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**„Multiphysics simulation of negative ions in a gas filled
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Abstract

Accelerator mass spectrometry (AMS) is an ultra sensitive technique for measuring smallest amounts of trace isotopes with isotopic ratios down to 10^{-12} - 10^{-16} . The main challenge of AMS is the effective suppression of unwanted isobars. These isotopes of different elements with almost equal mass cannot be separated by conventional mass analyzers and are usually many orders of magnitude more intense.

Laser photo detachment of anions is a novel technique for isobar suppression based on the selective neutralization of isobaric background by the means of a suitable intense laser. The Ion Laser Interaction Mass Spectrometry (ILIAMS) project at the University of Vienna is the first implementation of this approach as part of an AMS-facility, in this case the Vienna Environmental Research Accelerator (VERA). The key component of ILIAMS is a negative ion cooler based on a gas filled radio frequency quadrupole (RFQ).

This master's thesis describes the development of a simulation model capable of accurately computing trajectories of ions transiting the RFQ cooler using the multiphysics simulations software COMSOL. The generated model includes results of buffer gas distributions based on rarefied gas dynamics. Formerly unresolved geometric effects on pressure and density are presented along with experimental validation. Gathered information is combined with FEM-calculations of applied static and time dependent electric fields. Further simulations of ion-gas collisions apply an elastic scattering variable hard sphere (VHS) collisional cross section. The developed model reproduces experimental $^{63}\text{Cu}^-$ residence times of 8.2 ms at low beam currents with high accuracy. Furthermore, changes with varying buffer gas pressure are in good agreement with experimental data. An additional approach with the purpose of approximating occurring space charge effects gives insight into the possible formation process of such charge distributions, known to strongly affect ion residence times. The constructed model will extend the possibilities to perform modifications and optimizations on the RFQ cooler, improving the ongoing research on the ILIAMS setup at VERA.

Zusammenfassung

Beschleuniger Massenspektrometrie (AMS) ist eine extrem sensitive Methode zum Nachweis kleinster Mengen von Spurenisotopen bis hin zu Isotopenverhältnissen von 10^{-12} - 10^{-16} . Die größte Herausforderung für AMS besteht in der effektiven Unterdrückung von unerwünschten Isobaren. Diese Isotope anderer Elemente, jedoch beinahe gleicher Masse, können mit konventionellen Masseseparatoren nicht abgetrennt werden und sind üblicherweise um mehrere Größenordnungen intensiver.

Laser photo detachment von Anionen ist ein neuartiges Verfahren zur selektiven Neutralisierung und Unterdrückung von isobarem Hintergrund mittels Einsatz eines intensiven Lasers geeigneter Wellenlänge. Das Ionen Laser Interaktions Massenspektrometrie (ILIAMS) Projekt an der Universität Wien ist die erste Umsetzung dieses Ansatzes als Teil einer AMS-Anlage, in diesem Fall des Vienna Environmental Research Accelerator (VERA). Das Kernstück von ILIAMS ist ein Anionenkühler auf der Grundlage eines gasgefüllten Radiofrequenz Quadrupols (RFQ).

Diese Masterarbeit beschreibt die Entwicklung eines Simulationsmodells zur präzisen Berechnung der Bahnen der Ionen, welche diesen RFQ Kühler durchqueren. Für diesen Zweck wurde die Multiphysik Simulationssoftware COMSOL benutzt. Das generierte Modell beinhaltet Resultate von Buffergas Verteilungen anhand der Theorie verdünnter Gasströmungen. Vormalig unbestimmte Einflüsse der verwendeten Geometrien auf Druck und Dichte werden zusammen mit einer Validation anhand experimenteller Daten vorgestellt. Diese Ergebnisse werden mit FEM-Berechnungen statischer bzw. zeitabhängiger elektrischer Felder kombiniert. Weitere Simulationen von Ionenkollisionen mit dem Buffer Gas verwenden einen variable harte Sphären (VHS) Wirkungsquerschnitt. Das entwickelte Modell reproduziert experimentelle $^{63}\text{Cu}^-$ Aufenthaltszeiten von 8.2 ms bei niedrigen Ionen-Stromstärken mit hoher Genauigkeit. Änderungen mit wechselndem Gasdruck stimmen gut mit experimentellen Daten überein. Ein zusätzlicher Modellierungsansatz zur Annäherung von auftretenden Raumladungseffekten liefert Einsicht in die mögliche Entstehung und Entwicklung solcher Ladungsverteilungen, welche die Aufenthaltszeiten stark beeinflussen. Das erstellte Modell erweitert die Möglichkeiten zur Anpassung und Verbesserung des RFQ Kühlers und sollte die permanente Forschung bei VERA und ILIAMS positiv beeinflussen.

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List of Abbreviations

Name	Abbreviation
electron affinity	<i>EA</i>
accelerator mass spectrometry	AMS
backwards differentiation formula	BDF
computer aided design	CAD
center of mass	CM
density functional theory	DFT
direct simulation Monte Carlo method	DSMCM
extraction high lens	EXTHI
extraction low lens	EXTLO
finite element method	FEM
full width half maximum	FWHM
graphical user interface	GUI
Holifield Radioactive Ion Beam Facility	HRIBF
hard sphere	HS
high voltage	HV
Ion Laser Interaction Mass Spectrometry	ILIAMS
Ion Laser Interaction Setup	ILIAS
injection high lens	INJHI
injection low lens	INJLO

List of Abbreviations

Name	Abbreviation
light amplification by stimulated emission of radiation	laser
mass spectrometry	MS
random access memory	RAM
radio frequency	RF
radio frequency quadrupole	RFQ
root mean square	RMS
the stopping and range of ions in matter	SRIM
Vienna Environmental Research Accelerator	VERA
variable hard sphere	VHS

List of mathematical Symbols

Name	Symbol	Unit
Boltzmann constant	k_B	J/K
electrical conductivity	σ_e	S m^{-1}
electron affinity	EA	eV
elementary charge	e	C
Knudsen number	Kn	1
mass to charge ratio	m/q	kg/C
Mathieu parameter	q_M	1
permeability of vacuum	μ_0	H/m
permittivity of vacuum	ε_0	F/m
rarefaction parameter	δ	1
relative permittivity	ϵ_r	1
speed of light in vacuum	c_0	m/s
universal gas constant	R	$\frac{\text{kg m}^2}{\text{s}^2 \text{mol K}}$

1 | Introduction

The behavior of a charged particle subjected to electrostatic and magnetic fields is determined by its charge state q , its kinetic energy E and its momentum p , respectively. At fixed fields, ions are deflected according to their masses, more precisely their mass to charge ratio m/q . This trait can be used to perform mass analysis on probing materials, a technique called mass spectrometry (MS). Accelerator mass spectrometry (AMS) unites conventional MS with the high kinetic energies accessible by particle accelerators to greatly increase sensitivity. The highly efficient suppression of unwanted isobaric background allows for measurement of isotopes with isotopic ratios as low as 10^{-12} to 10^{-16} [1, 2, 3].

Especially in the case of long lived radionuclides at their natural abundances, AMS is arguably the most sensitive method available [1, 3]. Conventional MS fails to separate signals of naturally produced, long lived radioisotopes from its neighbouring masses that are many orders of magnitude more intense [2]. Classical decay counting either requires long counting times or extremely large sample sizes in order to obtain signals above background noise [1].

The investigated isotopes of interest depend on the field of research, some examples include [1]:

- ^{14}C - radiocarbon dating of environmental and archaeological samples
- ^{26}Al , ^{10}Be , ^{36}Cl - geological dating, e.g. earth magnetic field, exposure dating of rocks
- ^{236}U , ^{135}Cs - tracers for transport processes of anthropogenic radionuclides, e.g. investigation of seawater currents
- ^{60}Fe - signal detection from past supernovae

A more detailed overview of AMS isotopes of interest can be found in Kutschera et al. 2013 [1].

1.1 AMS at VERA

The Vienna Environmental Research Accelerator (VERA) is a medium sized, state-of-the-art AMS facility at the University of Vienna, based on a 3 MV tandem accelerator and suited to measure the full range of known isotope species [4, 5]. A schematic drawing of the facility is given in Figure 1.1. On its low energy side, two sources each contain a target wheel that can hold up to 40 targets. These are sputtered with cesium, producing a negative ion beam on the order of some μA that is extracted and preaccelerated up to 75 keV or 30 keV, depending on the source. A low energy mass spectrometer (combination of an electrostatic analyzer and a bending magnet) passes the fraction of desired mass to charge ratio (m/q) on into the accelerator's terminal, which can be set to a voltage of up to 3 MV. Therein, the charge state of the beam is changed by removing electrons either via passing of a foil stripper, or through collisions with a stripper gas. The resulting positive ion beam contains, apart from the ions of interest, broken fragments of molecules of different charge states and masses. It is filtered again at the high energy mass spectrometer, followed by a measurement on the detector side [4, 5]. Selecting the charge states after stripping needs to take into account fragments of the same m/q ratio. When measuring ^{36}Cl for example, the 6+ charge state is restricted due to its strong interference with $^{12}\text{C}^{2+}$ injected as C_3 [6].

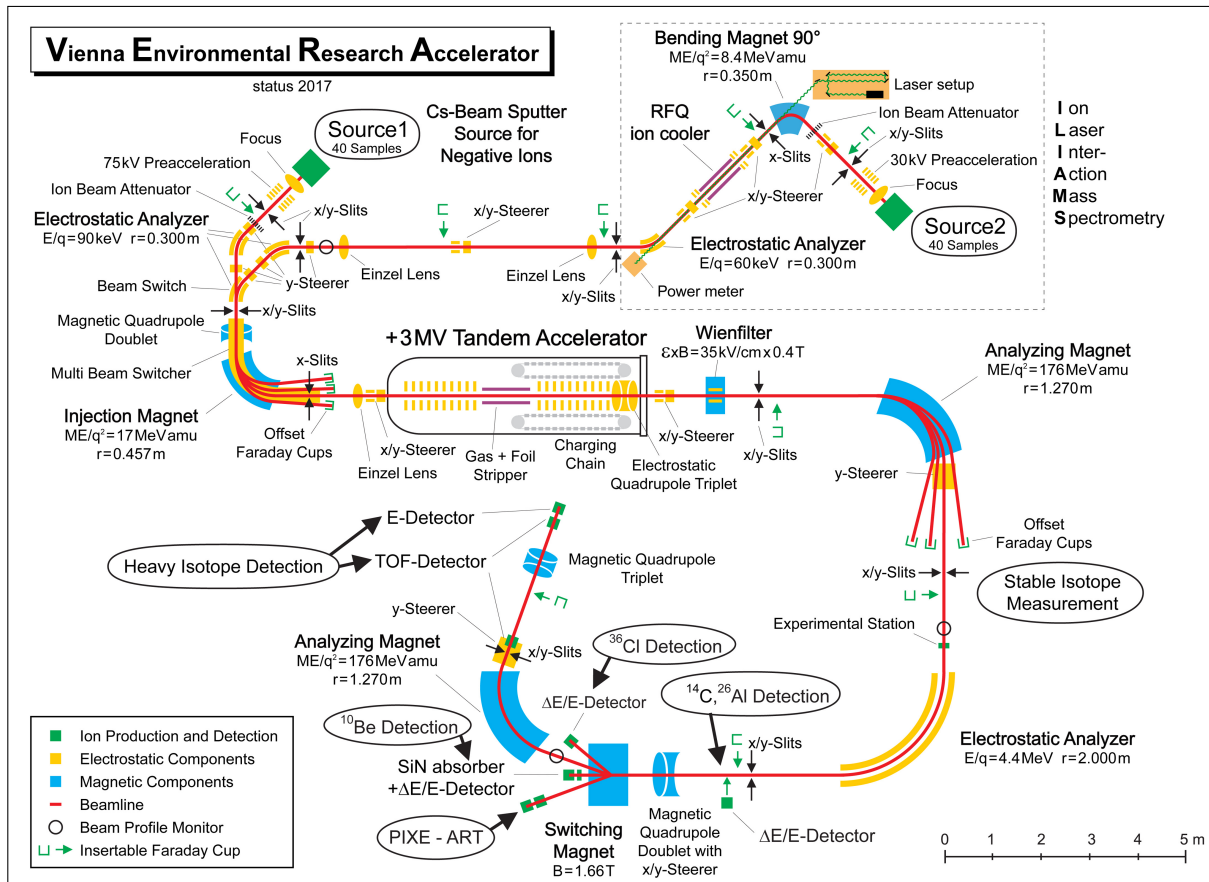


Figure 1.1: Schematic of the VERA facility (taken from [4]).

1.2 Isobar suppression in AMS

Isobars differ in atomic number Z but share the same mass number A . If isobars of a target isotope are present in the same charge state, they cannot be separated using electrostatic and magnetic analyzers, assembling an unwanted *isobaric background* when performing AMS. At comparable intensities, mass separation of nuclear isobars at high resolution is feasible. In reality, disturbing isobars are many orders of magnitude more intense [3].

In most cases when measuring long lived nuclear isotopes at their natural abundances, the suppression of at least one stable isobar is necessary [2]. A variety of different methods and techniques are applied in the frame of AMS to fathom this problem, including but not limited to:

- The usage of a negative ion source, which already excludes elements with negative electron affinity. However, this only applies to the minority of elements, such as ^{14}N and ^{26}Mg , isobars of ^{14}C and ^{26}Al respectively [3].
- Choosing a molecular form of the desired isotope during injection for which the formation is suppressed or not possible in the case of the isobar. An example would be the suppression of ^{41}K against ^{41}Ca when extracting CaH_3^- [7].
- Fully stripping the isotope of interest in case its isobar has a lower Z and therefore cannot reach the same charge state [8]. This is of course limited by the acceleration energy and only pursued at the largest AMS facilities. Fully stripped ^{36}Cl measurements have been performed using a 25 MV accelerator [9].
- Exploiting the different energy loss behavior in matter dependent on the atomic number Z . According to the situation, this is either realized by recording the energy loss spectra of the ions when passing through matter, or by using an absorber only passable by ions with a Z below a certain threshold [10].

Feasibility of the described separation techniques depends on the $\Delta Z/Z$ ratio between isotope of interest and its isobar. In addition, the performance of the above techniques, as well as the accessible charge states, are limited by the available acceleration energy. The range of radioisotopes accessible for AMS is therefore limited, slightly more for smaller and middle sized facilities such as VERA [3]. While a few isotopes (e.g. ^{60}Fe , ^{99}Tc) can still be measured at larger facilities, separation from isobaric background is not possible for mass numbers above 100. As a result, about half the radionuclides of interest for AMS are not accessible with conventional isobar suppression techniques [4].

1.3 Motivation of this master's thesis

Within recent years, the development of new isobar suppression techniques has been pushed to overcome the limitation described in Section 1.2, yielding two promising approaches. One of them exploits the selective chemical reaction of interfering isobars with a reactive gas and was first explored and developed by a group in Toronto [11].

The second method uses optical filtering and is based on the selective neutralization of isobaric background via laser photo detachment. Within the past ten years, a novel injection setup, the Ion Laser Interaction Mass Spectrometry (ILIAMS), has been developed and installed as an extension to VERA by the Isotope Physics Group [4]. This setup enables the measurement of isotopes suffering from too strong isobaric background [12, 13, 14]. Its centerpiece is a RFQ negative ion cooler filled with a low pressure buffer gas (see Section 2.2, The ILIAMS RFQ negative ion cooler).

This master's thesis describes a computational approach to model the behavior of negative ion beams in the RFQ ion cooler. Former works by Johanna Pitters (2015, [15]) and Tobias Moreau (2016, [16]) already generated simulation models using the software SIMION ([17]). However, some important processes and influences could not be properly included in these models, which hampered further development of the setup. A source of great uncertainty emerges from the lack of knowledge regarding the buffer gas pressure distribution inside the cooler. Measurements at the gas inlet suggest it to reside in the range of several tenths of mbar. However, comparable experimental realizations of selective laser photo detachment with a negative ion cooler performed by Liu et al. [18] at the Oak Ridge National Laboratory report an operational pressure of only 6 Pa.

This thesis uses the multiphysics simulation software COMSOL to model the ILIAMS RFQ negative ion cooler [19]. Contrary to SIMION, it allows not only for the computation of ion trajectories and electric fields, but also for rarefied gas dynamics simulations [20]. Moreover, none of the former models considered the influence of emerging space charge effects, which experimentally have been shown to strongly influence the residence times of ions within the RFQ cooler for beam currents above several nA [14]. Although the final model presented in this thesis does not include a fully developed tool to precisely describe emerging space charge effects, a lot of effort has been put into tackling this problem. Parts of it are presented in Section 3.7.

