Study of the ISOLTRAP Laser Ablation Source

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An investigation into the principles of a laser ion source (LIS) and its optimized implementation in the ISOLTRAP setup will be provided. The current status of the LIS will be described and improvements will be discussed. A new configuration will be introduced along with ion transport simulations, and an outline will be made for future improvements and upgrades along with corresponding simulations.

I. INTRODUCTION & MOTIVATION

The mass of an atom can be expressed as:

\[ m = Zm_p + Nm_n + Zm_e - \frac{E_{B\text{nuc}}}{e^2} - \frac{E_{Be^-}}{e^2}. \] (1)

where \( m \) is the total mass of the atom, \( Z \) is the atomic number and \( N \) is the mass number. Furthermore \( m_p, m_n \) and \( m_e \) are describing the mass of the proton, neutron and electron, respectively. The speed of light is denoted as \( c \) and the binding energy of the nucleus and the electron is denoted with \( E_{B\text{nuc}} \) and \( E_{Be^-} \) respectively. The highly accurate determination of atomic masses provides detailed invaluable inputs to a wide range of physics communities. Indeed, determining the mass of an atom with a relative precision in the \( 10^{-6} \) to \( 10^{-8} \) range gives access to the nuclear binding energy, a quantity which allows tracking the evolution of nuclear structure phenomenon as a function of \( Z \) and \( N \) and enters as a primary input in all astrophysical nucleo-synthesis calculations. Bellow a relative precision of \( 10^{-8} \), high precision atomic mass values can also significantly contribute to the study of the weak interaction [1].

The advancement of the Penning-trap based mass measurement techniques, roughly four decades ago, opened the field of mass spectrometry into the high precision era. Nonetheless, a comprehensive assessment of the various sources of systematic errors along with their unavoidable respective magnitudes is required. Since the atomic mass unit is defined as 1/12 of the mass of \(^{12}\text{C}\) the use of carbon clusters of various sizes was proposed to carry out such a study [2] since the carbon cluster of various sizes are covering all of the mass range available to an experiment as seen in Fig. [1] In this case, the ions can be provided by a laser ablation source similar to the one which have been implemented in 2002 at the pioneering Penning-trap mass spectrometer ISOLTRAP [2]. In the light of the recent upgrade of the ISOLTRAP setup during CERN’s long shutdown 2 (LS2), a redesign of the laser ion source (LIS) will be presented in this report. Such a source of carbon cluster reference ions, is especially crucial since both the vertical and horizontal part of the ISOLTRAP beamline have been completely maintained during long shutdown 1 and 2, respectively. Furthermore, with the recent implementation of phase-imaging ion-cyclotron-resonance technique for cyclotron-frequency measurements [3] at ISOLTRAP, such a source will also be used to study the systematic effects of this particular technique. In the following, the ISOLTRAP experimental setup and an overview of the various iteration of the laser ions source will be presented. Additionally a short description of the production of ions and clusters by laser-induced plasma will be highlighted. Finally the present configuration of the source will be presented together with the corresponding optimization by ion optics simulations.

II. THE ISOLTRAP EXPERIMENT

The high-precision mass spectrometer ISOLTRAP has obtained data of more than 400 exotic nuclides with half-lives as short as 48 ms [4] and reached a record relative mass uncertainty of \( 9 \times 10^{-10} \) [5].

This experimental setup receives ions from CERN’s radioactive ion beam facility ISOLDE. At ISOLDE a wide range of short-lived nuclides are produced through spallation-, fragmentation- or fission-reactions using a 1.4 GeV proton beam (\( \sim 3 \times 10^{13} \) protons per pulse) delivered by CERN’s Proton Synchrotron Booster (PSB). In this way, a large variety of nuclides from \( Z = 9 \) to \( Z = 92 \) can be produced with half-lives down to few ms. The flow of neutral radioactive atoms diffusing out of the target material is subsequently ionised (using surface, plasma or resonant laser ionisation) before being accelerated to an energy of 30 keV to 50 keV. The isobar of interest can then be separated from the contaminants using one of two dipole magnets HRS (High Resolution Spectrometer) and GPS (General Purpose Spectrometer) with varying mass resolving power [6].

