EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Preparation of negative ion beams for the determination of the electron affinity of polonium by laser photodetachment

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Abstract

Polonium, element 84, is a radioactive element of which many fundamental properties still remain unknown. One example is the electron affinity, the energy gained when attaching an electron to a neutral atom. The electron affinity determines the chemical reactivity and, indirectly, the compounds the element can form as well as the stability of its chemical bonds. From a theoretical standpoint, electron affinity investigations are of interest because they can serve as a precise probe for electron correlation. Polonium, a relatively heavy element, is of particular interest due to the prevalence of relativistic effects in its atomic structure as well as its two-hole electron configuration, rendering it more complex to study than the one electron hole system of astatine.
In a previous letter of intent (INTC-I-148) and proposal (INTC-P-462), an experiment was outlined to measure the electron affinities of both polonium and astatine. The electron affinity of astatine was successfully determined in a beam time in 2018, and now steps are proposed to determine the electron affinity of polonium. A technical obstacle here is that polonium cannot be produced efficiently in the negative surface ion source used in the astatine experiment. However, positively charged polonium beams are readily available and have been delivered reliably from actinide targets. Therefore, we propose to investigate negative ion beam production of polonium by means of charge exchange at the CRIS experiment. The feasibility of the method to determine electron affinities by attaching the GANDALPH detector to CRIS will be demonstrated off-line by measuring the electron affinity of stable and isoelectronic elements to polonium.

Requested shifts: 6 shifts, (split into 2 runs over 1-2 days)

1 INTRODUCTION

Polonium (Po) is the first even-Z element in the periodic table for which all of the isotopes are radioactive, and most are short-lived. Polonium-210 ($t_{1/2}$=138.4 d) naturally occurs in uranium ores as a product of the uranium-238 decay chain, making it the most common isotope of Po. As an alpha emitter (E_α=5.29 MeV) with a high specific activity (166 TBq/g), ^{210}Po is highly toxic and potentially fatal when ingested[1]. Although Marie and Pierre Curie discovered Po more than a century ago, many of its characteristics remain elusive. Further investigations of Po radiotoxicity can help develop efficient decorporation treatments in medicine and long-term management plans for abandoned uranium mines[2].

We can learn more about the behavior of Po through studying its negative ion. Negative ions have both practical and fundamental applications in medicine[3,4], chemical reactivity[4], astrophysics[5], and they can serve as benchmarks for atomic structure calculations[6]. At ISOLDE, the development of negative ion beams would also benefit future experiments like PUMA[7], which aims to investigate the density distribution of neutrons and protons on the surface of the nucleus of exotic ions using antiprotons.

The ionization potential (IP) of Po was recently measured with high precision at TRIUMF-ISAC[8] and ISOLDE[9]. The electron affinity (EA), on the other hand, remains experimentally unknown. Following the EA measurement of astatine (At) at ISOLDE[4], the next natural progression is Po. Polonium is the lighter neighbor of At, the heavier homolog of tellurium (Te), and is expected to have a smaller EA than both. Theory predicts that the EA will be positive and that it could form stable negative ions[10].

The laser photodetachment threshold (LPT) method[11] is the standard technique for precision determination of EAs. The LPT method is routinely applied for stable isotopes of elements such as Te[12], and has been successfully demonstrated at ISOLDE for radioactive At[4] and ^{128}I[13]. This is the method that will also be used to eventually measure the EA of Po.

High production yields of Po isotopes have been demonstrated at ISOLDE by using actinide targets[9,14]. Beam contamination was previously addressed by pre-irradiating the target and then stopping protons to allow the main contaminants to decay[9]. However, this should not be a problem for us as we plan to use ^{202}Po which has shorter lived contaminants. Additionally, this measurement campaign could run in conjunction with any other approved campaign to study Po, such as the experiment to investigate its ground state properties by in-source spectroscopy[15].