The COMSOL model constructed during the scope of this thesis unites the simulations of rarefied gas dynamics, generated static and time dependent electric fields, as well as the transit of charged particles through the ILIAMS RFQ negative ion cooler currently operated at VERA. It allows for a realistic prediction of residence times of ions at low beam currents and can provide a basis for the testing of eventual modifications and optimizations of the installation.

2 | Theory

2.1 Laser photo detachment of negative ions

As described in Section 1.2, conventional isobar suppression methods require the kinetic energy and the ratio $\Delta Z/Z$ to be sufficiently high. An element-selective technique not restricted by this is the principle of laser (light amplification by stimulated emission of radiation) photo detachment of negative ions. This means the removal of the additional electron from a negatively charged ion by its interaction with a photon and is described by



The binding energy between the surplus electron and the neutral core atom or molecule is given by the electron affinity (EA). A detachment can occur, if the energy of the involved photon E_γ exceeds the EA of the respective atom or molecule. Such a reaction is a non resonant threshold process [21, 22]

$$E_\gamma = h \cdot \nu \geq EA \quad (2.2)$$

with h being the Planck-constant and ν the photon frequency. The cross section σ around the threshold energy can be described by Wigner's law [22]

$$\sigma(E_\gamma) = \begin{cases} 0, & E_\gamma < EA \\ C \cdot (E_\gamma - EA)^{L+1/2}, & E_\gamma \geq EA \end{cases} \quad (2.3)$$

with L representing the angular momentum of the photoelectron and C some proportionality constant. If the EA of the isotope of interest is higher than that of the isobar, a selective neutralization of the isobaric background can be achieved. This is realized by collinearly overlapping the ion beam with an intense laser of a suitable wavelength. Unwanted isobars can thus be neutralized, unable to pass a mass spectrometer, while the isotope of interest stays negatively charged [23]. A graphical representation of such a setting is given and described in Figure 2.1.

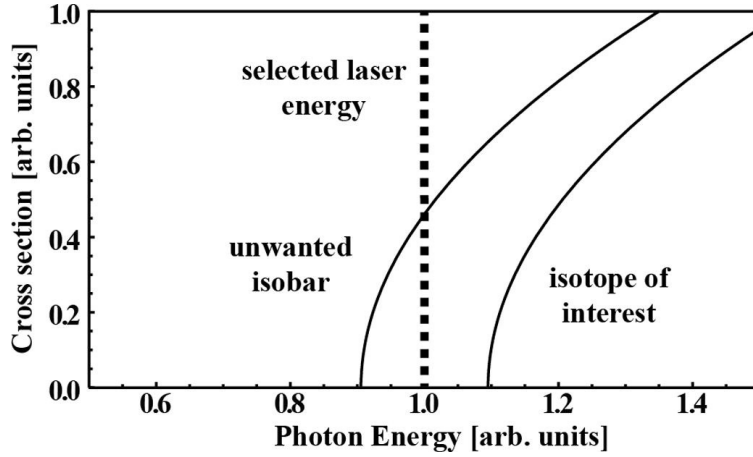


Figure 2.1: Photo detachment cross section of an isotope of interest and its unwanted isobar (solid lines). When choosing a laser with suitable photon energy (dashed line, Equation 2.2), only the unwanted isobar can be photo detached while the isotope of interest stays unaffected (taken from [23]).

Requirements regarding the respective EA of the trace isotope and its isobar are not always matched. In such cases, one can exploit an injection in molecular form. For example, the EA of ^{60}Fe is lower than that of its stable isobar ^{60}Ni . However when considering their corresponding single hydrides ^{60}FeH and ^{60}NiH , the opposite is the case [21].

For an initial number of ions N_0 , a photo detachment cross section σ , a given photon flux ϕ and an interaction time t , the number of remaining ions is given by

$$N(t) = N_0 \cdot e^{-\sigma\phi t} \quad (2.4)$$

The method of selective laser photo detachment in AMS was initially suggested and described more than 30 years ago by Berkovits et al. 1989 [24]. However, cross sections for photo detachment are rather small, typically several 10^{-17} cm^2 . Therefore, effective depletion using conventional lasers requires an interaction time of at least several hundreds of μs , which had not been feasible until the development of an ion cooler for anions [14, 25].

2.2 The ILIAMS RFQ negative ion cooler

The Ion Laser Interaction Mass Spectrometry (ILIAMS) setup is located at the low energy side after the second ion source at VERA. It allows for isobaric separation of ion beams via photo detachment. Following about five years of development and modification on an independent test bench decoupled from the accelerator, it was installed as part of a new injection beamline in 2015 [14]. After extraction from the source, the 30 keV negative ion beam is m/q separated by a double focusing 90° bending magnet and injected into the cooler section. Therein it undergoes electrostatic deceleration down to some tenths of eV, before entering the radio frequency quadrupole (RFQ) negative ion cooler. In the RFQ cooler it is further decelerated (cooled down)

by interacting with the buffer gas and overlapped with a laser for photo detachment. After exiting the cooler section, the ion beam, ideally cleared from isobaric background, is re-accelerated up to 30 keV. Following a set of 45° spherical electrostatic analyzers [14], the ions are passed on to the regular AMS injector. A schematic drawing of the ILIAMS setup is shown in Figure 2.2. Figure 1.1 also highlights the ILIAMS beamline as part of the entire VERA facility.

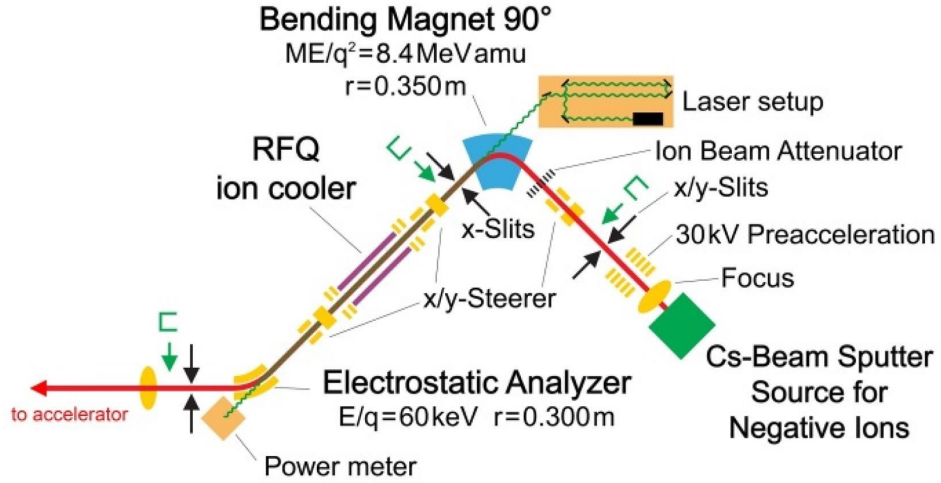


Figure 2.2: Schematic of the ILIAMS beamline at the VERA facility and its components. Shown in red is the direction of the negative ion beam, the laser is drawn in green (adapted from [14]).

The key component of ILIAMS is the RFQ negative ion cooler, which serves the purpose of increasing interaction times between laser and ion beam into the range of several milliseconds. Doing so, the RFQ cooler enables almost total detachment of isobaric background during ion transition. Experiments performed by Johannes Lachner et al. 2019/21 [12, 13] on negatively charged ^{36}Cl and ^{26}AlO ions yielded suppression factors of $f_{sup} > 10^{11}$ for the respective isobars ^{36}S and ^{26}MgO .

The RFQ cooler consists of a 950 mm long quadrupole (linear Paul trap) keeping the ion beam on the laser axis. Four rectangular guiding electrodes create a linear drift field of a few volts per meter to direct the ion beam towards the exit. The electrodes are held in position by six ceramic spacers, equally distributed and aligned within a cylindrical cooler tube of 960 mm length and an inner diameter of 40 mm. The cooler tube is filled with a low pressure buffer gas (typically pure He) to obtain near-thermal ion energies via collisions. The entire tube is located inside a casing connected to a differential pumping system consisting of three turbomolecular pumps. These ensure ultra high vacuum around the cooler unit to avoid collisional beam losses outside the quadrupole. To improve pumping efficiency and minimize buffer gas leakage into the neighbouring beam section, pumping holes have been added at the ends of the cooler tube. Their size can be controlled via movable sleeves, (see section 2.2). The entire setup is placed inside a safety cage that can be set to a cooler high voltage (HV) potential of up to -30 keV to facilitate electrostatic deceleration and re-acceleration of the beam [14, 23, 26]. A schematic drawing of the cooler section is shown in figure 2.3(a).

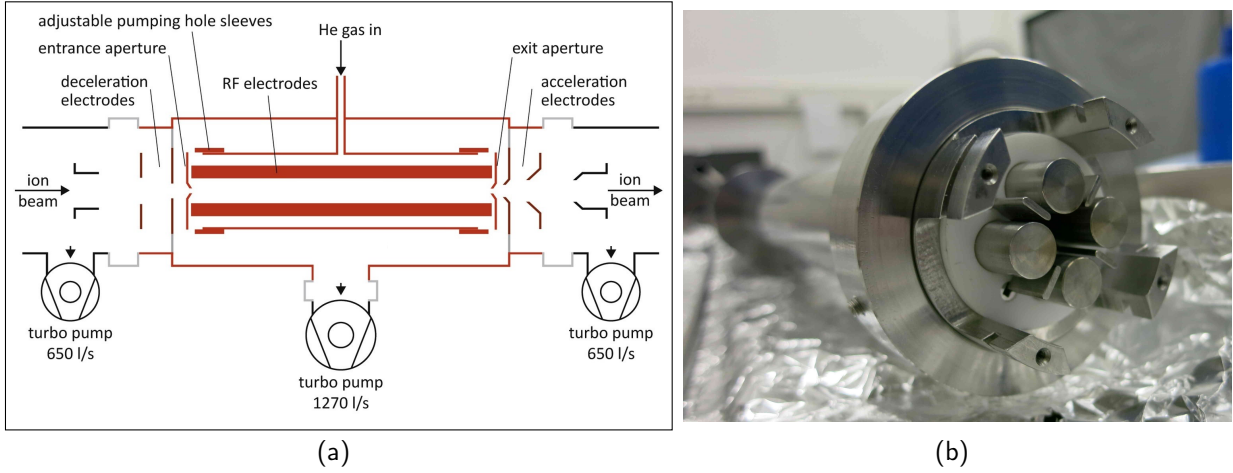


Figure 2.3: (a) Schematic of the RFQ ion cooler section (adapted from [26]) and the different adjusted potentials: cooler HV-ground potential (red), deceleration and re-acceleration lens potentials (dark red), lab ground (black) and insulators (gray); locations of the pumping components as well as their respective pumping strength; INJHI and INJLO lenses are given as deceleration electrodes, EXTLO and EXTHI as acceleration electrodes. (b) Structure of the RFQ ion cooler (taken from [26]) seen at its entrance. Also shown are the additional pumping holes and the corresponding sleeve. One of the ceramic spacers is visible directly at the entrance.

2.2.1 Linear Paul trap

A Paul trap can capture charged particles within a confined region by the combination of a periodic and time dependent as well as a static electric field, generated by electrodes of a certain geometry. By choosing suitable frequencies and voltage amplitudes, it can function as a mass filter. The linear case consists of four parallel hyperbolic electrodes, whose field forces ions towards their enclosed axis on which they can move along unaffectedly. The linear Paul trap therefore is a two dimensional ion trap. For an applied static voltage U_0 and an amplitude of the RF-voltage V_{rf} , the time dependent radial potential ϕ on the (x, y) plane orthogonal to the beam axis yields [27]

$$\phi(r, t) = \frac{\phi_0(t)r}{r_0^2} \quad (2.5)$$

$$\phi_0(t) = U_0 + V_{rf} \cdot \cos(\omega t)$$

where r_0 represents the shortest distance from the axis to the electrodes. Only an approximation of the quadrupole field can be made for Paul traps like the RFQ cooler, which uses cylindrical RF-electrodes of radius r inside a grounded tube. A ratio of r_{rf}/r_0 has to be chosen in order to gain the best approximation, as described by Denison et al. 1971 [25]. The electrode structure of the ILIAMS RFQ cooler measures $r_{rf} = 5$ mm and $r_0 = 4.37$ mm, yielding a slightly different ratio of 1.144. Because the RFQ negative ion cooler is just used as an ion guide, mass filtering and therefore static field contribution is not necessary ($U_0 = 0$). The governing equations of motion, called *Mathieu equations*, therefore reduce to Equation 2.6

$$\begin{aligned} \frac{\partial^2 u_i}{\partial t^2} - 2q_M \omega t &= 0 \\ q_{M,x} &= -q_{M,y} = -\frac{4QV_{rf}}{mr_0^2\omega} \end{aligned} \quad (2.6)$$

With $u_i = (x, y)$ and ω being the angular frequency of the periodic RF-field. The dimensionless Mathieu parameter q_M represents the correlation between ion characteristics (charge Q and mass m) and operation parameters of the quadrupole [27]. Stable solutions for Equation 2.6 exist for all $|q_M| < 0.908$ [27], although experiments on the Ion Laser Interaction Setup (ILIAS) test bench showed that most stable ion currents could be reached in the range of $q_M \approx 0.5$ for the RFQ cooler [26]. Experiments used for comparison in this thesis were realized at $q_M = 0.42$ [14]. Within this region ($q_M \lesssim 0.4$), the average trapping field can be approximated by a radial pseudo-potential-well of average potential depth D [28]:

$$\phi_{rf}(r) = D \frac{r_0^2}{r^2} = \frac{q_M V_{rf}}{4} \cdot \frac{r_0^2}{r^2} \quad (2.7)$$

2.2.2 Guiding field

As mentioned, the linear Paul trap only confines ions perpendicular to the beam axis. To ensure that trapped and cooled ions exit the cooler at the intended side, an additional static guiding field has to be applied. This is realized by four so called *guiding electrodes*, 1.5 mm thick plates with rounded edges. These are placed symmetrically between the RF-electrodes. A tilting angle in beam direction of 0.036° with respect to the beam axis results in an axial field gradient along the cooler of a few volts per meter [26]. This linear drift field forces a preferred direction towards the extraction side onto the trapped negative ions.

2.2.3 Injection/Extraction

In order to avoid losses due to collisional neutralization or scattering, the negative ion beam has to be decelerated to some 10 eV before injection into the cooler. On the one hand, this is realized by setting the ground potential of the cooler section a few volts above cathode potential of the ion source. On the other hand, two additional flat aperture lenses provide a smooth field gradient for deceleration, while mitigating beam expansion. The outer „high“ lens (INJHI) has an inner diameter of 20 mm, the opening of the inner „low“ lens (INJLO) measures 10 mm. Their counterparts on the extraction side (EXTLO, EXTHI) are of conical shape to provide additional beam focusing during re-acceleration. The low lenses are fully enclosed by insulators connected to the beamline tube, thus limiting buffer gas leakage out of the cooler area into the neighbouring beamline [26].

Furthermore, two conical apertures set to ground potential (cooler-HV) are placed with their tips towards the respective endings of the ion trap. They shield the decelerated ion beam from the

deflecting fringe fields of the RF and guiding electrodes. Due to their form, an additional beam focusing effect is obtained. All mentioned components are highlighted in color in figure 2.3(a) according to their corresponding applied potential [26].

2.3 Buffer gas cooling of negative ions

The technique of buffer gas cooling for ion beams is widely used [29]. Trapped ions are surrounded by a low pressure neutral buffer gas (usually He) and lose their kinetic energy via frequent collisions with the gas particles. After a sufficient number of collisions, the ions approach the equilibrium conditions of the buffer gas, they thermalize. In the case of a linear Paul trap, cooled ions accumulate at the trapping center [27], a welcomed effect at the ILIAMS beamline, as the laser is aligned at the beam axis.

However, when dealing with negative ions, the choice of the injection energy is of great influence. Because of their usually low electron affinities (EA) they can easily be neutralized by collisional detachment. To avoid such beam losses, the center of mass (CM) energy of the collision should not surpass the EA of the anion [28]. The CM energy is given by

$$E_{CM} = E_i \cdot \frac{m_g}{m_i + m_g} \quad (2.8)$$

where m_i and m_g are the respective masses of the ion and the buffer gas particle and E_i denotes the ion's kinetic energy in the lab frame. Equation 2.8 describes the maximum energy that can be transferred in a single collision. It shows the benefit of using a light buffer gas like He, as the maximum collisional energy transfer remains rather small compared to heavier gases like N₂ or Ar. Avoiding beam losses due to collisional detachment is the main reason the negative ions are electrostatically decelerated before entering the RFQ cooler at ILIAMS.