The ions that arrive to the ISOLTRAP, are stored inside an helium buffer-gas filled Radio frequency quadrupole (RFQ) for accumulation, cooling and bunching (typical trapping time 10 – 20 ms) [7]. The bunched beam is subsequently injected into the ISOLTRAP multi-reflection time-of-flight mass separator (MR-ToF MS) for fast isobaric separation (typically \( R = m/\Delta m = 100.000 \) in 22 ms for mass 85) [8, 9]. In the low production and short half life regime, this device can also used as a mass spectrometer in its own rights reaching relative mass precision in the \( 10^{-6} \) regime [8]. After this, the ion bunch is transferred to a cylindrical Penning trap used for further cooling [10]. Finally, the ion beam is delivered to a hyperbolic Penning trap where the measurement is performed.
The measurement techniques rely upon the determination of the true cyclotron frequency of the ions of interest which is given by $v_c = qB/(2\pi m_{\text{ion}})$ where $B$ is the strength of the confining magnetic field, $q$ is the ion charge and $m_{\text{ion}}$ is the mass to be determined \[1\]. The measurement procedure is beyond the scope of this report, for further information detailed descriptions of the procedures can be found in Refs. \[3, 12–14\]. A schematic representation of the current experimental setup is shown in Fig. 2.

Since the original design of the LIS of Ref. \[2\] the laser ions source design has continuously been updated to improve performance. As a result, the work presented in this report is a direct continuation of the work disrobed in Refs. \[16–21\].

### III. LASER ABLATION AND IONIZATION FOR ION PRODUCTION

The production of ion clusters by emission from laser-induced plasma is an essential tool for calibration in multiple state of the art mass spectrometers \[2, 22–24\]. In this section we will shortly be detailing the working principles of a laser ablation source (for further details see Ref. \[25\]). In the case of using a long laser pulse in the nanosecond regime, the various features of the laser-target interaction region during irradiation in vacuum are presented on Fig. 3. The predominant effect arise from the pressure gradients, which give an ion acceleration in the direction perpendicular to the target surface \[26–28\], as seen in region I of Fig. 3. The velocity distribution of the ions form a Maxwell-Boltzmann distribution, however effected by a shifted center of mass velocity due to the Knudsen layer \[29\]. In region II of Fig. 3 after the laser impact, a plume will adiabatically expand in the direction perpendicular to the sample. This plume, which typically fulfil the Anisimov model \[30\] for hydrodynamic motion (i.e. neglecting the electrostatic interaction). In region III of Fig. 3 the electrostatic interaction between the electrons and the ions created in the ablation process becomes non-negligible when the ions are further away from the sample as indicated in Ref. \[31\] \[32\]. Here the kinetic energy-distribution of ions was reported larger than that of neutral atoms, suggesting an electrodynamic acceleration process for ions due to a high electric field in the non-equilibrium laser induced plasma.

The time response of the electrons to the laser is much faster than that of the ions and the laser pulse itself. As the electrons and the ions have the same thermal temperature, the lighter electrons will reach the highest thermal velocities and will escape the plasma first. This separation will induce an electric field, which results in a proportionality between the kinetic energy and the charge of the accelerated ions. Since the electrons and ion initial motion is dominated by the hydrodynamic model, they are distributed perpendicular to the surface of the target as illustrated in Fig. 3.

However these phenomena are strongly dependent on the laser intensity. At fluences of $10^5 - 10^7$ W/cm$^2$ the kinetic energy distribution of the ions are often less than 5 eV, leading to a Maxwell-Boltzmann-like kinetic energy distribution. At higher fluences however, the kinetic energy of the ions shift towards higher energies and the distribution broadens. This is also observed in the angular distribution due to non-linear interactions between the laser beam and the plasma as sketched in Fig. 4. This however only happens until a certain fluence threshold \[32\].

The laser wavelength will also be an important pa-
parameter. The laser which will be used at ISOLTRAP is a frequency double 532 nm Nd:YAG laser, which has shown in experiments to give a narrower angular distribution with a better defined and lower average kinetic energy distribution than a comparable CO₂ laser [33]. In general, the shorter wavelength seems to imply a higher production of singly charged ions [34].