A variety of laser systems and laser/ion beam interaction lines are available at the ISOLDE facility, making it a most suitable place for these studies. Our goal is to measure the EA of Po by use of an approach similar to the one used for the corresponding study of the At measurement at ISOLDE[4]. In the At study, the Gothenburg ANion Detector for Affinity measurements by Laser PHotodetachment (GANDALPH) was attached to the GLM branch at ISOLDE. We now intend to place GANDALPH at the end of the CRIS beamline, where negative ions can be produced through a double charge exchange reaction of Po^+. Beforehand, we will demonstrate the feasibility of the laser photodetachment method off-line at CRIS with a stable, isoelectronic element such as Te.

The accompanying theoretical calculations of the EA of Po will be carried out using the state-of-the-art relativistic coupled cluster approach, which is considered the most accurate method for studies of heavy many-electron atoms. Higher order electronic excitations and quantum electrodynamic (QED) corrections will be included in these calculations, allowing for meV accuracy[16]. The newly developed approach will then be used to estimate uncertainties on the theoretical predictions, to guide the experimental wavelength scan range, and facilitate comparison[4]. Furthermore, parallel calculations of the EA of Te will be made and compared with the experimental value. This should provide an excellent model system to test the precision in the many-body calculation where both QED and relativistic effects need to be taken into account.

Within this letter of intent, we request beam time for determining the yield of Po^- by means of charge exchange as a milestone towards the determination of the EA of Po.

2 TECHNIQUE AND EXPERIMENTAL SETUP AT ISOLDE

2.1 Laser photodetachment in collinear geometry

The EA of an element can be determined from the threshold photon energy required to induce a photodetachment process on the negative ion. This represents a transition from the ground state of the negative ion to the ground state of the neutral atom releasing an electron[11]. For a LPT measurement, a beam of negative ions is overlapped with a laser beam in a collinear geometry. The 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 EA, the photodetachment cross-section increases dramatically, following the Wigner law[17]. Hence, a precise value for the EA can be extracted from the energy of the onset in the photodetachment

cross-section. The relative detachment cross-section is measured as the neutralization rate by detecting secondary electrons released after created neutral atoms impinge on a graphene coated glass plate[18]. A sketch of the experimental layout of the spectrometer, GANDALPH[4,13,19], is shown in Figure 1. Figure 2 shows a schematic diagram of the CRIS beamline in its current functionality and future placement of GANDALPH. Minor changes to the CRIS setup will need to be made in order to study the Po negative ions including the wavelength of the lasers and the polarity of the electric fields after the charge exchange cell. A typical photodetachment spectrum, as obtained for Te[12], is shown in Figure 3. A similar result is expected for the Po spectrum.

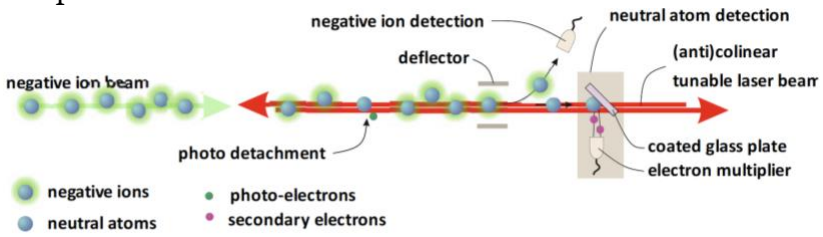

Figure 1 – Collinear laser photodetachment spectroscopy within GANDALPH. The frequency tunable laser beam is overlapped with the negative ion beam. The photodetachment takes place in the overlap region. Non-detached ions are deflected, and the photo-neutrals are detected via secondary electron generation in a graphene coated glass plate.