For ⁶³Cu⁻ (used for simulations, see subsection 3.6.3) the EA , i.e. the binding energy of the surplus electron is for example 1.23578(4) eV [30]. This corresponds to a maximum injection energy of about 20.7 eV in the lab frame. Still, experiments performed on the ILIAS test bench yielded highest transmission through the cooler at around 50 eV [26]. Also, for several other ions in the same study, the optimum injection energy has been found at collisional energies in the CM roughly twice the EA . However, a general rule concerning this matter cannot be given [26].

To accurately simulate particle trajectories inside the RFQ ion cooler, a suitable model to account for collisions between ions and buffer gas needs to be applied. The COMSOL particle tracing module comes with a variety of options depending on the requirements. The first intention had been to apply the theory of nuclear and electronic stopping, describing the energy dependent drag of charged particles flying through matter. It is based on the work and calculations published by Ziegler et al. 1985 [31], whose simulation software *SRIM* and *TRIM* are widely used to calculate

material specific ion transport properties. However, accuracy of a default model implemented in COMSOL breaks down below kinetic energies of 8.6 eV [31]. For comparison, thermal energies of the buffer gas inside the RFQ cooler reside at $E_{th} = 3/2k_B T \approx 39$ meV at a temperature of 300 K.

As a consequence, an elastic scattering model has been chosen to calculate interactions between ions and buffer gas. The elastic collision node implemented in COMSOL's particle tracing interface is based on the theory of the hard sphere (HS) elastic collision model. Due to its simple structure, this model is known to be rather imprecise. A much more realistic model is the so called variable hard sphere (VHS) model, which is an extension of the HS-model [32, 33]. In order to accurately model trajectories of ions passing the RFQ cooler, this extension has been implemented into COMSOL manually. A short summary of the mathematical foundations for both models is given in the following sections.

2.3.1 Hard sphere (HS) model

The *hard sphere (HS)-model* is an oversimplified collision theory that is widely used in combination with the direct simulation Monte Carlo method, which allows for fast computation and easy implementation. Particles are approximated as simple elastic hard spheres with specific diameter d_i . A collision occurs at points in space and time where the distance of two trajectories becomes small enough for their spheres to overlap. The total collisional cross section between two species 1 and 2 can therefore simply be written as a constant function dependent on the respective particle diameters d_1 and d_2 [33]:

$$\begin{aligned}\sigma_{HS,12} &= \pi d_{12}^2 \\ d_{12} &= \frac{(d_1 + d_2)}{2}\end{aligned}\tag{2.9}$$

Effective HS-diameters for different atoms and molecules can be obtained experimentally by measuring mean free path and mean velocity at fixed temperature. Considering two particles with respective masses m_i and velocities v_i , an elastic collision per definition requires conservation of total energy and momentum

$$\begin{aligned}E &= \sum_{i=1}^2 E_i = \sum_{i=1}^2 \frac{m_i v_i^2}{2} = \sum_{i=1}^2 \frac{m_i v_i'^2}{2} \\ P &= \sum_{i=1}^2 p_i = \sum_{i=1}^2 m_i v_i = \sum_{i=1}^2 m_i v_i' = m_i v_m \\ v_m &= \frac{\sum_{i=1}^2 m_i v_i}{\sum_{i=1}^2 m_i}\end{aligned}\tag{2.10}$$

where „'“ indicates post collisional variables and v_m represents the center of mass velocity. A geometrical visualization of such an interaction is shown in Figure 2.4.

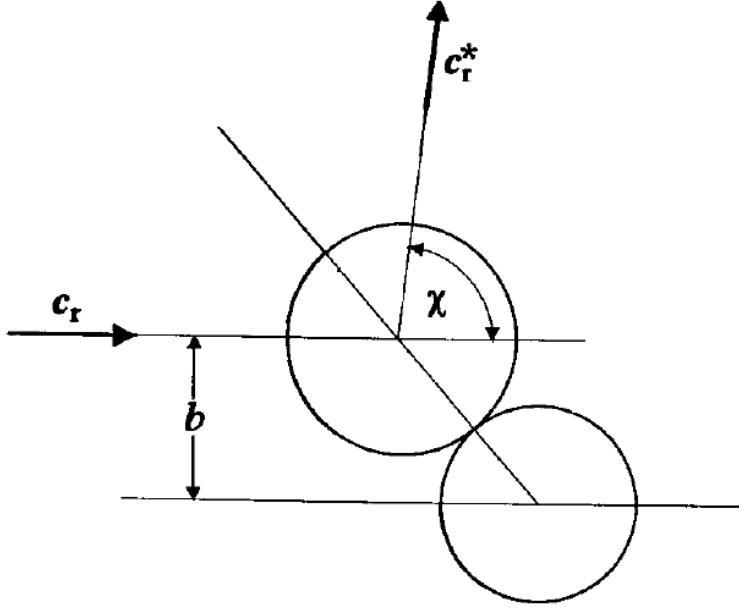


Figure 2.4: Center of mass collision geometry of two hard sphere particles in the plane of pre- and post collision relative velocities c_r and c_r^* (adapted from [33]); b and χ are called impact parameters.

From Equation 2.10 we obtain the post collision velocities as

$$\begin{aligned} v'_1 &= v_m + \frac{m_2}{m_1 + m_2} v'_r \\ v'_2 &= v_m - \frac{m_1}{m_1 + m_2} v'_r \\ v'_r &= v'_1 - v'_2 \end{aligned} \quad (2.11)$$

where v'_r denotes the relative velocity of the collision particles after collision, whose directions are determined by the so called *impact parameters*, namely the distance of closest approach b and the deflection angle χ , as can be seen in Figure 2.4. In the case of hard sphere particles the impact parameters correlate according to

$$\cos \frac{\chi}{2} = \frac{b}{d_{12}} \Rightarrow \cos \chi = 2 \left(\frac{b}{d_{12}} \right)^2 - 1 \quad (2.12)$$

Equation 2.9 and Equation 2.12 show that the nature of scattering is isotropic in the center of mass frame for hard sphere particles, meaning that all directions of v'_r are equally likely.

2.3.2 Variable hard sphere (VHS) model

The utilization of a collision cross section that is totally independent from the translational energy ($E_t = m_r v_r/2$, where m_r is the reduced mass) available during scattering can cause inaccuracies

between HS-simulations and experimental data by a large margin. For real gases, the scattering cross section decreases as v_r and E_t increase, which originates from the temperature dependence of the coefficient of viscosity (μ). The *variable hard sphere (VHS)-model*, developed in 1981 by Bird [33], introduced a velocity dependent scattering cross section based on experimentally gained reference parameters, that follows an inverse power law, given as

$$\frac{\sigma_{VHS}}{\sigma_{ref}} = \left(\frac{d_{VHS}}{d_{ref}} \right)^2 = \left(\frac{v_{r,ref}}{v_r} \right)^{2\omega_{ref}-1} \quad (2.13)$$

where the subscript „*ref*“ denotes experimental reference values. For a VHS particle species at temperature T_{ref} , that has a viscosity μ_{ref} with a corresponding temperature coefficient ω_{ref} , the respective average relative velocity v_{ref} and reference diameter d_{ref} can be calculated as

$$v_{ref}^{2\omega_{ref}-1} = \frac{\left(\frac{2k_B T_{ref}}{m_r} \right)^{\omega_{ref}-1/2}}{\Gamma\left(\frac{5}{2} - \omega_{ref}\right)} \quad (2.14)$$

and

$$d_{ref} = \sqrt{\frac{15\sqrt{\frac{m_r k_B T}{\pi}}}{2(5 - 2\omega_{ref})(7 - 2\omega_{ref})\mu_{ref}}} \quad (2.15)$$

respectively, where $\Gamma(x) = \int_0^\infty s^{x-1} e^{-s} ds$ is known as gamma function [33] and m_r denotes the reduced mass, given by

$$m_r = \frac{m_1 \cdot m_2}{m_1 + m_2} \quad (2.16)$$

Although the cross section is not constant anymore, the scattering angle still behaves according to Equation 2.12 and is therefore uniformly distributed across all possible angles within the center of mass frame.

2.4 Rarefied gas flow

In 1909, Martin Knudsen encountered derivations from Poiseuille's law [34] when measuring the flow rate through a tube at low pressure. He stated that below a certain pressure a gas does not behave as a medium anymore, but rather its particles move independently from each other, which he described as rarefied gas flow. Within this regime, the mean free path of the gas particles becomes sufficiently large such that particle-particle collisions are negligible with respect to interactions at surrounding surfaces [35].

2.4.1 Knudsen criterion

The crucial parameter to determine the present flow regime is the dimensionless *Knudsen number* Kn , defined as the ratio of the mean free path λ of the gas particles and the characteristic length L of the geometry they are restricted to [36].

$$Kn = \frac{\lambda}{L} = \frac{1}{\delta} \quad (2.17)$$

Often the *rarefaction parameter* δ is used instead for practical reasons. The mean free path for a simple gas with mass m and viscosity μ is given by

$$\begin{aligned} \lambda &= \frac{\mu \bar{v}}{\rho k_B T} \\ \bar{v} &= \sqrt{\frac{2k_B T}{m}} \end{aligned} \quad (2.18)$$

where $\bar{v}$ is the most probable molecular speed and ρ , k_B and T denote the number density, the Boltzmann constant and the temperature respectively [37]. Although the change from ordinary continuum gas dynamics to rarefied flow is of gradual nature, a classification for flow regimes dependent on the Knudsen number can be made according to Chambre et al. 1961 [38] and Karniadakis, Beskok and Aluru 2005 [36]:

- Continuum flow ($Kn < 0.01$)
- Slip flow ($0.01 < Kn < 0.1$)
- Transitional flow ($0.1 < Kn < 10$)
- Free molecular flow ($Kn > 10$)

While the continuum flow regime describes a fully developed medium where Navier Stokes equations can be applied, a statistical approach summing over single particle fluxes is required for the free molecular flow. The slip flow and transitional flow regime are transition regions and rather extensive in computation and simulation because they need to consider both cases. A demonstrative plot of the main flow regimes as a result of pressure and characteristic length with correspondent Kn is given in Figure 2.5.

As a consequence of the strong pumping units, the gas flow inside the RFQ ion cooler section was expected to completely remain within the free molecular respectively the transitional flow regime. COMSOL offers a Molecular Flow package sufficient to solve both systems [20]. However, early attempts to simulate transitional flows soon revealed excessive demands of computation power and RAM in the case of more detailed geometries. Even for clusters accessible at larger research facilities such as CERN, comparable models had to be restricted to the two dimensional case [39]. All gas simulations shown within this master's thesis therefore have been performed using the free molecular flow interface.

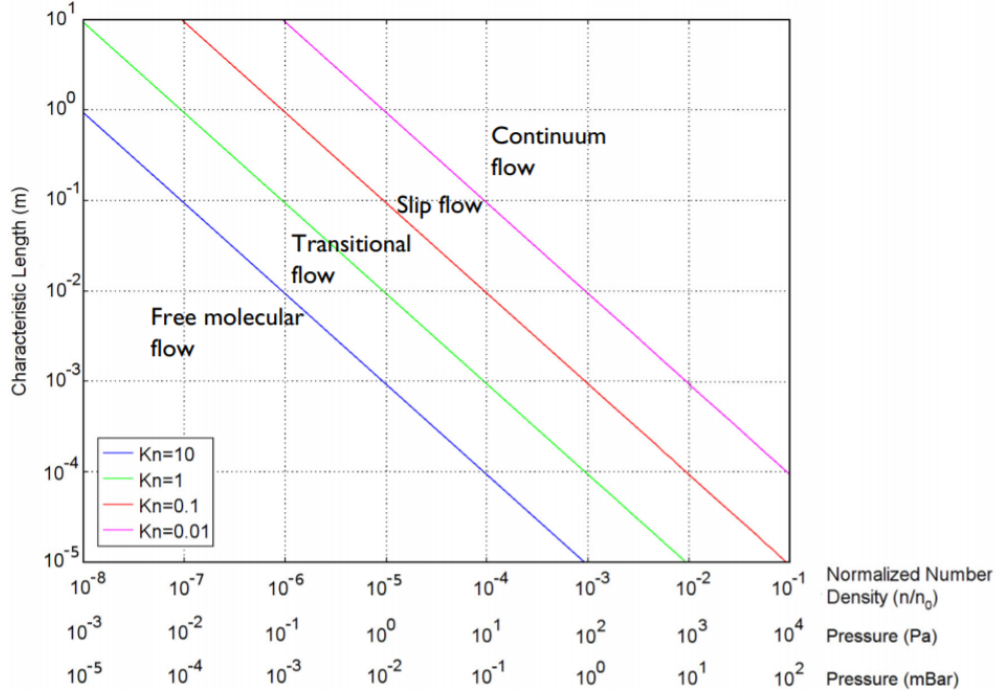


Figure 2.5: Example of the main flow regimes for gas flows (taken from [20]). The colored lines represent constant Knudsen numbers. The number density of the gas is normalized to that of an ideal gas at standard conditions, the equivalent pressure is given in Pa and mbar. Although the lines indicate an instantaneous change of the flow regimes, transition regions are present.

2.4.2 Free molecular flow

In the continuum regime, particle momentum near a channel wall is reduced with respect to the center because it is transferred by molecular collisions, resulting in a parabolic momentum distribution in flow direction. For the free molecular flow regime, the lack of particle-particle collisions prevents momentum transfer leading to a uniform distribution over the whole channel diameter. When Knudsen first derived the theory of free molecular gas flow, he made a fundamental assumption for the scattering of gas particles at surfaces under free molecular conditions, known as *Knudsen's cosine law of scattering*. It states that the probability ds for a scattered particle leaving the surface at solid angle $d\omega$ and an angle θ with respect to the surface normal is described by

$$ds = d\omega \frac{\cos \theta}{\pi}$$

$$\rho(\theta) = \frac{ds}{d\omega} = \frac{\cos \theta}{\pi} \quad (2.19)$$

implying no contribution from the direction of incidence. Here, $\rho(\theta)$ is the corresponding polar distribution, while the azimuthal distribution is uniform over all angles of $\phi \in [0, \pi]$. Although he himself had no feasible explanation at that time, his experiments were in very good agreement [40]. Later he could show that this law is a direct consequence of the second law of thermodynamics.

2.5 Space charge contribution

As indicated in Section 1.3, space charge effects emerging from the decelerated ion beam inside the cooler are assumed to have a large effect on transmission through and the ion residence time within the cooler. Experiments with $^{63}\text{Cu}^-$ have shown a drastic decrease in transmission when operating at beam currents in the 100 nA range. While ion residence times of 8.2 ms have been measured for low injected beam currents in the pA range, where space charge becomes negligible, they decrease below 1 ms for beam currents on the order of several hundred nA [26]. An illustration of this effect is given in the results section in Subfigure 3.17(a). Charged particles interact with each other electrostatically. The scalar force between two point-like sources with respective charges q_1 and q_2 separated by the distance r is described by Coulomb's law

$$F = \frac{1}{4\pi\epsilon_0} \frac{q_1 q_2}{r^2} \quad (2.20)$$

where ϵ_0 denotes the permittivity of vacuum. A single ion can be considered as such a distinct point-like charge source. However, a high ion density as present in the case of ion beams can be treated as a charge distributed over a region of space, the space charge. Such a space charge density ρ_{SC} generates an electric potential Φ_{SC} , satisfying Poisson's equation (Equation 3.9). Assuming static conditions of present electric fields and beam current, the generated space charge potential is of static nature. The total potential Φ acting on a charged particle can then be described as the sum of some present static potential Φ_0 and the space charge potential Φ_{SC} [41]. The accumulated space charge inside gas filled ion guides is an important factor, especially when performing experiments at higher beam currents. Based on theoretical calculations by Guan and Marshall 1993 [41], Guo et al. 2014 [42] introduced a computational model approach for the simulation of space charge effects, including results for linear Paul traps such as the RFQ negative ion cooler. For Mathieu parameters $q_M < 0.4$ the radial ion density distribution can be approximated as a Gaussian distribution

$$n(r) = \frac{\rho_l}{2a^2\pi} \exp\left(-\frac{r^2}{2a^2}\right) \quad (2.21)$$

where r represents the distance to the beam axis and ρ_l denotes the line density given by the ratio of total ion number and effective length of the Paul trap. The parameter a is the standard deviation and is given by [42]

$$a = \frac{r_0}{2} \sqrt{\frac{4k_B T \epsilon_0 + q^2 \rho_l}{2\pi q \epsilon_0 D}} \quad (2.22)$$

Here r_0 and q are the distance between beam axis and RF-electrodes and the ion charge respectively, D is given by Equation 2.5.