At ISOLTRAP the laser system provides a laser pulse length in the nanosecond regime. This is a lot slower than the picosecond electron-lattice relaxation time, and therefore allows for the thermodynamic ablation [35] (i.e laser evaporation and plasma heating). A shorter laser pulse (e.g in the femtosecond regime) would result in larger energy distribution [36] due to thermal diffusion energy. Hence, the ISOLTRAP laser system allows the production of laser ablated ions with a high production rate with a relatively cold and well defined kinetic energy distribution. From these considerations as well as the ablation knowledge from Refs. [31, 37], one can consider that the ions’ velocity distribution can be described as a shifted Maxwell Boltzmann distribution. The laser ablated ion beam profile is assumed to follow the same profile as the incoming laser pulse, i.e it is assumed to follow a Gaussian distribution.

IV. HISTORICAL OUTLINE OF THE LASER ION SOURCE

Since the implementation of the LIS in Ref. [2] a few upgrades has been made. The initial LIS system is depicted in Fig. 5.

The first work done by Boehm [19] consists of adding a steerer between the einzel lens and the quadrupole bender as well as constructing an attenuating filter for the laser setup and changing some insulating components to avoid shortcuts. Due to laser fluctuations resulting in ion fluctuations, a complete redesign of the laser system was done again by Fink [20] who changed the LIS to a pierce setup as depicted in Fig. 6 with a slight displacement of the rotating feed through to make sure, that the laser did not ablate the same spot on the carbon pellet.

In order to further improve the yield of the laser ablation process Lommen [18] did a redesign of the formation chamber varying the variables in Fig. 7 for a most efficient pierce electrode configuration. Furthermore Lommen added a telescope lens to the laser-system and optimized its impact on the performance of the LIS.

Additionally Murray [16] and another Summer student [17] did further small correction to the potentials and
A summary of the performance of the LIS was made by Manea [21] to determine what improvements were to be done for the LIS in the future. Large cluster production fluctuations still persist for the LIS. To solve this an increase in laser fluence is made to stabilise the ion production, but the fluence-increase lead to a much warmer ion-distribution with a much broader energy distribution. This severely reduce the effect of the cooling and separation done in the lower trap as well as reduce the transport efficiency of the ions between the two Penning traps. This situation require a trade-off between having a significant cluster-count in the upper trap, while still having a cold and well defined cluster-distribution. The problems of poor cooling in the lower trap is hereby intertwined with the problem of having count-rate fluctuations. In addition to this Manea suggests a recommission of the pellet-turning motor, that would often break down, which together with the modifications made by Fink [20] in a pierce geometry should ensure a faster extraction of the ion cloud and eliminate any high count bursts from sparks or charging up the insulator.

V. PRESENT AND FUTURE LIS MODIFICATIONS

In this section, a new implementation of the LIS system as well as the corresponding ion transport simulation will be presented.

A. New positioning of the Laser Ion Source

The recent addition of a second high performance time-of-flight detector in the ISOLTRAP beamline is such that a modification of the layout of the laser ablation setup is required. An easy, solution would be to remove this detector. However being able to use such a device to characterise and optimise the laser source output would be highly beneficial. In addition, the most recent modification of the laser ion source itself is such that the laser path is blocked and therefore cannot be shine from the side as it once was. Therefore a total redesign of the laser chamber would be necessary in order to keep a relatively stable setup, and not to return to the lass stable setup of Fig. 5. By acknowledging the considerations and optimizations that previous studies have done. This complete redesign might be a very time
consuming procedure. A much simpler solution consists in placing the source parallel to the ISOLTRAP horizontal beam line and to keep the incoming laser beam normal to the pellet surface. The simple addition of an electrostatic quadrupole bender should allow for the laser ablated ion beam to be bend towards the time-of-flight detector. Such a setup is an ideal solution, since we will be able to keep the previous optimizations for the LIS system. The transport of ions from the source to the time-of-flight detector has been simulated and optimised using the SIMION simulation software. The details of the simulation will described in the next section.

**B. Simulations of the redesigned setup.**

With the new setup we are able to send the carbon clusters both straight to the time-of-flight detector for characterization and right for the calibration of the tandem Penning traps. Fig. 7 shows a representation of the simulated section of the ISOLTRAP beamline within the 3-D SIMION software.