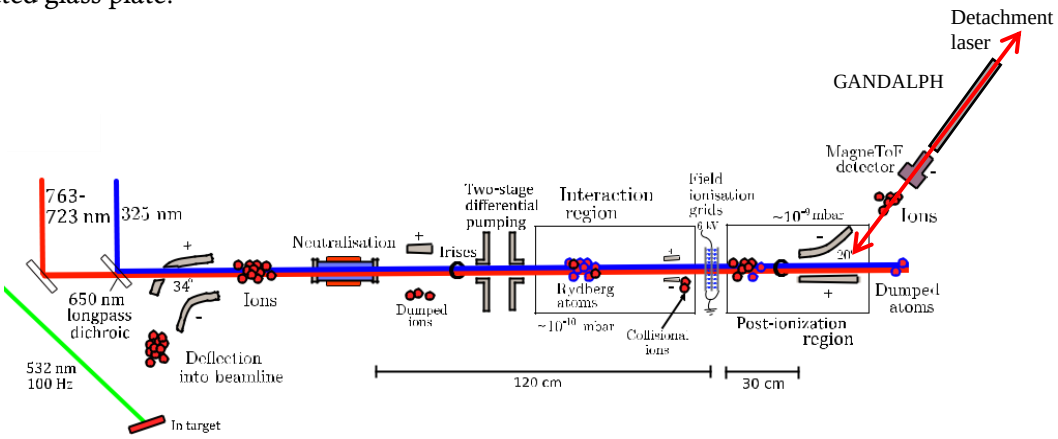

Figure 2 – Schematic diagram of the current CRIS beamline and plans for GANDALPH placement and detachment laser.

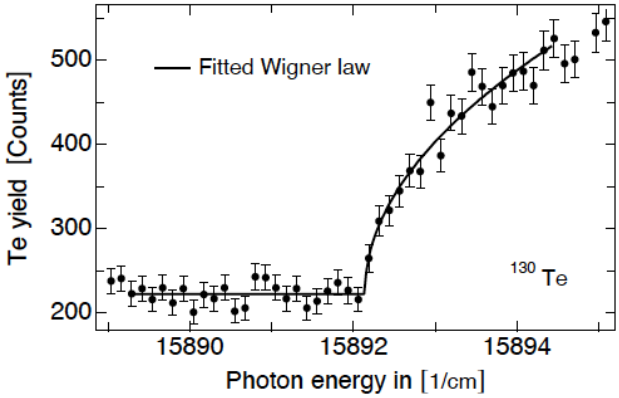

Figure 3 – Measurement of the total photodetachment cross section for Te. The solid line is a fit of the Wigner law to the experimental data[12].

2.2 Future beam and laser requirements

From the experiments for the determination of the EA of At[4] and ^{128}I[13], we can estimate the average ion beam current required for a future experiment on Po. An average ion beam current of $^{211}At^-$ was measured before the experimental chamber to be about 600 fA with an estimated transmission of about 1%[4]. For the radioactive ^{128}I measurement, the beam of stable ^{127}I was used for tuning where the average current was measured to be around 200 pA with a maximum transmission efficiency of 25%[13]. These two experiments both used the GPS separator and were therefore limited by the duty cycle losses of the pulsed laser, with a loss factor as great as 10 000.

The pulsed laser loss factor will not be a concern at CRIS because the ISCOOL RFQ cooler buncher ensures temporal overlap between the laser and ion beams[20]. Even with losses due to the efficiency of the double charge exchange process, a large improvement in sensitivity can be expected using ISCOOL compared to GLM. Oxygen is the only homologue of Po for which the double charge exchange cross sections have been investigated. The expected efficiencies of negative oxygen ion formation in sodium (Na) vapour, using beam energies of 20-30 keV, were reported to be 32-25%[21]. This process will be further tested off-line with Te prior to studying Po.

The laser wavelength scanning region of interest is defined as 830-890 nm from extrapolation of theoretical calculations[10,22,23]. These values as well as experimental values of Te[12], Bi[24], and At[4] are shown in Table 1. We plan to complete the high precision calculations prior to the experiment, which will allow us to narrow the wavelength range to 10 nm for the investigation of the threshold energy.

This preparatory work sets the scan region for laser photodetachment within well-defined and narrow limits, for which the required laser wavelengths can be obtained easily using Ti:Sapphire lasers that are available at ISOLDE.