3 | Methods and Results

3.1 COMSOL Multiphysics

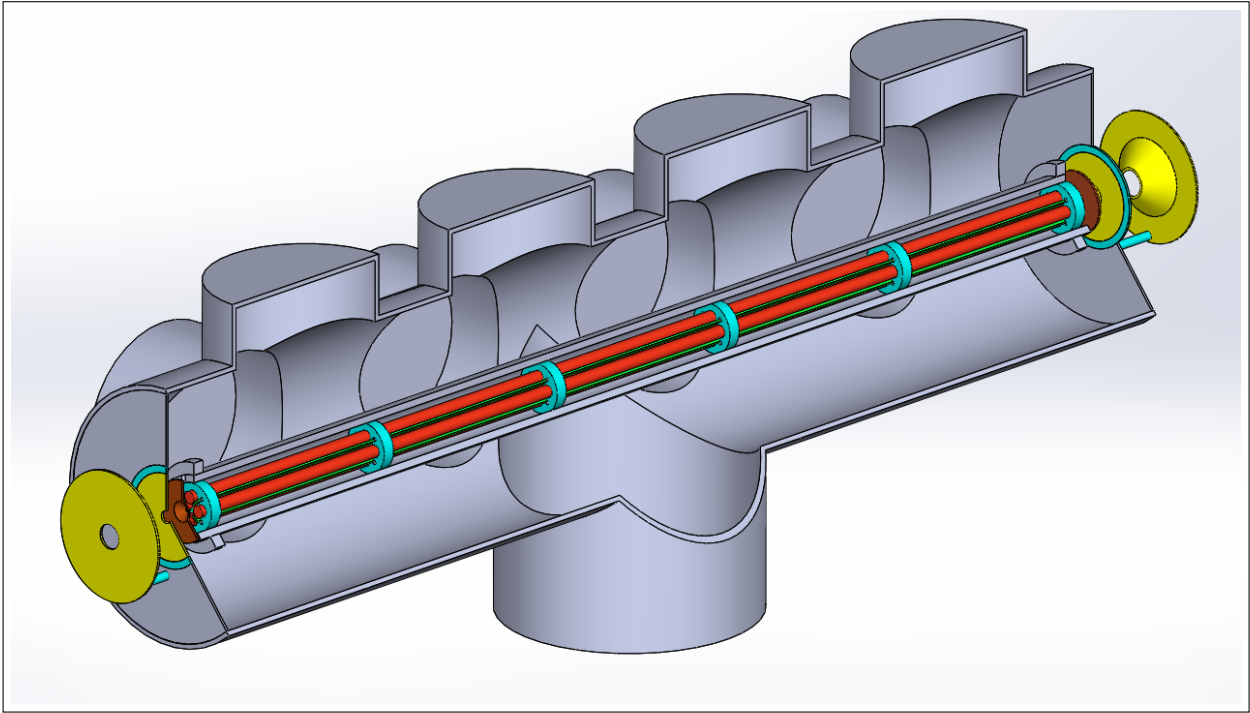
COMSOL is a cross-platform finite element analysis solver and multiphysics simulation software that covers all necessary steps of common modeling flows. It includes tools to create geometries using the integrated CAD functionality and mesh them with respect to the used fields of physics as well as precision requirements. The graphical user interface (GUI) allows for easy application of boundary conditions depending on the specified physics modules, while a variety of customized solvers are accessible to solve for the physical systems as needed. Result data can be post processed numerically and graphically directly within the working environment. Although any of these steps can be performed using the GUI, COMSOL also allows for LiveLink application with numerical programming software like MATLAB as well as with CAD software such as SolidWorks or AutoCAD [19].

However, the greatest benefit of using COMSOL certainly is the possibility to combine a large number of different physics interfaces sharing dependent variables within a single simulation. This allows for the calculation of bidirectional models, as well as the preprocessing of initial conditions in a rather simple way [19]. A reliable simulation model of the RFQ negative ion cooler needs to combine accurate calculations of the buffer gas dynamics and present electrical fields, as well as their respective influence on ion trajectories. These are requirements that are conveniently matched by COMSOL.

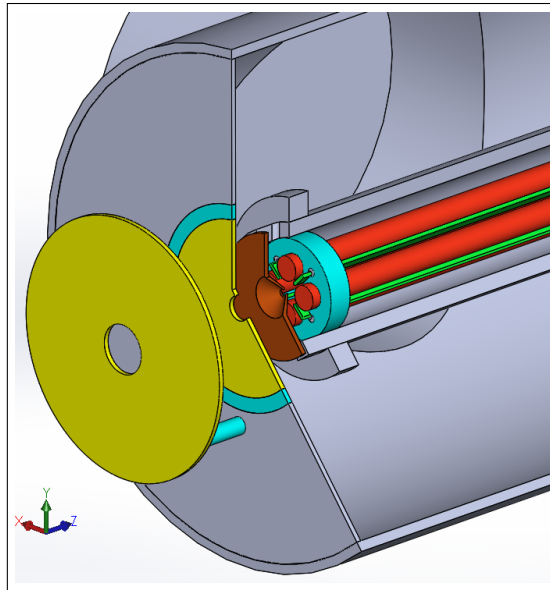
Packages used in this master's thesis were the *Molecular Flow Module* [20] to generate stationary solutions of buffer gas dynamics, the *AC/DC Module* [43] for applying proper stationary and time dependent electrostatic fields and the *Particle Tracing Module* [32] to calculate trajectories of charged particles affected by these fields and interacting with the buffer gas. To allow for direct fitting of result data, also the *Optimization Module* [44] has been utilized on occasion.

All simulations presented within the frame of this master's thesis have been performed using the simulation software COMSOL Multiphysics® (Version 5.6). Theoretical information such as used equations or solver configurations are provided.

3.2 3D-model of the RFQ-negative ion cooler



(a)



(b)

Figure 3.1: 3D model of the total cooler section generated with SOLIDWORKS [45]. A sectional view is given for better visualization. (a) shows the total model of the cooler; (b) illustrates the injection side for a more detailed view of included components.

Casing and RFQ-cooler tube are given in gray, important components are highlighted in color: RF-electrodes (red), guiding electrodes (green), apertures (brown), high and low lenses (yellow) ceramic spacers and insulation material (turquoise). Attached with insulated rods on both sides are the injection and extraction high lenses (INJHI, EXTHI) not included in any COMSOL simulation.

At the very beginning, a 3D model of the RFQ negative ion cooler had to be created to serve as simulation geometry. Dimensions and distances for all necessary cooler components have been collected from sketches dating back to the development of ILIAS, as well as from former works that included modifications of the cooler geometry [15, 16]. Unfortunately no documentation regarding the dimensions of the pumping holes located at the respective ends of the cooler tube has been found. Those have been enlarged previous to installation at VERA, as described in [15]. An estimation has been made on the basis of photographic material keeping record of the latest opening.

A first detailed model has been generated using the CAD-software SOLIDWORKS [45]. This model also includes parts that are non essential within the context of simulation, for the simple purpose of future presentations. The result is presented in Figure 3.1. Subfigure 3.1(a) shows the whole model of the RFQ cooler, while Subfigure 3.1(b) focuses on the injection side for a better impression of included components.

Although COMSOL supports CAD-import for various formats, independent models have been constructed later on using the internal CAD-tools. Those have been used solely to perform COMSOL-simulations and have been modified according to the respective computational requirements in order to enable/optimize simulation performance and improve output quality [19]. Adaptions performed are explained in place.

3.3 Buffer gas flow determination

Although former works have attempted to simulate ion behavior inside the RFQ cooler, incomplete knowledge of the actual buffer gas pressure inside the cooler tube has been a source of great uncertainty. An InstruTech® CVG101 Worker Bee™ convection-enhanced Pirani vacuum gauge sensor, located outside the cooler section, gives information about the pressure at the gas inlet into the cooler [15, 16]. This pressure can be varied by a valve placed before this section. Operation parameters are in the region of some tenths of mbar, depending on the buffer gas species (He, N₂ and Ar). On the other side of the inlet valve, an operational pressure of 2 bar is applied. Figure 3.2 shows the location of inlet valve and inlet pressure measurement.

Additionally, three separate ATMION® Wide Range vacuum gauge pressure devices located next to the vacuum pumps provide information about the outgoing pressure. However, no information about the pressure distribution within the cooler tube is known. A practical experiment to link the displayed inlet pressure on the outside with the actual mass flow rate into the cooler was carried out to allow for appropriate boundary conditions when simulating buffer gas dynamics. This was realized by measuring the constant pressure decline out of a fixed and predetermined volume over time. The obtained results were taken as initial conditions for all future gas dynamics simulations presented in this thesis.

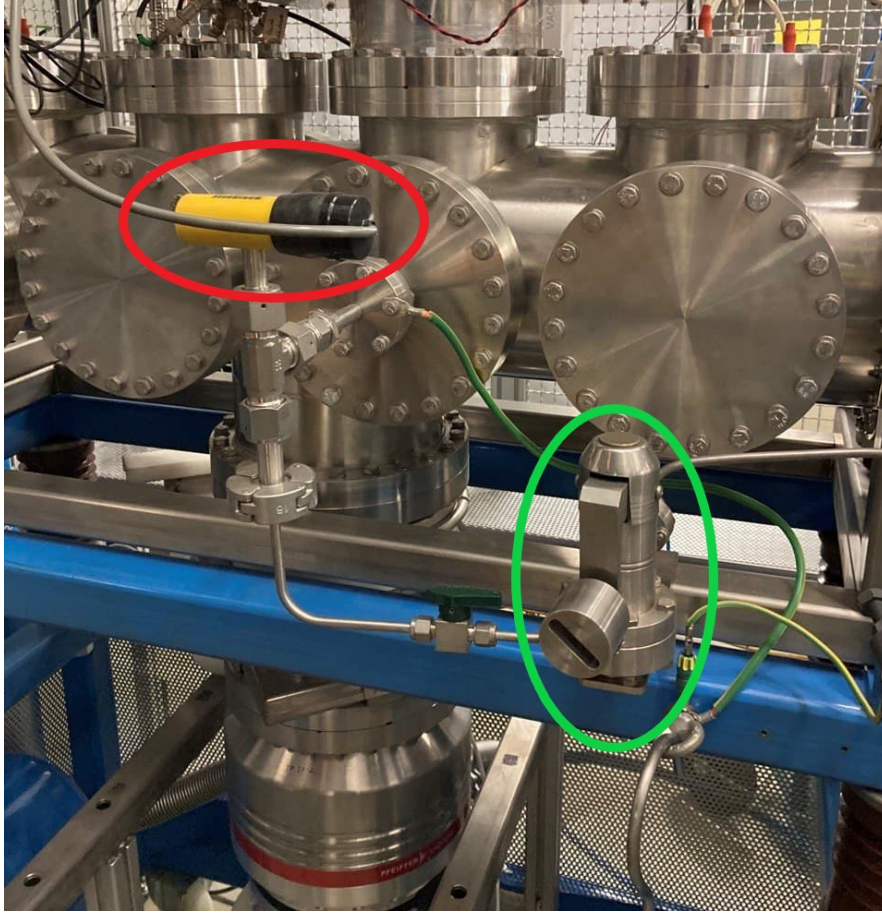


Figure 3.2: Location of the InstruTech[®] CVG101 Worker Bee[™] convection-enhanced Pirani vacuum gauge sensor used to measure the inlet pressure of the buffer gas into the cooler. The pressure gauge and the inlet valve are circled in red and green respectively.

Furthermore, outlet pressures at the vacuum pumps for all currently used gases were measured across the range of inlet pressures. This serves the purpose to compare simulation results to the experimental data and identify deviations from the free molecular flow regime.

3.3.1 Experimental setup

Figure 3.3 shows a schematic drawing of the experimental setup. The pressure device currently used to control the inlet pressure, an InstruTech[®] CVG101 Worker Bee[™] convection-enhanced Pirani vacuum gauge sensor, is highlighted in green and marked as p_{in} and will be referred to as *inlet-gauge* [46]. Shown in blue and connected to the vessel (yellow) is a reservoir serving as gas source. A THYRA CONT VD81 Compact vacuum gauge (green) with digital output independent of the gas specimen and a maximum operation pressure of 1600 mbar was used to measure pressure inside the reservoir (p_{res}). Highlighted in red are the ATMION[®] Wide Range vacuum gauge pressure devices located next to the the vacuum pumps (p_{center} and p_{side}), referred to as *vacuum-gauge* [47].

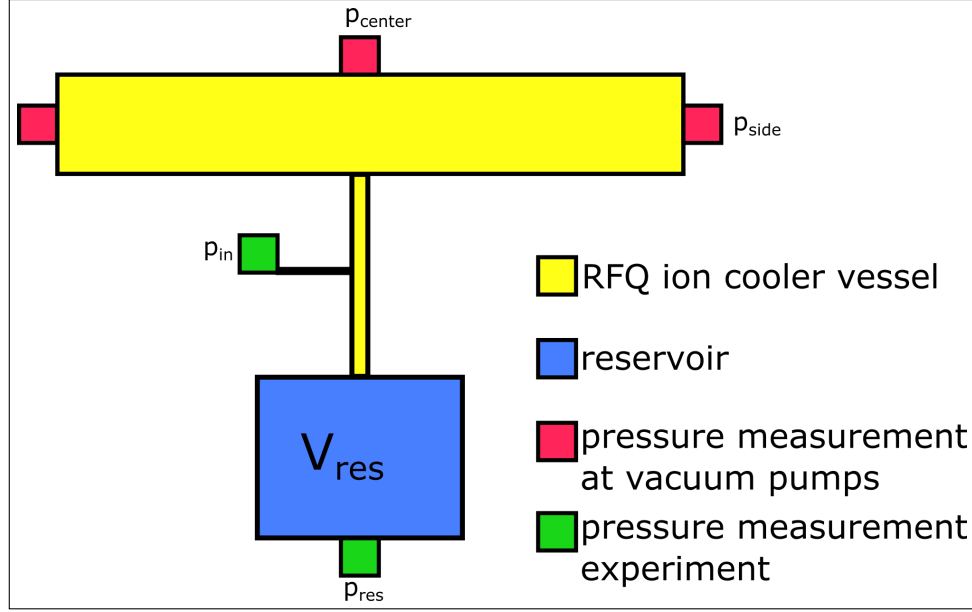


Figure 3.3: Schematic drawing of the experimental setup for the flow measurements into the cooler, not true to scale. The cooler section and the inlet tube are given in yellow, the reservoir connected to the other end of this tube is shown in blue. Highlighted in red are measurement locations for the outlet pressure (p_{center}, p_{side}) placed directly in front of the vacuum pumps. The measurement locations for inlet pressure (p_{in}) and the pressure inside the reservoir with fixed volume (p_{res}) are highlighted in green. Gas flow is directed from the reservoir into the cooler section, from where it is pumped by the vacuum pumps.

3.3.2 Theory

Basis for all calculations is the commonly known ideal gas law

$$pV = Nk_B T \quad (3.1)$$

where p , V , N and T denote the pressure, volume, particle number and temperature respectively. Considering that two parameters of Equation 3.1 remain constant, one can express the remaining as functions of each other. For a fixed volume and temperature, one can therefore calculate the particle flux out of a reservoir via the resulting pressure decline.

$$\Delta N = -(N_f - N_i) = -(p_f - p_i) \frac{V}{k_B T} \quad (3.2)$$

Subscripts „i“ and „f“ denote initial and final values respectively, the negative sign ensures a positive value for a flux pointing outwards the considered system. Similar, if an unknown volume V_x is connected with a defined volume V_{def} without changing the particle number and under constant temperature, V_x writes as:

$$V_x p_i = (V_x + V_{def}) p_f \Rightarrow V_x = \frac{V_{def} \cdot p_f}{p_i - p_f} \quad (3.3)$$

3.3.3 Output correction

While the inlet gauge functions by sensing the heat loss from an electrically conducting wire, the vacuum gauge emits electrons and measures the resulting ionization of the gas atoms/molecules. Consequently their outputs strongly depend on the intrinsic gas properties of heat conductance, convective properties and ionization probability. As both of them are calibrated for the use of N_2 , correction factors have to be applied when measuring different gases. In the case of the vacuum-gauge, a constant conversion factor is given in the corresponding data sheet, while the inlet-gauge user guide states exemplary displayed and real values for different pressures, which were used to interpolate a linear correlation function [46, 47]. The corresponding conversion parameters are summarized in Table A.1 in the appendix and have been used to differ between *displayed* and *real* values. For the inlet-gauge no uncertainty is stated, whereas the vacuum-gauge measures with an accuracy of $\pm 10\%$ for pressures below 0.01 mbar [46, 47].