The ion velocity distribution is defined as done by Manea in Ref. 21, as a shifted Maxwell Boltzmann velocity distribution, that are formed on the Gaussian laser spot. This ion distribution is transported towards the time-of-flight detector by the defined electric fields to reach the highest transport efficiency. In order to find the optimal electrode voltage configuration, an optimization algorithm was used. The optimization algorithm is described in the following subsection and will be the basis of all of the ion-optics simulations presented in this report.
FIG. 9. Simion 2-D drawing of the LIS from the initial ion chamber with the extraction electrodes, einzel lense and steerer. From here a quadrupole bender (QP-1) will be attached to the newly commissioned quadrupole bender (QP-2) from where it can be sent right to the tandem Penning traps or straight to the time-of-flight detector as shown in Fig. 2.

1. **Downhill Simplex Method for Optimization**

The optimization algorithm have been written as a luar-user-program, which is intertwined with the features of SIMION. The working principles of this algorithm is that of a downhill simplex. The algorithm, is described in length in the literature see e.g. Ref. [38]. It is a method of minimizing a function in an \( n \)-dimensional space by transforming a simplex (a polytope of \( n + 1 \) vertexes). These transformations are based on the function values at each vertex and will let the simplex progress through reflections, contractions and expansions as described in the following algorithm.

REPEAT:
- find highest, lowest, and centroid points of the simplex
- try reflection
  - IF \( \psi(\text{reflected}) < \psi(\text{lowest}) \):
    - try expansion
    - IF \( \psi(\text{expanded}) < \psi(\text{reflected}) \):
      * accept expansion
    - ELSE:
      * accept reflection
  - ELSE:
    - IF \( \psi(\text{reflected}) < \psi(\text{highest}) \):
      * accept reflection
    - ELSE:
      * try contraction
      * IF \( \psi(\text{contracted}) < \psi(\text{highest}) \):
        * accept contraction
      * ELSE:
        * do reduction
  UNTIL converged (e.g. size (simplex) < tolerance).

The downhill simplex has been acknowledged for it’s stability and for the fact that the procedure, do not need to calculate derivatives. The calculation of numerical derivatives requires to call the function multiple times and this is very ineffective for a complicated SIMION simulation, therefore the downhill simplex is very well suited for these optimizations, but is relatively slow for simpler systems in few dimensions. The algorithm which has been developed uses the newest version of the integrated SIMION downhill simplex library as well as the integrated SIMION-test-plane library for diagnostics of the ion-beam. The exact criteria for convergence differs a bit depending on the simulation and will be described in relation to the exact situation in use, as done in the following section.

2. **Results of the Simion Optimization**

In the case of the transport optimization towards the time of flight detector one would want maximum ion transmission towards the active window of the detector (35 mm \( \times \) 25 mm). Hence, the cost function for the optimization was chosen as

\[
f(\text{\( N_{loss}, \sigma_{yn}, \sigma_{zn} \)}) = N_{loss} - \left(1 - \frac{\sigma_{yn}}{2} - \frac{\sigma_{zn}}{2}\right),
\]

where the factor which contributes the most is \( N_{loss} \) which is the integer number of ions that is lost during transport. This number is calculated in SIMION
as the number of initial particles minus the number of particles which reaches the detector. Since the algorithm should not only minimise the number of lost ions but also find optimal voltage settings for the beam to go through the entrance aperture of the detector, the loss function should also carry information regarding the focusing of the beam. In practice, this is taken care of by the second term of the loss function \(L\), which is weighted to at largest be an integer, so it is always more efficient to get an additional particle to hit the detector compared to any additional focusing of the beam. The standard deviations of the second term must therefore be normalized by the normalization

\[
\sigma_{in} = \frac{\sigma_i}{\Delta_i/2}
\]

where \(\sigma_i\) is the statistical standard deviation of the particles hitting the detector with a detector-size \(\Delta_i\) in the corresponding dimension.