Table 1

Element (Method)	*EA (eV)*	*Reference*
Bi (experimental)	0.942362(13)	24
Te (experimental)	1.970876(7)	12
Po (MCDHF)	1.405	23
Po (DC-CCSD(T)+ Breit +QED)	1.469	22
Po (CBS-DC-CCSD(T)+Gaunt+QED)	1.484(26)	10
At (experimental)	2.41578(7)	4

3 INVESTIGATION OF THE PRODUCTION OF NEGATIVE ION BEAMS

The crucial part of the experiment is the efficient production and transport of negative ions to the laser interaction region. While the EA measurement of At was successful with negative ion production using low work function surface ionizers[4], this will not be feasible for Po. The EA of Po is expected to be smaller than the work function of the LaB_6 surface ionizer, resulting in a drastic decrease of the ionization efficiency. Furthermore, there is no data on the production of Po negative ion beams at ISOLDE yet. For these reasons, we propose to investigate the use of a charge exchange cell to convert positive Po ions to negative ions.

3.1 Conversion of positive ions to negative ions by charge exchange in a metal vapour cell

The CRIS experimental setup is equipped with a charge exchange cell, allowing for the neutralization of positively charged ion beams by means of interaction with a vapour of alkaline elements (Na or K in the case of CRIS)[25,26]. It is also possible to produce negative ions in two separate electron capture processes.

The main variables when investigating the production rate of negative ions are the density of the alkali vapour, determined by the temperature, and the incoming beam energy. Figure 4 shows the dependence of negative equilibrium charge state fractions on energy of projectile elements in Na vapour[21]. Oxygen has a similar EA as the predicted value for Po and could be used to compare negative equilibrium charge state fractions.

The charge exchange cell installed at CRIS is already producing negative ions. As no experiments at CRIS currently use negative ions, these are being deflected away and dumped. The beam of negative ions can be studied by reversing the polarity of the electric field that separates the charge states in the beamline after the charge exchange cell.

A measurement of the charge exchange efficiency, X^+ to X^-, and the equilibrium ratios can be performed for a homologous element of Po. Finally, the charge exchange efficiency for Po has to be determined. The experimental configuration foreseen for this is shown in Figure 5. A beam of positive ions is sent through a charge exchange cell. An electrostatic deflector separates the different charge states (+1, 0, -1) and different particle detectors measure the ratios of ions to atoms and thus the conversion efficiencies.

All the necessary tests can be performed off-line with Te to then determine the yield of Po^- in the shifts requested in this proposal. Afterwards, beam time will be requested in a separate proposal.

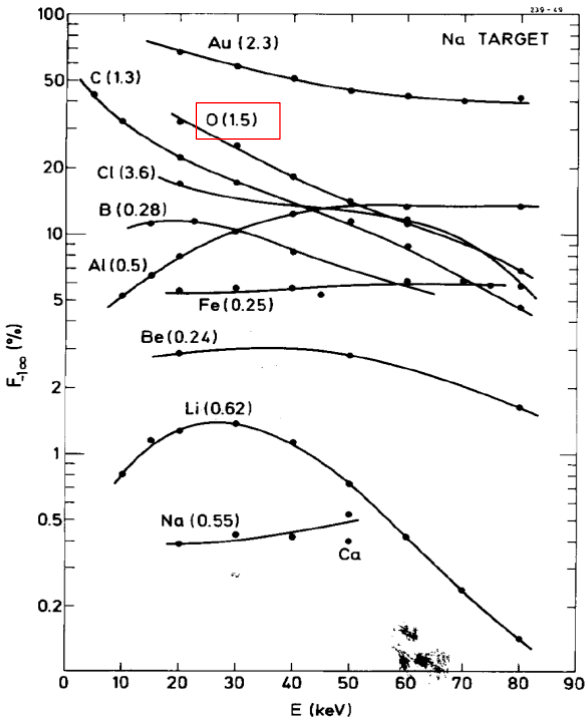

Figure 4 – The graph shows the negative equilibrium charge state fraction (%) vs. projectile energy (keV) in a sodium vapour. The element of interest here is O, where the optimal projectile energy is $\leq$20 keV, yielding a $\geq$32% negative fraction[21].