3.3.4 Implementation and results

Before introducing any gas into the cooler section, the combined volume of the reservoir and connecting tubes was measured. This was done by expanding air from the reservoir into an evacuated balloon of known volume and measuring the corresponding pressure decline. The procedure was carried out two times with gas containers of 1 l and 22.3 l in volume, while the valve leading into the cooler remained closed. Applying Equation 3.3 the fixed volume was determined as $V_{res} = (184 \pm 12) \text{ cm}^3$. The elapsed time to reach a pressure decline of 10 mbar within the reservoir was measured for all gases. As pressure levels were around 1500 mbar, the rate was assumed to be constant within this time. The results, as shown in Table 3.1, were used to correct all results with respect to leakage out of the reservoir.

Table 3.1: Leakage rate out of the reservoir at an operational pressure level of about 1500 mbar.

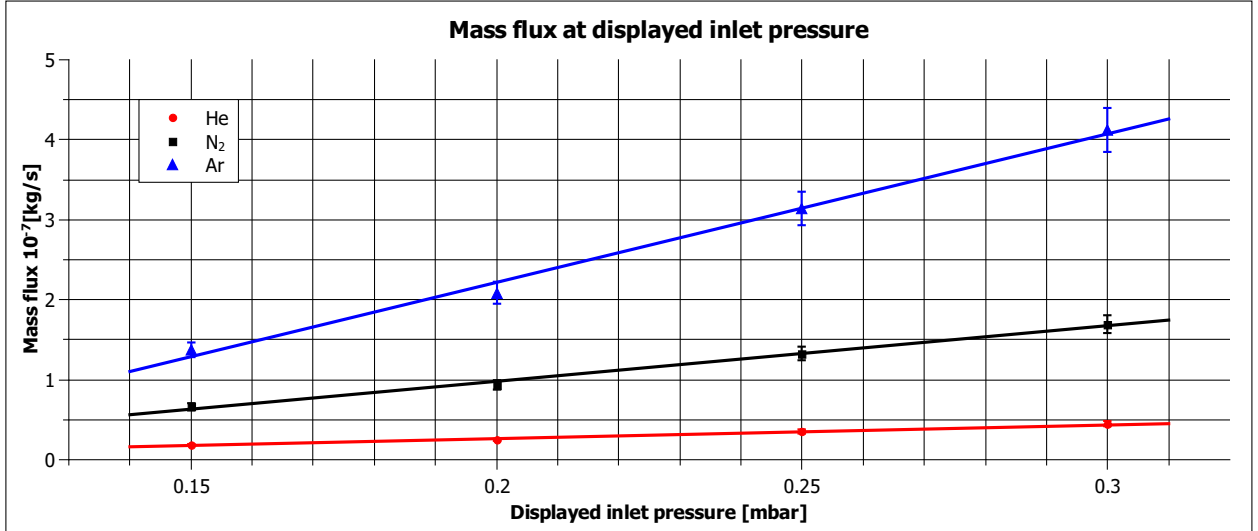
Gas	He	N_2	Ar
Leakage rate [(mbar l)/s]	0.018	0.015	0.013

Afterwards, the reservoir was filled to 1500 mbar. While controlling the displayed inlet pressure by manually adjusting the valve, the time until the pressure decline reached about 50 mbar was measured. This cycle was repeated three times for all three gas species and with four different inlet pressures, all of them within typical operational values. A graphical representation of mass flow rates into the cooler is depicted in Figure 3.4, the corresponding calculated and averaged results are summarized in Table A.2. A linear regression was done for all measurement results to allow for future corrections when performing ILIAMS experiments. The parameters of the linear approximation, providing mass flux values dependent on the displayed inlet pressure, are given in Table 3.2. The corresponding parameters regarding the real inlet pressure can be found in Table A.3.

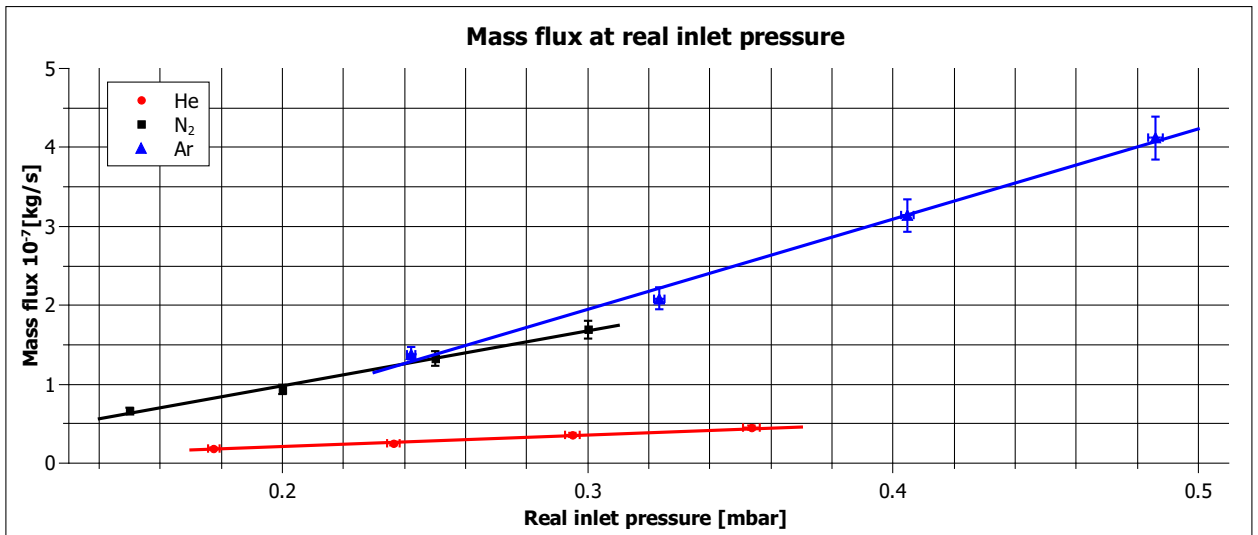
Table 3.2: Parameters for the linear regression function linking the displayed inlet pressure p_{disp} for the respective buffer gas to the corresponding mass flux rate f (in [kg/s]) into the cooler. The function reads: $f = a \cdot p_{disp} + b$

The inlet pressure p_{disp} has to be inserted in mbar.

Gas	a	Δa	b	Δb
He	$1.70 \cdot 10^{-7}$	$0.15 \cdot 10^{-7}$	$-7.67 \cdot 10^{-9}$	$3.41 \cdot 10^{-9}$
N ₂	$6.96 \cdot 10^{-7}$	$0.37 \cdot 10^{-7}$	$-4.12 \cdot 10^{-8}$	$0.85 \cdot 10^{-8}$
Ar	$1.86 \cdot 10^{-6}$	$0.11 \cdot 10^{-6}$	$-1.50 \cdot 10^{-7}$	$0.25 \cdot 10^{-7}$



(a)



(b)

Figure 3.4: Graphical representation of the data summarized in Table A.2. The calculated mass fluxes for He (red), N₂ (black) and Ar (blue) (in kg/s) into the cooler as functions of (a) the displayed inlet pressure, and (b) the real inlet pressure converted according to Table A.1. The data points represent the measured values. Included errors emerge from the uncertainties of the reservoir volume V_{res} , the conversion error of the inlet-gauge and the standard deviation of the three respective measurement cycles. Linear fits according to the data given in Table 3.2 are added as solid lines of matching color.

The mass flux through a geometry strongly depends on the flow regime, thus on the distribution of pressure and particle density, as well as the dimensions of the geometry [37]. The gas flow within the cooler itself is expected to be characteristically rarefied due to the strong vacuum pumping within the cooler vessel. Unfortunately, this is not the case along the inlet geometry connecting the buffer gas reservoir with the cooler vessel. Pressure values on the order of some tenths of mbar in combination with small tube diameters of about 1 mm can lead to different flow conditions prior to entering the cooler. Mathematical predictions based on the theory of rarefied gas dynamics thus cannot be applied to predict the mass flux rates into the cooler, solely by knowing the inlet pressure.

Although a linear fit has been chosen to describe the correlation in Figure 3.4, a function of higher order or exponential nature may be more applicable. Such a nonlinear growth would indicate a divergence from the free molecular flow regime, most probably occurring within the inlet geometry. However, as only four data points have been obtained for each buffer gas, the linear case has been retained to avoid overfitting. Nevertheless, the correlations described in Table 3.2 are crucial to perform predictive simulations on the buffer gas dynamics inside the RFQ cooler, which need to be based on realistic initial conditions.

Independent from the flux determination, the resulting pressure at the vacuum-gauges over the whole operational displayed inlet pressure range was measured for later comparison purposes with COMSOL models. Only one of the two vacuum-gauges at the side of the cooler section was available during this experiment. However, gas flow is expected to be approximately equal at both sides of the cooler. Figure 3.5 shows the results for the displayed center vacuum gauge pressure as a function of the displayed inlet pressure.

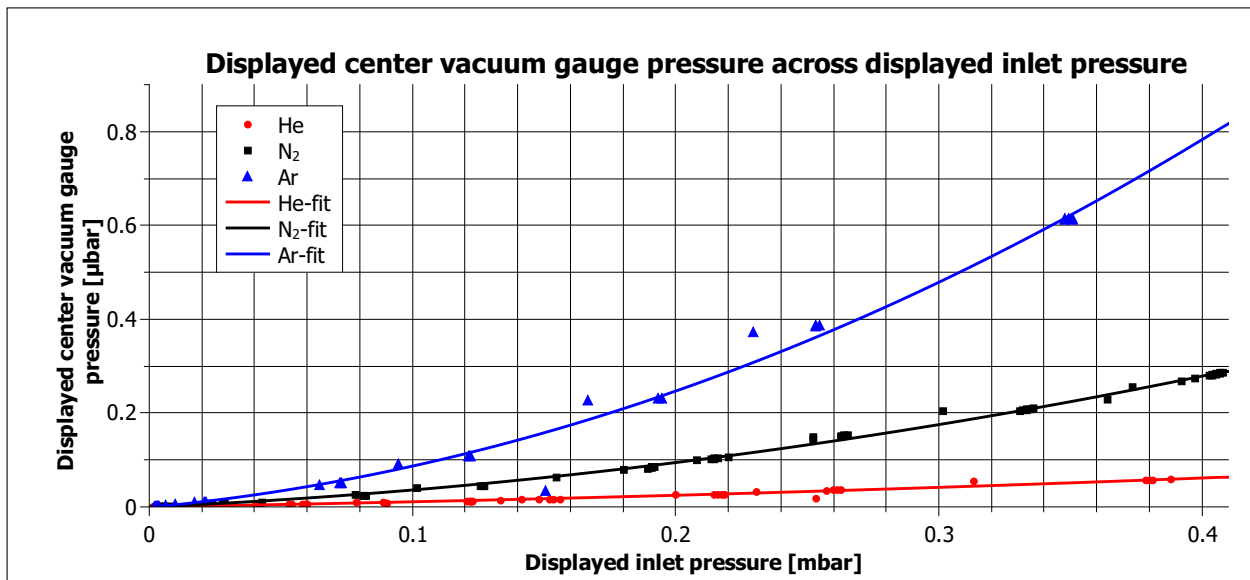


Figure 3.5: Pressure (in μbar) displayed at the center vacuum gauge for He (red), N₂ (black) and Ar (blue) as a function of displayed inlet pressures (in mbar). Polynomial fits of order two are included as solid lines of matching color.

As can be seen, the outlet pressure shows a nonlinear trend with increasing inlet pressure. This again indicates a transition from the free molecular flow regime to the transitional or even continuum flow regime, most probably within the inlet geometry [37]. A second order polynomial has been chosen to fit the data, resulting fit parameters can be found in Table A.4. The obtained data had been of great importance to monitor the accuracy of later generated gas dynamic models, as an accurate simulation should be able to predict outlet pressure values comparable to experimental measurements.

3.4 Buffer gas dynamics simulation

COMSOL simulations using the *Free Molecular Flow Module* periodically apply a direct solver that solves a matrix inhibiting the incoming flux of all surface mesh elements according to Equation 3.6. Results are taken as initial condition for the following cycle until the rate of change of entry values falls below a certain threshold. This relative tolerance in COMSOL is set to 0.1% per default and has not been changed. The computation time and the required RAM increase significantly with model size and mesh quality [20].

In order to obtain precise results with an available RAM of 15 GB, the number of computable mesh elements is restricted to a maximum of about $5 \cdot 10^5$. To still guarantee for high accuracy of results, the modeling geometry has been modified and optimized step by step. By decreasing model size, i.e. by exploiting symmetries and omitting negligible details, the mesh density had been improved constantly. Each new model has been checked on the basis of results such as pressure and particle density with respect to prior models. To verify the reliability of the resulting model, its predictions for the outlet pressure at the vacuum pumps has been compared with the experimental results obtained in subsection 3.3.4 and shown in Figure 3.5.

3.4.1 Underlying equations for COMSOL simulations

There are two main methods to approach simulations regarding the free molecular flow regime. The *direct simulation Monte Carlo method (DSMCM)* combines the results of a large number of randomized particle trajectories. The angular coefficient method computes the incoming flux at each surface by summing up the flux arriving from all other surfaces in the line of sight. Macroscopic conditions of the enclosed volume can then be derived from the surface solutions. COMSOL uses the latter [20].

The particle speed distribution $\rho(v)$ of a system adhering to Equation 2.19 can be derived as

$$\rho(v) = \left(\frac{m}{k_B T} \right)^2 \frac{v^3}{2} \exp \left(-\frac{mv^2}{2k_B T} \right) \quad (3.4)$$

resulting in an average absolute speed of

$$\langle v \rangle = \sqrt{\frac{9\pi k_B T}{8m}} \quad (3.5)$$

with m , T and k_B being the particle's mass, the temperature and the Boltzmann constant respectively [20]. Equation 2.19 and Equation 3.4 can be used to calculate the incoming flux at a finite element arriving from all others in the line of sight. Considering a finite element x' with a particle emission rate of J' ($[J'] = 1/(\text{m}^2\text{s})$) and a surface area S' , the emitted flux leaving at angles $[\theta', \theta' + d\theta']$ and $[\phi', \phi' + d\phi']$ and velocities between $[v', v' + dv']$ is

$$dJ'_{x'} = J' dS' \rho(\theta') \sin |\theta'| d\theta' d\phi' \rho(v') dv'$$

The covered projected area of an element x at distance r and a surface angle θ with respect to r is then given by

$$A_{x' \rightarrow x} = \frac{r^2 \sin |\phi'| d\theta' d\phi'}{\cos \theta}$$

This results in a flux per unit area arriving at x from x' of

$$dG_x = \frac{dJ'}{A_{x' \rightarrow x}} = \frac{J' dS' \cos \theta \rho(\theta') \rho(v') dv'}{r^2}$$

By integrating over all possible speeds dv' and all surfaces S of the elements in sight, one obtains the total flux:

$$\begin{aligned} G &= \int_S \frac{J' \cos \theta}{r^2} \overbrace{\rho(\theta')}^{\text{Equation 2.19}} dS \cdot \underbrace{\int_0^\infty \rho(v') dv'}_{=1} = \\ &= \int_S \frac{J' \cos \theta \cos \theta'}{\pi r^2} dS = \\ &= - \int_S \frac{J' (n \cdot r)(n' \cdot r)}{\pi r^4} \end{aligned} \quad (3.6)$$

where n and n' represent the normal vectors of the respective finite element surfaces [20]. The method itself operates relatively straight forward, especially when calculating a stationary solution. However, the computational costs to obtain realistic solutions can be rather RAM and time consuming due to quite fine mesh requirements and rapid increase in matrix size.

3.4.2 Settings and boundary conditions

In order to obtain a modeling domain for gas or particle dynamic simulations, it is necessary to invert a given geometry of the surrounding solids. Their interiors can be excluded since the gas flow only depends on the surfaces.

Alongside numerical parameters such as the particle mass and the temperature, proper inlet and outlet boundary conditions need to be applied. All performed gas simulations use the option of an outgassing wall in combination with realistic mass flux values given in Section 3.3 to model buffer gas entering the RFQ cooler. The particle flux is equally distributed across the boundary area and progressing according to the cosine law of scattering (Equation 2.19) [20]. Affected mesh elements are assigned a constant flux value in the resulting computation matrix. As outlet condition, vacuum pumps have been applied to the corresponding areas in the model using the known pumping strengths shown in Subfigure 2.3(a). The fraction of incident molecules that are extracted from the domain by a pump with area A and pumping speed S is given by

$$f = \sqrt{\frac{2\pi M}{RT}} \cdot \frac{S}{A} \quad (3.7)$$

where M and R represent the molar mass of the simulated particle species and the universal gas constant respectively [20]. As simulations including the regions outside the cooler section were not feasible due to a massive increase in mesh elements, pumping conditions have been applied to the section openings at injection and extraction lenses. As a consequence, only the predictions for the occurring pressure at the center vacuum gauge, given as p_{center} and illustrated in Figure 3.5, have been directly comparable to experimental results.