Additionally for the test of bending the beam directly towards the MR-ToF or the tandem Penning traps an extra factor have been added to this expression. This factor is making sure that the beam stays in the center of the path, since the 3-D models of the other parts of the setup has not been implemented in this simulation yet, and we therefore will not be able to define a detector to observe the particles as for the optimization into the time-of-flight detector. This additional factor will be described as

\[
\Delta f = -(1-w_x \times (\sigma_x - x_0) - w_y \times (y - y_0))
\]

Here the position is offset by the values \(x_0\) or \(y_0\) which is the desired position of the mean of the particles. Weights can be defined by normalizing to the beam-pipe-width. However in order to gain insight one must do a complete evaluation of different weights. This have for some cases been done, and the results are documented. However since this is only a test of the bending potentials a more in-depth analysis should be performed, when the system is assembled and a complete simulation can be done of all of the components.

The simulation of the ions flying straight towards the time-of-flight detector is a very complicated 27 dimensional minimization problem, for which in each iteration a large enough ion-simulation must be made to accurately simulate the ion-distributions from Sec. III. This can however be condensed to an 18 dimensional minimization problem through the use of symmetries, experimental knowledge, and restrictions on the electrostatic quadrupole bender and this optimization have been solved by the downhill simplex algorithm of Sec. V.B.1.

Using initial guess values as defined in Ref. [16–18, 20], the optimization have been run through multiple iterations. The results of the electrode potential are saved and an the result of the simulation is presented in Fig. 10. With this optimized electric fields one can achieve a transport efficiency of 77%. Starting with a 1000 ions, a 100% transmission is achieved through the first bending section. The first bottleneck is the injection side the second quadrupole bender. Indeed, the lack of focusing between the two quadrupole benders, can explain why 21% of the ions are lost at the entrance and exit of the new bender. This is the main source of ions loss in this configuration. A few percent of the remaining ions are lost before reaching the detector chamber and of these ions only 1% miss the detector in the detection chamber.

The simulations of bending towards the MR-ToF or towards the lower Penning trap are very similar to Fig. 10 just with an additional bending in the second quadrupole bender and a data-collection on an interaction plane located at the edge of the simulation grid. The transport efficiency is up to 93% for those simulations, however by adjusting the weights of the convergence criteria, a more narrow beam with the correct center position can be achieved by compromising on the efficiency. Such an estimation can however only be done satisfactory by introducing the additional parts of the setup into the simulation.

C. Current status of the LIS

By the results of the simulation in the previous section, the LIS are now ready to be assembled and attached in the new location. The immediate problem of implementing the LIS have hereby been solved, and the assembly is awaiting the alignment of the beamline to be completed.

However the problems of cluster production fluctuations, poor cooling and separation in the lower trap, and recommission of the pellet-turning motor as suggested by Manea [21] will be solved. The redesign of the ion position have been made with the cluster production fluctuations in mind, and the most efficient design have been suggested with the current equipment, as indicated by the studies of Fink [20].

The faulty pellet-turning motor have been investigated. The investigation have been done by compari-
son to the similar laser vaporization system used in the Cryogenic Storage Ring at the Max Planck Institute for Nuclear Physics (MPIK) in Germany of Ref. [39]. However the system in use at the MPIK have spacial restrictions on turning the pellet holder head-on with the engine and therefore require a vacuum rotary drive and a dedicated combination of gears in order to turn the pellet. Since such spacial restrictions are not present at the ISOLTRAP setup, such complication to the setup are not necessary. Therefore an evaluation of the present problems of the rotating pellet stage have been made in order to find a solution. The current pellet-holder of the ISOLTRAP LIS works by a slightly shifted rotateable sample holder, which is placed at the end of the LIS as depicted in Fig. 6. This rotateable sample holder lies on ball-bearings and are connected to an engine through an insulating connector as shown in the bottom of Fig. 11. The problem of the sample holder relies in the insulating rotating feedthrough. Since this is very flexible, if the screw connected to the engine starts to unscrew, then the insulator might be compressed enough for the two flexible metal plates to meet, thus creating a spark, or for it just to be sliding off the ball-bearing and de-attach from the motor, which will result in a breakdown, where one would need to open the chamber and break the vacuum in order to attach the sample-holder to the engine again. Since no insulated rotateable feedthrough are made by the regular suppliers that are tiny enough for the space between the engine and the sample holder, then several other solutions have been thought out. One option is to do a modification of the current rotateable feedthrough in order to make it more rigid. Another option would be to replace it with another connector like the ones shown above in figure 11. These connectors are however not insulated, so the insulation could in those cases be solved by a plastic or rubber coating on one or both ends. However when in operation this coating might tear up, and the 3kV extraction voltage on the sample holder might cause sparks to the grounded motor.