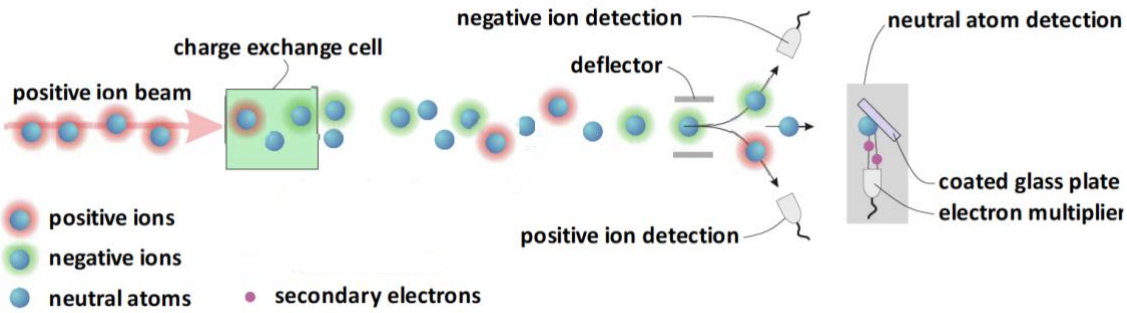

Figure 5 – Setup for the measurement of the conversion efficiency of the charge exchange cell from the negative/positive ion detection ratios. The neutral atom rate can be measured at the same time.

Summary of requested shifts

First, we plan to test the production of negative ions off-line at CRIS by reversing the polarity of the electric field using any element. Then we can begin off-line tests using Te to investigate the charge exchange efficiency. After, we will move to beam tuning where we will require an ISOLDE beam tune setup for 20, 30, and 40 keV. This can be achieved using ^{238}U (no protons/RILIS required).

For the investigation of charge exchange efficiency of Po^+ to Po^-, we request 6 shifts spilt into 2 runs of 3 shifts, separated by at least 1-2 days. For each run, the following series of tests will be done with a different alkali vapour (Na and K):

- Optimize the vapour thickness through measuring charge state production at different charge exchange cell temperatures. Each temperature increment will require 30 minutes to 1 hour to stabilize before each measurement.
- Investigate beam energy dependence of the charge exchange and transport efficiencies with the preset beam tunes at 20, 30, and 40 keV.
- Measure the positive and negative ion beams by changing polarity of electric fields.

The time separation between runs will allow the charge exchange cell to be safely cooled (temperature & short-lived radioactivity), vented, and cleaned before the other alkali metal is installed. The chamber will then be pumped down and heated for the next series of measurements.

We plan to use ^{202}Po as the main contaminants (^{202}Ra/^{202}Fr) are relatively short lived ($t_{1/2}$≈2ms/300ms) and are produced at slower rates compared to other Po isotopes (<1% ^{202}Po). Therefore, we would not have to stop the protons in order for the contaminants to decay to perform our measurements. Moreover, the decay chain of ^{202}Po is free from long-lived contaminants and present therefore a reduced radiological risk for accessing the beam line. From table 2, we can expect an ion yield of ~10^7/μC which should allow for sufficient beam intensity measurements before and after the experimental chamber with Faraday cups or with the CRIS MagneTOF ion detector. In the case that beam intensity is not sufficient to use the methods mentioned above, we could measure by single ion counting.

Table 2 – Summary of beam requirements

Beam	*Intensity*	*Target*	*Ion Source*	*Shifts*
^{202}Po	1.7 x 10^7 [ref. 14]	UC_X	RILIS	6

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: CRIS (fixed), GANDALPH (semi-fixed)

Part	Availability	Design and manufacturing
CRIS	☒ Existing	☐ To be used without any modification
Charge exchange cell	☒ Existing	☐ To be used without any modification ☒ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
Collinear laser beam line	☒ Existing	☐ To be used without any modification ☒ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
GANDALPH	☒ Existing	☒ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed CRIS installation.
Additional hazards: None