Simulations using the free molecular flow interface do not require to assign specific materials, as none of the involved variables depends on material properties.

3.4.3 Evolution of the gas dynamics model

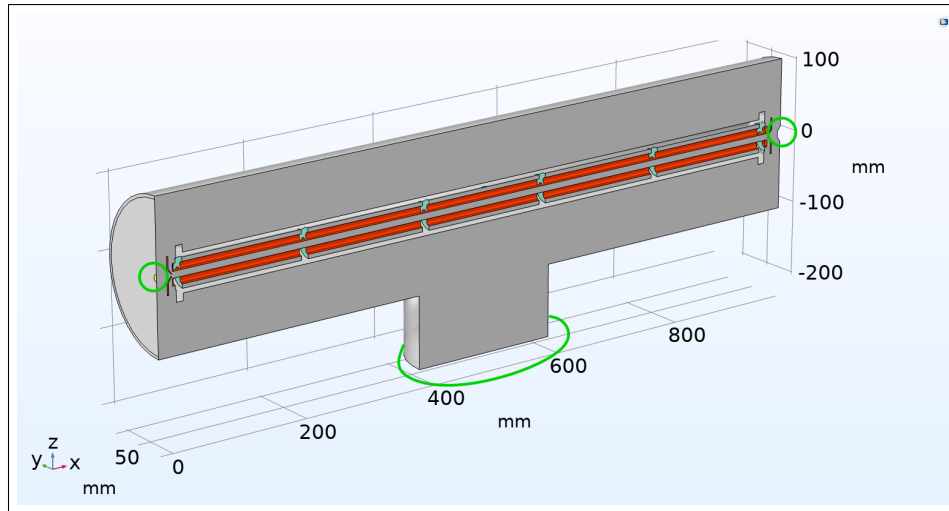
The quality of results increases with the mesh density, while computational resources only allowed for a total number of about $5 \cdot 10^5$ mesh elements. For this reason, the size of the modeling domain had to be reduced drastically without omitting relevant details. A crucial part to realize this has been the exploitation of symmetries. Although an axial symmetry condition has not been applied due to the presence of guiding and RF-electrodes, the implementation of several mirror symmetries has been possible. Such a boundary requires the incoming flux distribution to equal the outgoing one. In case a boundary with assigned pumping condition is partitioned, the corresponding pumping strength needs to be adapted such that the strength per unit area remains the same. Whenever such a condition has been applied, results at similar mesh quality have been checked to match with the former models before optimizing the mesh density. The same procedure has been performed whenever the geometry changed to a greater extent.

Figure 3.6 illustrates the evolution of the modeling geometry. A modeling domain needs to depict the regions where the simulated processes are occurring. To simulate the buffer gas flow in between the physical surfaces of the cooler tube, electrodes etc., the modeling domain shown in Figure 3.1 had to be inverted. Confining surfaces of important components such as electrodes or insulated spacers are highlighted in color in Figure 3.6.

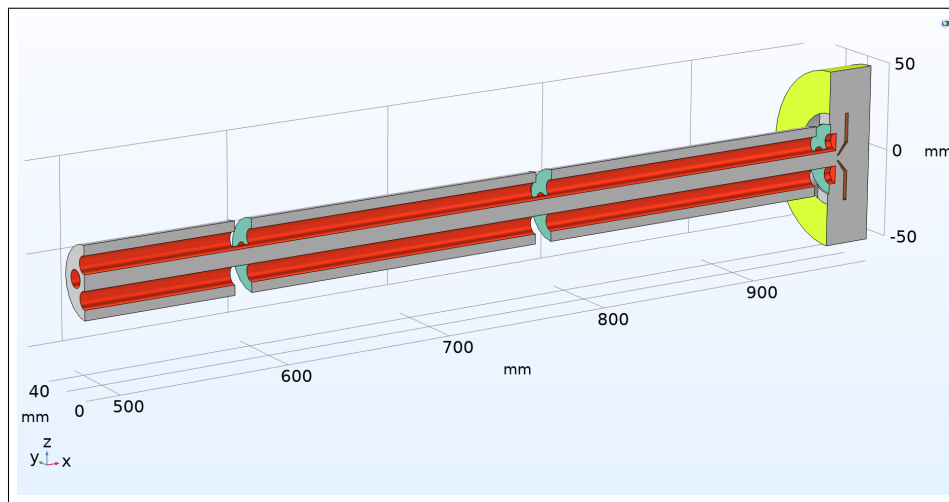
The initial model (Subfigure 3.6(a)) already shows significant simplifications when compared to Figure 3.1. While injection and extraction low lenses have been merged with the casing to serve as a single wall, high lenses have been removed from the geometry. This is because they are located outside the cooler vessel and thus do not considerably contribute to the resulting gas flow. Although the influence of the guiding electrodes on the gas flow within the cooler tube is not negligible, they had to be removed from the original model, as they increased the number of mesh elements to an extent not feasible for computation. The locations of the vacuum pump boundary conditions are circled in green. The domain has been cut in half vertically, while a mirror symmetry condition has been applied on the created surface.

Due to the extremely small contribution of particles reentering the cooler tube from the outside, the cooler vessel has been simplified drastically as can be seen in Subfigure 3.6(b). It has been approximated as a vacuum pump (highlighted in yellow) surrounding the cooler tube opening. The pumping strength on each ending had to be changed to half the value of the turbomolecular pump located at the center of the cooler vessel. Furthermore the geometry has been divided once more. A mirror boundary condition has been applied to the surface at the cooler center. As a matter of fact, attempts to also include a third symmetry condition had to be discarded due to highly unrealistic results.

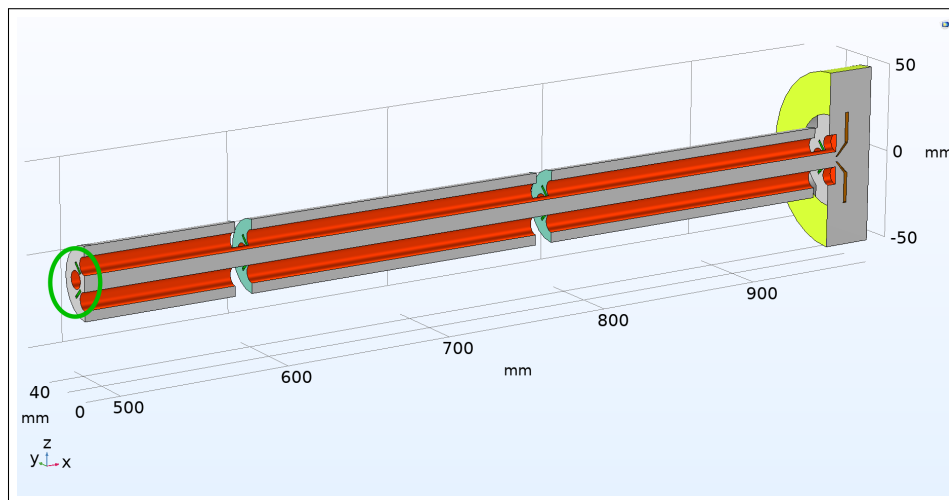
For the resulting model, shown in Subfigure 3.6(c), the mesh generating process has been done manually, assuring high density near the beam axis while tolerating lower density with increasing distance. This step made it possible to include the initially omitted guiding electrodes (highlighted and circled in green). Parameters of this model have been tuned until exceeding computational limits of the hardware to assure for high quality of results.



(a)



(b)



(c)

Figure 3.6: Evolution of the gas model geometries. Highlighted in yellow are the boundaries where a vacuum pump condition is applied. For better orientation, boundaries at important components are highlighted: RF electrodes (red), quad holders (light blue) (a) shows the initial geometry including the casing, (b) omits the casing to allow for a smaller geometry and therefore higher mesh quality, and (c) includes the guiding electrodes (hardly visible at the cooler center and highlighted in green).

3.4.4 Results

Figure 3.7 shows computational predictions of the final gas model for the outgoing pressure at the centered vacuum gauge (red) together with the experimental results (black, including uncertainties highlighted in green). Computational results increase linearly. On the one hand, this is a consequence of the linear correlation between inlet pressure and inserted mass flux which has been used as initial condition (see Table 3.2). On the other hand, the computational flux matrix based on free molecular flow dynamics described in subsection 3.4.1 scales linearly with applied boundary conditions. Experimental results show a nonlinear progression, especially for higher buffer gas masses. This behavior has already been described in subsection 3.3.4. For He, the model is in reasonable agreement with experimental data. Although the outlet pressure is underestimated, the relative error occurring in the range of typical operation conditions is below 10%. In the case of other buffer gases, deviations become rather high. More precisely, the outlet pressure for displayed inlet pressures in the range of 0.10 mbar - 0.30 mbar is underestimated about 43% and 53% in the case of N₂ and Ar respectively. As available data for comparison, regarding the ion behavior inside the RFQ cooler, only uses He as buffer gas, all following simulations have been performed for He. Due to lower rates of collisional detachment when using lighter buffer gases, the cooler is operated mainly with He anyway.

Figure 3.8 displays the simulated on-axis He number density for different displayed inlet pressure values. Note the step wise decline towards the cooler tube openings. This is a direct consequence of the six equally distributed ceramic spacers mentioned in Section 2.2. These act as local reductions in the cross sectional area with respect to the cooler tube diameter, hindering the buffer gas from flowing. This leads to the creation of five sections of almost constant buffer gas density. This is an important discovery based on the performed simulations, as future modifications of the RFQ cooler can now be planned under consideration of these geometry effects.

Figure 3.9 represents the simulated surface pressure within the cooler for He and a displayed inlet pressure of 0.25 mbar. Again, the step wise decrease towards the cooler openings caused by the quad holders is clearly visible. Pressure values are on the order of some Pa, with the peak located at the gas inlet in the cooler center. An overview of peak and average pressure values inside the cooler tube for different displayed inlet pressures is given in Table 3.3.

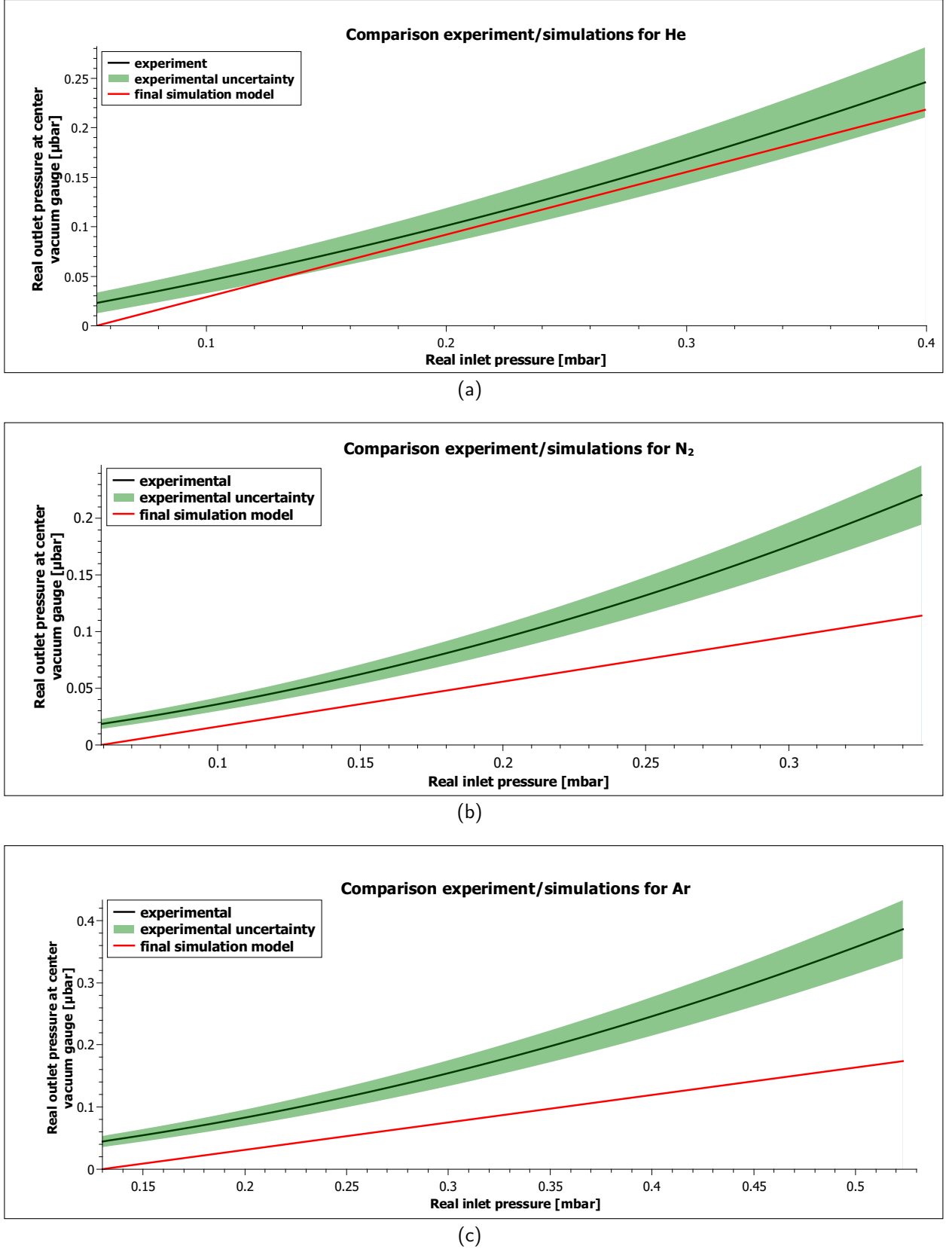


Figure 3.7: Comparison of simulated (red solid line) and experimental (black solid line) results of the outlet pressure at the vacuum gauge (in μbar) as a function of the real inlet pressure (in mbar) for (a) He, (b) N_2 and (c) Ar. The region of uncertainty for experimental results is given in green.

3. Methods and Results

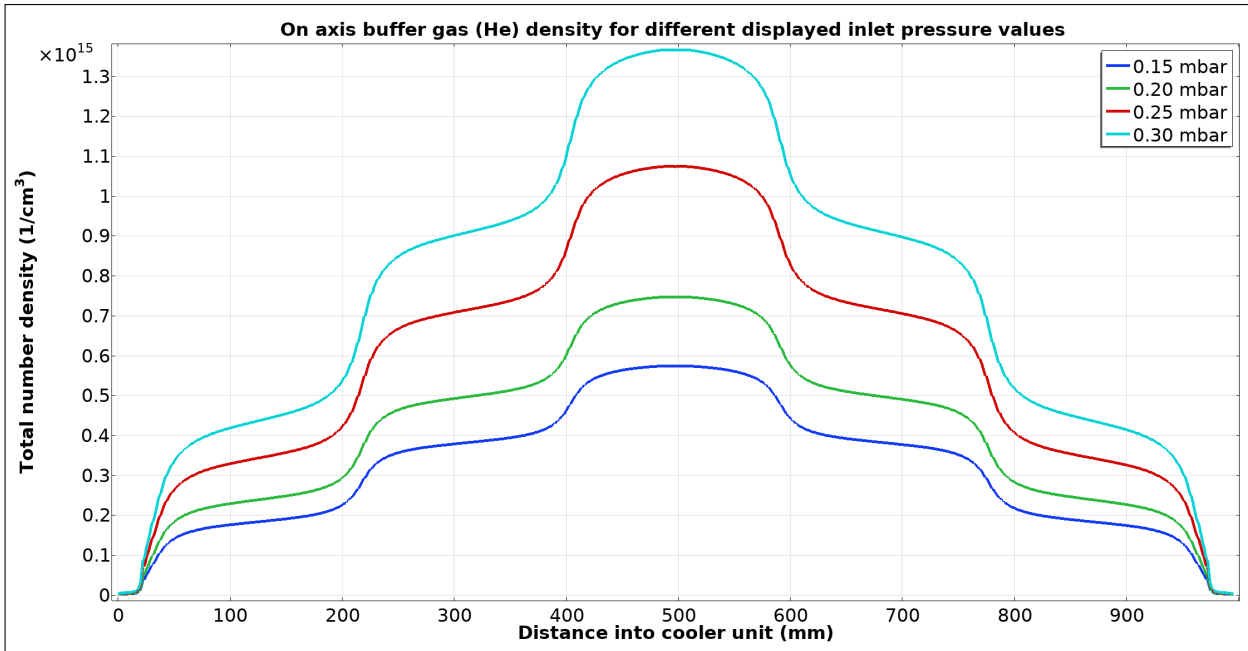


Figure 3.8: COMSOL results for the on-axis buffer gas number density for different displayed inlet pressures. The illustrated results are given for the buffer gas He.