The most stable solution might therefore be to simply replace the rotating feedthrough by either one of the metal connectors shown in Fig. 11 or by a metal rod. This could simply be achieved by building a small high voltage cage around the engine and float the equipment by the extraction voltage. There were unfortunately no time for setting up such a cage.

D. Future development

The reattachment and implementation of the LIS as described in the previous sections should be performed. Considering any improvements to the current setup, a new Nd:Yag laser have been kindly provided by the ISOLDE RICIS team and will allow for a more stable beam and higher quality TEM(0,0) Gaussian mode for low intensities. This would be predicted to have a stabilizing effect on the carbon cluster production, and would be the ideal laser for the carbon cluster production.

Furthermore from the simulations in Sec. V B 2 we see that ion-transportation could be made more efficient especially around the second quadrupole bender, where a lens between the two benders might be able to increase the transport efficiency.

The next major upgrade to the system could be introducing a RF-cavity before the extraction electrode in the LIS. This way, the ions can be trapped, as they are produced, and sent as a cooled bunch to the different parts of the set up. This might possibly solve many of the problems with the current setup, since the ions can be produced at lower laser intensities, giving a colder and fewer ions with a more narrow velocity distribution. By initially bunching ions from multiple laser-pulses the LIS would be able to produce bunches of cold ions, with low production fluctuations, which can be transported more efficiently and will improve the resolution of the Penning traps significantly to further improve the calibration of the ISOLTRAP. A sketch of such an implementation is shown in Fig. 12.

Using such a setup one would be able to produce ions with a narrower energy distribution. To test whether this procedure will work, a simulation has been done using the parameters and optimization algorithm from the simulation of Fig. 10. This simulation as seen in Fig. 13 show that with such a set up a transport efficiency of 72% can be achieved of the ion bunches, without the steerers of the present setup. This transport efficiency could go even higher if one would take into account that the ions
FIG. 12. Simion 3-D drawing of future LIS with the initial RFQ and einzel lense. From here a quadrupole bender (QP-1) will be attached to the newly commissioned quadrupole bender (QP-2) from where it can be sent right to the tandem Penning traps or straight to the time-of-flight detector as shown in Fig. 12.

in that case could be produced with a lower kinetic energy spread. The implementation of such a system must therefore be considered, and could be implemented as in Fig. 12. By such a system all of the modifications of this project will still be valid, since the consideration of the rotation targetholder will still be in use as well as the pierce geometry requiring the additional bending. Once such a system would be implemented, an updated 3-D sketch could be made and the downhill simplex algorithm could be reused with small modifications. Unfortunately no time could be dedicated to the simulation of the ion trapping and cooling with buffer gas in the RF cavity.

FIG. 13. Simulation of the ion transport from the proposed future LIS to the time-of-flight detector on the electric field surface. The ions are produced as defined in previous simulation through electrodes with optimized voltages using the downhill simplex optimizer. For scale, the formation electrode is typically 3kV.

VI. SUMMARY

During this project a detailed investigation have been done in the underlying theory and the many experimental advances that have been made to create the LIS for the ISOLTRAP. The decision was made to keep the current LIS implemented and to modify it for present use. The proposed solution are backed up by ion transport simulations, optimized by an implementation of the downhill simplex method, which is easy do adapt for future simulations and investigations of other parts of the experiment. With the results of the simulation everything is now ready for the actual implementation and tests once the alignment and reassembly of the ISOLTRAP setup is finished. Furthermore an analysis of the weaknesses of the current LIS have been done, and several solutions has been addressed, including the simulation of a completely redesigned LIS with a RF quadrupole in the formation chamber although without proper cooling of the carbon ions in the RFQ. With the implementation of the equipment as described in this report and using the optimal voltages from the simulation data, then one are be able to efficiently implement the LIS to function for current use, and have a guideline for future improvements and upgrades.

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