the feasibility of the system by determining the photodetachment threshold energy for the radioactive ^{128}I isotope [6].
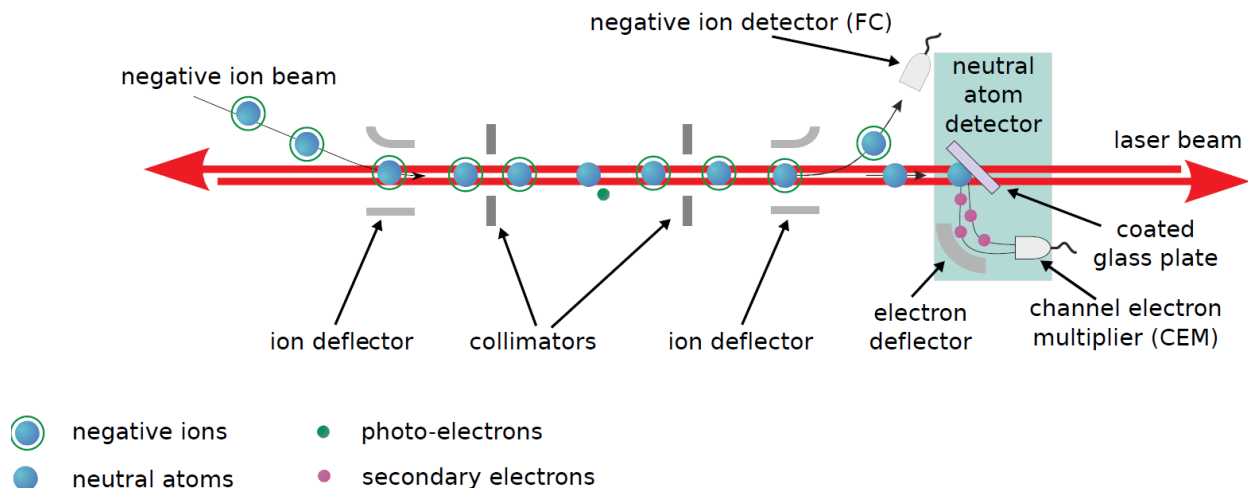

Figure 1: Schematic layout of GANDALPH. Negative ions, extracted from an ISOLDE target and ion source, and merged with a laser beam. Residual atoms from the photodetachment process impinge on a coated glass substrate and create secondary electrons which are detected.

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.

The data acquisition system is built upon the modular and well established RILIS DAQ commonly used for in-source laser spectroscopy. 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 ion beam intensity on the detector Faraday cup will be recorded for normalization.

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/µA $^1H^+$]	
		2005	*2016*
^{38}Cl	37.2 m	$1.6\ 10^5$	$1\ 10^5$
^{40}Cl	81 s	$4.3\ 10^4$	$9\ 10^4$
^{41}Cl	38.4 s	$1.4\ 10^4$	$3.5\ 10^2$
^{42}Cl	6.9 s	$1.1\ 10^3$	$1.4\ 10^1$

The database yield for ^{38}Cl using a Nb foil target lists 1.5×10^7 ions/µC. Because of the spread of yields and depending which target material and ion source configuration is available for the experiment, we would request 2 shifts 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 intensities in the order of 2×10^4 ions/s in our beam line. Based on experience from ^{128}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 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.

Ideally, the compatibility of the laser wavelength, laser line width and RILIS scanning capabilities to our requirements would be confirmed in a separate run earlier to the on-line experiment. For this we would request 3 shifts of off-line beam time with stable chlorine isotopes.

We therefore request a total number of 8 shifts with radioactive beam, 8 shifts for the compatibility test, off-line setup and reference scans split into two runs.

Description	Element	Number of shifts (offline)	Protons
Run 1			
Compatibility tests	stable chlorine	(3)	no
Run 2			
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

4. CONCLUSION

We propose an experimental investigation of the isotope shifts between two chlorine isotopes. We foresee that these first investigations will be followed by an addendum for a sequence of chlorine isotopes as well as further proposals to study isotope shifts in electron affinities of other elements. For this, GANDALPH can be eventually connected to the CRIS beam line which enables to extend the list of available elements through the use of double charge exchange.

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. The isoelectronic fluorine isotope would be very interesting since the smaller number of electrons make a more accurate theoretical treatment feasible. Other interesting candidates for further studies 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.

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6. APPENDIX

6.1. DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises:

Part of the Choose an item.	Availability	Design and manufacturing
RILIS	☒ Existing	☒ To be used in non standard operation. Requires narrow bandwidth operation of dye lasers
GANDALPH	☒ Existing	☐ To be used without any modification ☒ To be modified

6.2. 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:

Hazards	*RILIS*	*GANDALPH*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Vacuum		10^{-9} mbar	
Electrical and electromagnetic			
Static electricity		5000 V	
Ionizing radiation			
Beam particle type		negative chlorine ions	
Beam energy		10-20 keV	
Cooling liquids		water, closed loop	
Non-ionizing radiation			
● Laser	Class 4, 330-1000 nm	Class 4, 330-1000 nm	

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