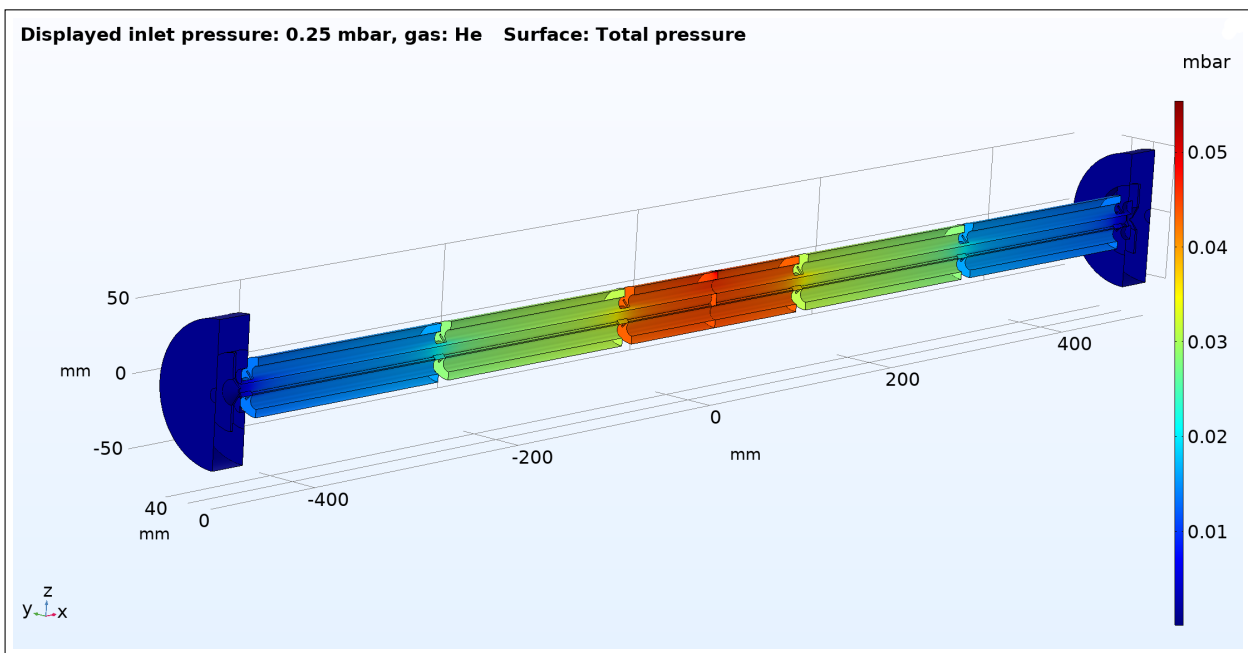


Figure 3.9: COMSOL results for the buffer gas pressure distribution inside the cooler tube, acting on the surface. Illustrated results are given for the buffer gas He and a displayed inlet pressure of 0.25 mbar.

Table 3.3: Simulation results for the peak and average pressure values (in Pa) inside the cooler tube for different displayed inlet pressures (in mbar).

Displayed inlet pressure [mbar]	0.15	0.20	0.25	0.30
Peak pressure inside the cooler tube [Pa]	2.98	3.88	5.58	7.10
Average pressure inside the cooler tube [Pa]	1.40	1.83	2.63	3.34

The COMSOL simulation model predicts pressure levels inside the RFQ cooler on the order of a few Pa at typical settings. This is in very good agreement with operation conditions of comparable buffer gas filled ion guides. Experiments performed at the IsoTrace Laboratory in Toronto by Eliades et al. 2011 [48], who used a negative ion cooler to yield selective chemical isobaric suppression, report operation pressures between 0.2 Pa and 1.4 Pa. Furthermore, publications by Liu et al. 2005 [18] describe the application of selective photo detachment at the Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory. Operation pressure of the He filled RF-quadrupole ion guide is stated at around 6 Pa. This is a major improvement compared to former simulations, which usually assumed constant buffer gas pressure across the whole cooler equal to the displayed inlet pressure and thus overestimated the real pressure by about an order of magnitude [15].

3.4.5 Implementation into presented model

The essential parameter needed for realistic particle simulations in rarefied gas environment is the buffer gas density across the modeled geometry. As indicated by the source equations of the free molecular flow package summarized in subsection 2.4.2, the flux matrix, and therefore the resulting particle density, scales linearly with the incoming particle flow f_N . Additionally, the particle density increases by a factor of $\sqrt{m_{gas}/m_{He}}$, due to the mass term in Equation 2.18, as slower particles result in longer spatial occupation and thus higher density (the normalization using m_{He} is solely to ensure consistent units).

This feature of scalability is a crucial part in the realization of a complete cooler model. It enables properly obtained and normalized gas dynamics data of a single simulation to be reused over the whole range of incoming flux values and buffer gas masses. Of course, this is only valid provided that all other factors such as the modeling domain and temperature stay the same. The normalization procedure for a specific particle density data set ρ_{spec} is of the form:

$$\rho_{norm} = \frac{\rho_{spec}}{f_N} \sqrt{\frac{m_{He}}{m_{gas}}} = \frac{\rho_{spec} m_{gas}}{f_{mass}} \sqrt{\frac{m_{He}}{m_{gas}}} = \frac{\rho_{spec} \sqrt{m_{gas} m_{He}}}{f_{mass}} \quad (3.8)$$

Here f_{mass} denotes the mass flux corresponding to f_N . Equation 3.8 has been used to treat the presented ion simulation model independently from the gas dynamics model, avoiding frequent gas dynamics simulations with computation times of several days. Furthermore it has been sufficient to implement an interpolated one dimensional function describing the buffer gas density along the beam axis for ongoing ion simulations. This has been possible because pressure and density are uniformly distributed along an axisymmetrical object due to the absence of gas currents. However, the application of such a simplified process requires the gas particles to reside within the free molecular flow regime. As comparison with experiments in Figure 3.7 show, this assumption holds only when using He, while for Ar and N₂ deviations are expected to be much stronger.

3.5 Electric fields simulation

COMSOL offers an *Electrostatics* as well as an *Electric Currents* interface as part of the *AC/DC Module* to model static and time dependent electrical fields [43]. Both of them use the finite element method (FEM), a numerical procedure for obtaining solutions to boundary-value problems by subdividing a geometry into sufficiently small finite elements. Derivatives are resolved spatially at every nodal point considering the contribution of all its neighbours. Using well defined boundary conditions, a solution matrix can be generated and reiterated until its residual becomes sufficiently small. A global solution can then be obtained via a suitable interpolation procedure between the nodal points [49]. Both interfaces solve for the electric potential V based on the classical Poisson's equation

$$E = -\nabla V \quad (3.9)$$

where E is the electric field intensity and ∇ is the common partial derivative operator [43, 49]. As the static guiding field is superimposed with the periodic RF-field, these two have been modeled independent of each other to include in later particle simulations.

3.5.1 Geometry

As the buffer gas density along the beam axis can be imported as a function derived from former gas simulations, the used geometry has been reduced to just the cooler tube and components within, extending up to injection and extraction low lenses (INJLO, EXTLO). Unnecessary features regarding ion simulations, such as the gas inlet, have been omitted. Also excluded are the ceramic spacers distributed inside the cooler tube. Although the presence of surface charges on these is likely, the COMSOL model does not include such effects. Furthermore, the high lenses (INJHI, EXTHI) have not been included in any ion simulations. Even though they contribute to the deceleration and focusing of the ion beam upon injection, the beam is immediately defocused upon entering the RFQ cooler and ion trajectories become randomized due to collisions with the buffer gas. For this reason, the influence of the high lenses has been assumed to be negligible. A representation of the resulting modeling geometry as well as the generated mesh can be seen in Figure 3.10. As the cooler itself is symmetric, only the injection section is shown to allow for more details. For better visualization of all involved components, the geometry is cut open. Subfigure 3.10(a) illustrates the different components of the geometry used to model electric fields and ion movement.

Similar to the gas dynamics simulations, the „empty“ space between the electrodes had to be included, as ion trajectories can only be computed within a domain. For this purpose, an inverted geometry filling the cooler tube has been added. This geometry has been split into three sub-domains, given in light blue, yellow and purple. They allow to apply a high quality mesh at the

beam axis, where it is most needed, while accepting a coarser mesh with increasing distance from the beam axis. Computation times have been reduced to roughly a third compared to simulations with an automatically generated mesh. It is necessary to say that occurring surfaces separating the different particle subdomains do not represent any physical surfaces and are not treated as such during simulations.

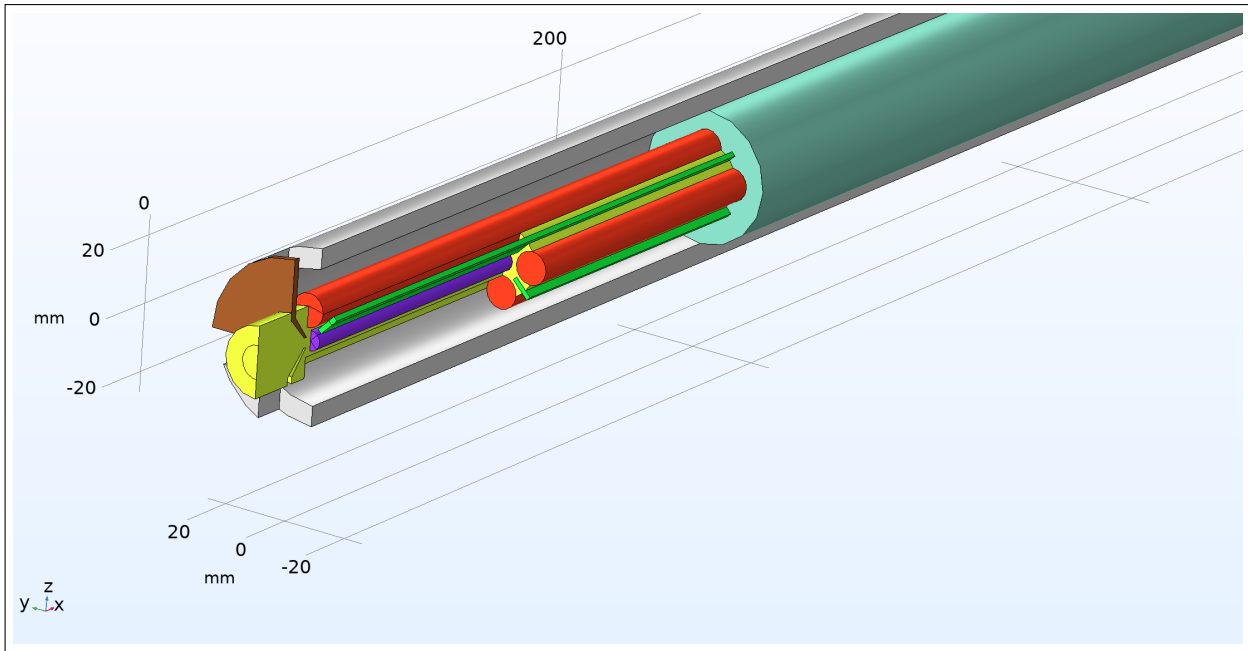
Subfigure 3.10(b) shows the mesh element volume for the resulting mesh. The color coding illustrates the volume of the mesh elements in mm^3 on a logarithmic scale. A step wise decrease in the mesh quality with increasing on-axis distance is evident, while the highest mesh quality is present at the tips of the aperture lenses as well as directly on the beam axis. The mesh of the remaining aperture lens appears to be non uniform. This can be explained by the presence of other structures distributed around them. The fine mesh of the nearby guiding and RF electrodes forces parts of the aperture to be meshed finer, as COMSOL only allows for a certain gradient of mesh volume to avoid convergence issues.

To obtain realistic results, material specific properties regarding the electrical behavior of involved materials are necessary. More precisely, both COMSOL interfaces require a value for the relative permittivity ϵ_r , while the electric currents interface also demands an entry for the electrical conductivity σ_e . The used material constants for the two assigned materials, the predefined *Steel AISI 4040* and the newly custom-defined *helium gas*, are given in Table 3.4.

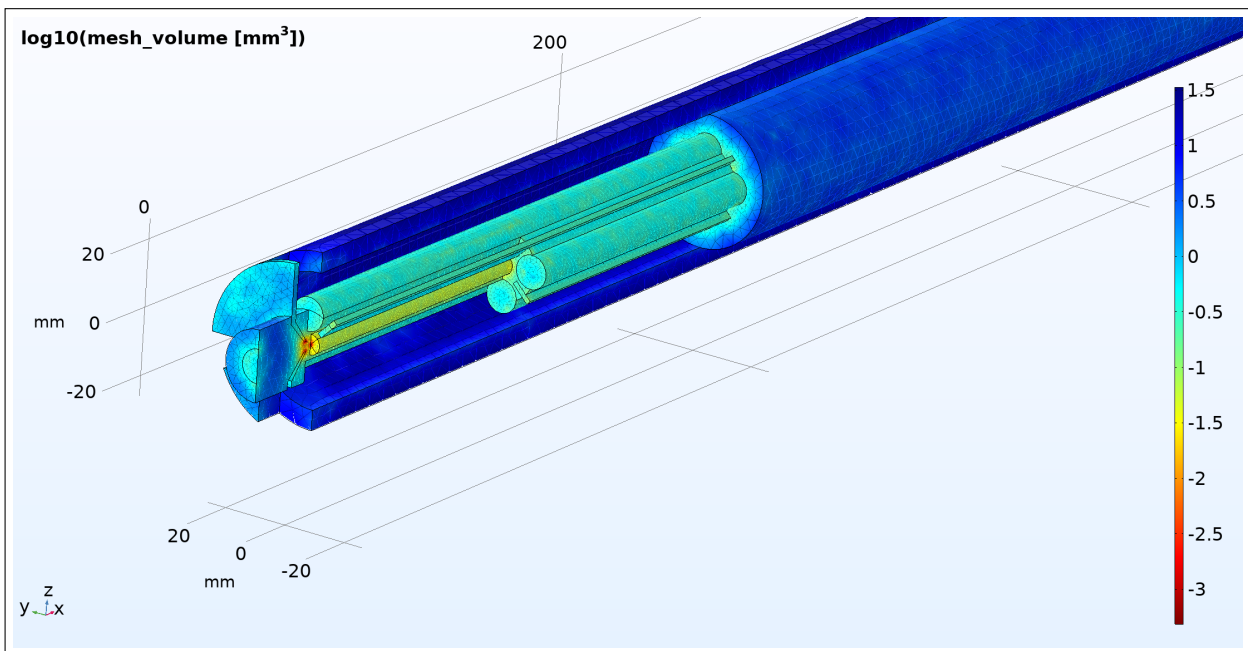
Table 3.4: Relative permittivity (ϵ_r) and electrical conductivity (σ_e) values for the materials Steel AISI 4340 and helium gas, taken from [50].

Material	ϵ_r [1]	σ_e [S m^{-1}]
Steel AISI 4340	2000	$4.0320 \cdot 10^{-6}$
helium gas	1	$2.9209 \cdot 10^{-24}$

It has to be noted that literature values for the relative permittivity of various steel alloys are hardly documented. Different sources state values between 100 and 4000 [50]. The choice in Table 3.4 is merely an average. However, as a precaution, simulations of the static and the RF-field have been performed to estimate the influence of this parameter. Results showed a maximal potential difference on the order of 10^{-13} V when changing ϵ_r from 100 to 4000. Therefore, it seems very likely that the actual value is not of great importance.



(a)



(b)

Figure 3.10: Representation of the COMSOL geometry and mesh used to simulate electric fields as well as ion trajectories. Due to the symmetry of the geometry, only the injection side is given to allow for a detailed view. The geometry has been cut open for better visualization of the present components.

(a) shows the pure geometry. Involved components are highlighted in different colors: Cooler tube (gray), RF-electrodes (red), guiding electrodes (green), aperture(s) (brown), subdivided „vacuum“ domains for later particle simulations (purple, yellow and light blue, going from finer to coarser mesh).

(b) illustrates the magnitude of the volume for single mesh elements (in mm^3). The implementation of different mesh qualities decreasing almost step wise with increasing distance to the beam axis is evident. Note the logarithmic scale.

3.5.2 Static guiding field

3.5.2.1 Underlying equations and simulation settings

Under static conditions, the charge density ρ remains constant at every point. Gauß's law describes ρ as

$$\nabla D = \rho \quad (3.10)$$

D represents the electric displacement field given by the constitutive relation

$$D = \varepsilon_0 E + P \quad (3.11)$$

where P is the electrical polarization vector and ε_0 the permittivity of vacuum. It is defined by the fundamental relation to the ideal constants μ_0 (permeability of vacuum) and c_0 (speed of light in vacuum) [43, 49].

$$\varepsilon_0 = \frac{1}{c_0^2 \mu_0} \text{ F/m} \approx \frac{1}{36\pi} \cdot 10^{-9} \text{ F/m} \quad (3.12)$$

Using Equation 3.9 and Equation 3.11, we can rewrite Equation 3.10 as

$$-\nabla(\varepsilon_0 \nabla V - P) = \rho \quad (3.13)$$

which is the source equation the COMSOL electrostatics interface solves for, although P could be set to zero in all present cases [43]. To model only the static guiding field - the time dependent RF-field is simulated independently - all components but the guiding electrodes have been set to ground level (cooler-HV). A charge conservation condition across the whole domain is automatically applied by COMSOL.

3.5.2.2 Results

Simulations have been performed to benchmark the guiding field for different applied voltages. Various mesh qualities and convergence criteria have been tested to assure reliable output. A cross sectional potential distribution of the purely static guiding field is shown in Figure 3.11, the results for different on-axis guiding potentials as functions of the applied guiding voltage are given in Figure 3.12.

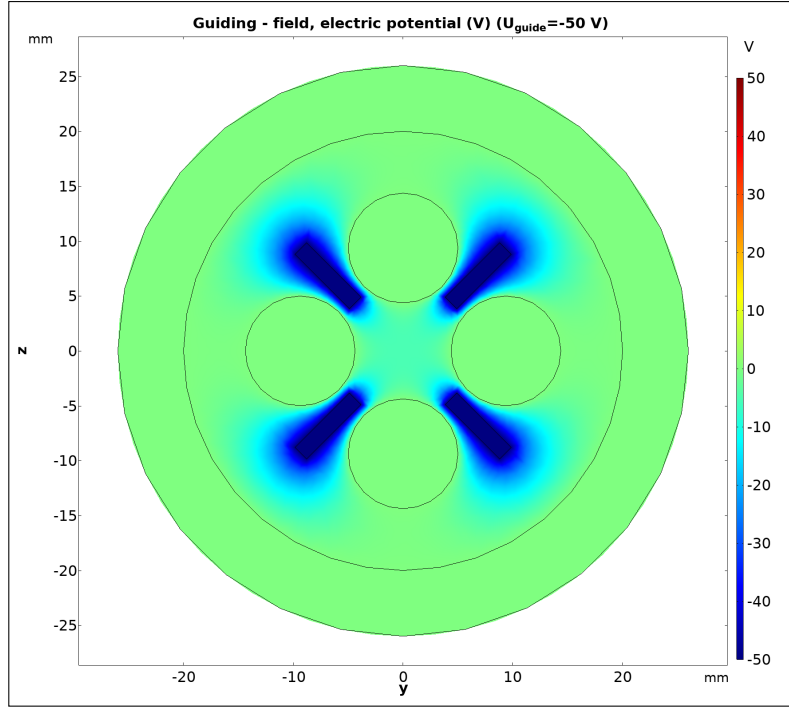


Figure 3.11: Cross sectional representation of the simulated guiding potential in direction of an incoming ion beam for an applied guiding voltage of -50 V. The edges of electrode structures have been included.

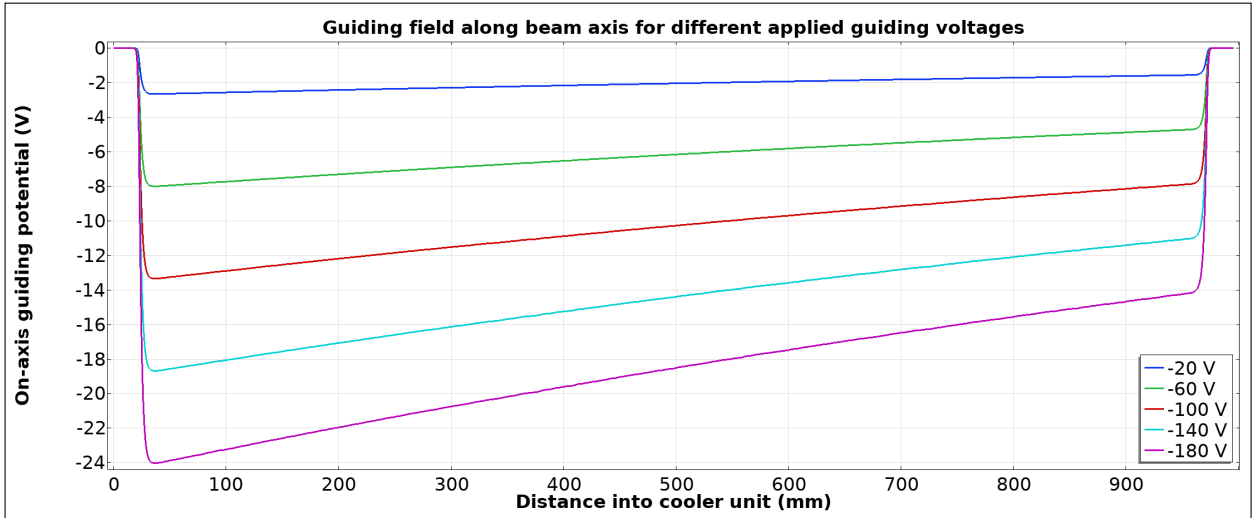


Figure 3.12: COMSOL results for the on-axis static guiding potential along the RFQ ion cooler for different applied guiding voltages.

As can be seen in Figure 3.12, the guiding field strength increases linearly with applied voltage as expected. The steep slopes that arise at the injection and extraction region are a direct consequence of the grounded apertures. Their conical shape penetrates the repelling fringe field of the guiding and RF electrodes. Incoming particles are therefore not defocused until they are confined by the RFQ trapping field. This has been a modification to avoid beam losses during the injection process [15].

The computed guiding field strength has been determined via linear fits of the slopes in Figure 3.12, using the evaluation software QTIPlot. The resulting linear correlation between applied guiding voltage U_{guide} and generated guiding field strength E_{guide} for trapped ions of charge number Z behaves according to

$$E_{guide} = -0.0576 \text{ m}^{-1} \cdot U_{guide} \cdot Z \quad (3.14)$$

This is in very good agreement with former simulations of Johanna Pitters [15], which yielded a correlation constant of -0.0588 m^{-1} . Typical operation conditions use a guiding field strength of about $2 \text{ V/m} - 3 \text{ V/m}$ [14].

3.5.3 RFQ-field

3.5.3.1 Underlying equations and simulation settings

The current density J in a static coordinate system can be described by the point form of Ohm's law

$$J = \sigma E + J_E \quad (3.15)$$

where σ is the electrical conductivity of the material and J_E an externally generated current density (zero in the given case). The time harmonic equation of continuity then yields

$$\nabla J = \nabla (\sigma E + J_E) = -i\omega \rho = -i\omega \nabla D \quad (3.16)$$

with ρ being the current density, i the common imaginary unit and ω the angular frequency of the time harmonic system to describe [43]. The last step in Equation 3.16 made use of Equation 3.10. Combining Equation 3.9, Equation 3.11 and Equation 3.16 one obtains

$$-\nabla [(\sigma + i\omega\epsilon_0) \nabla V - (J_E + i\omega P)] = 0 \quad (3.17)$$

which is the source equation of the COMSOL electric currents interface used for time dependent studies within the frequency domain [43].

Similar to the electrostatics interface, COMSOL by default applies a current conservation condition. As the generated time dependent electric field is periodic, the system can be computed using a study node that solves within the frequency domain. The applied voltage is then given by the amplitude value. For initial simulations, an amplitude of 200 V with respect to cooler-HV and a frequency of 2.62 MHz , according to experimental conditions in [26] have been used. All other components have been grounded to cooler-HV.

3.5.3.2 Results

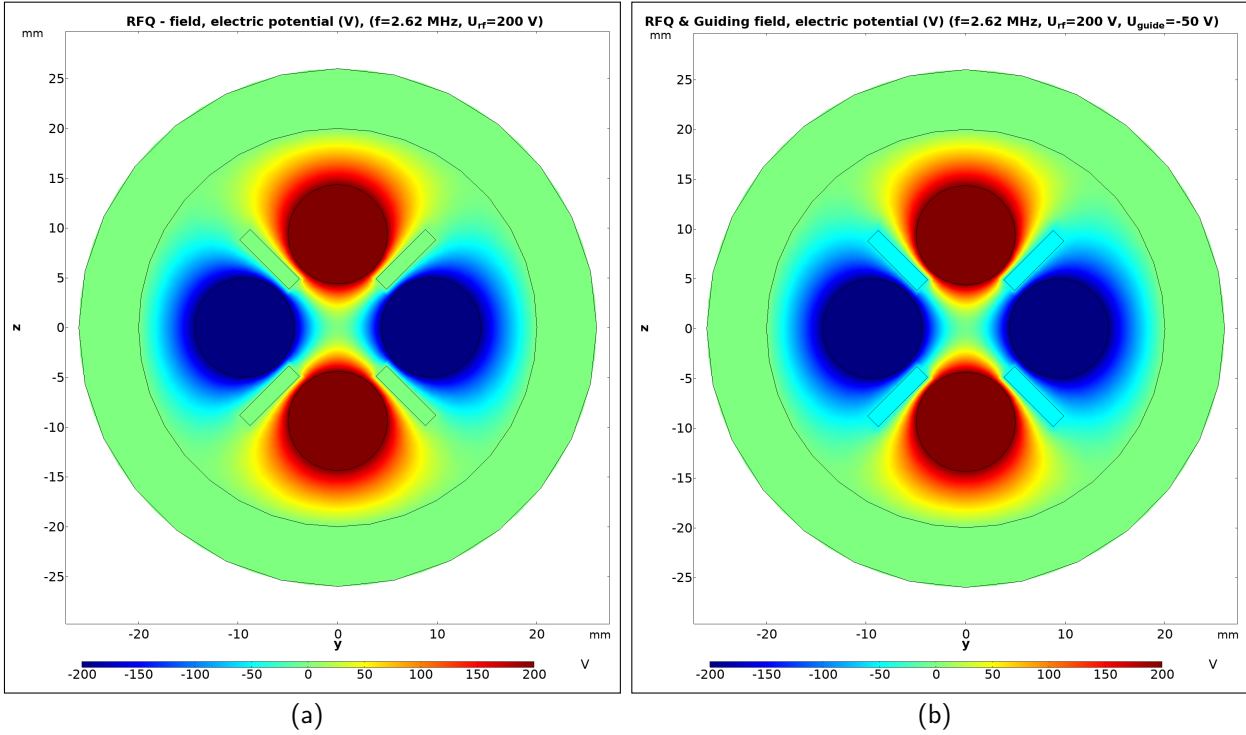


Figure 3.13: Cross sectional representation of the simulated RF-potential in direction of an incoming ion beam along the x-axis. Results are given for an applied frequency of 2.62 MHz, an RF-amplitude of 200 V and at phase $\varphi = 0^\circ$. The edges of electrode structures have been included. (a) shows only the RF-field while all other components are set to ground voltage; (b) illustrates the resulting superposition of the RF-field with the static guiding field for an applied guiding voltage of -50 V.

The resulting potential distribution for given parameters is shown in Figure 3.13. Subfigure 3.13(a) illustrates the pure RF-field while guiding electrodes are at ground potential. An overlay of both, the static guiding field and the time dependent RF-field at phase $\varphi = 0^\circ$, is given in Subfigure 3.13(b).

As expected, the potential on the surface of the RF-electrodes at phases $\varphi = (0^\circ, 180^\circ)$ adopts the positive and negative RF-amplitude values. For other phases, the potential then behaves according to a cosine function. In the case of symmetrically distributed electrodes, the resulting on-axis potential has to be zero over the whole cooler length and at all phases. This has been examined in order to check if the mesh quality has been high enough. The average magnitude of deviations has been on the order of 10^{-4} V, while single and rare outliers yielded about 0.02 V. Considering the magnitude of typical RF-amplitudes and guiding voltages, the quality of the mesh as well as the results have been considered sufficient.

Figure 3.14 illustrates cross sections of the simulated potentials of the RF and guiding field as well as their superposition. As one would expect, the potential at the respective electrodes equals

the applied voltage. In the case of the RF-field, electrodes at positive RF-amplitude values have been chosen. A phase of $\varphi = 180^\circ$ would cause a negative sign of the given RF-potential. The RF-field at beam center yields zero due to the opposite polarity of the generating electrodes. In contrast, the guiding field generates a negative potential at the beam axis, increasing with the distance into the cooler (see Figure 3.12).

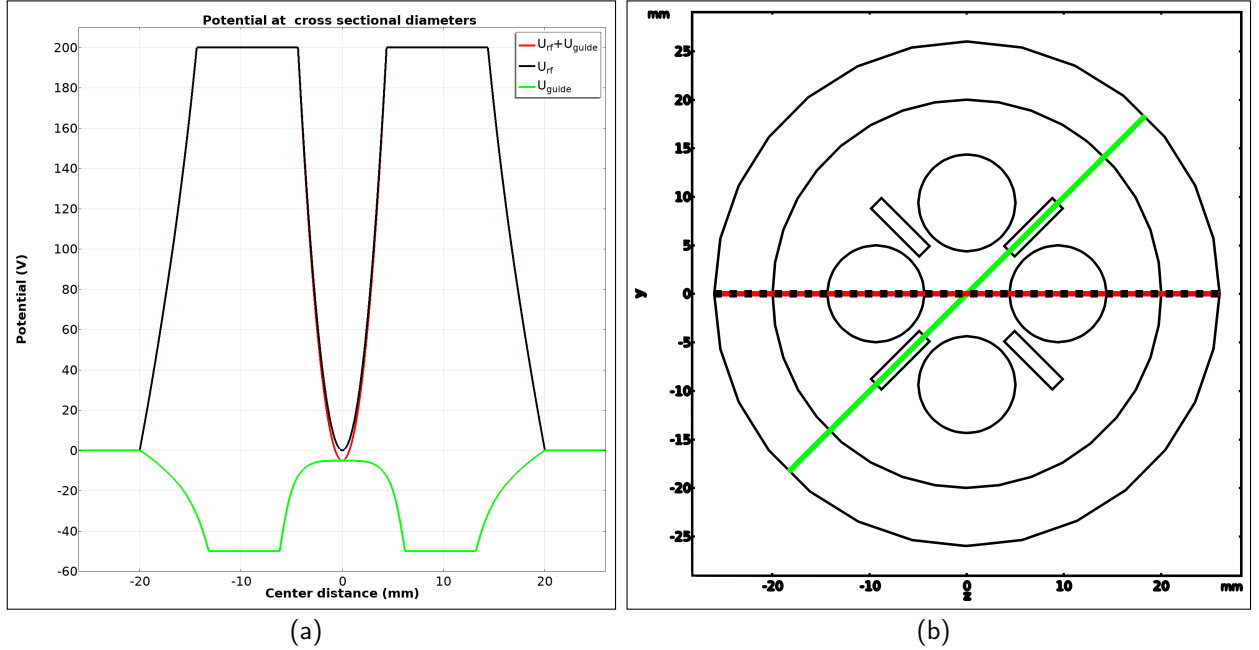


Figure 3.14: Various line potentials along diameters crossing the RF and guiding electrodes at the cooler tube center. Results are given for an RF-amplitude of 200 V, a guiding voltage of -50 V and at phase $\varphi = 0^\circ$ in the case of the RF-field.

(a) shows the different simulated potentials. The pure RF and guiding potential are shown as black and green respectively. Their superposition is highlighted in red. (b) displays the cross sectional diameters used for the potentials shown in (a). Lines have been chosen such that they eventually traverse the electrodes of the respective fields they show.

3.5.4 Implementation into presented model

Contrary to the gas dynamics case, FEM-computation of electric fields is very fast and storage efficient, as only one dependent variable needs to be evaluated. Therefore, it is feasible to combine simulations within a single model including both, the electrical components to calculate the generated fields prior to ion simulations, and the inverted geometry in which affected ion trajectories are then computed.

COMSOL allows to pass on the mesh based solutions of former studies as initial values of independent variables used for ongoing studies. This means that the solution of the electrostatic field can be passed on to a time dependent study of the RF-field. Both solutions can then be passed to a study of the charged particle tracing interface. Trajectories of ions can thereby be calculated under the influence of the formerly computed electric fields.

3.6 Ion-gas scattering simulation

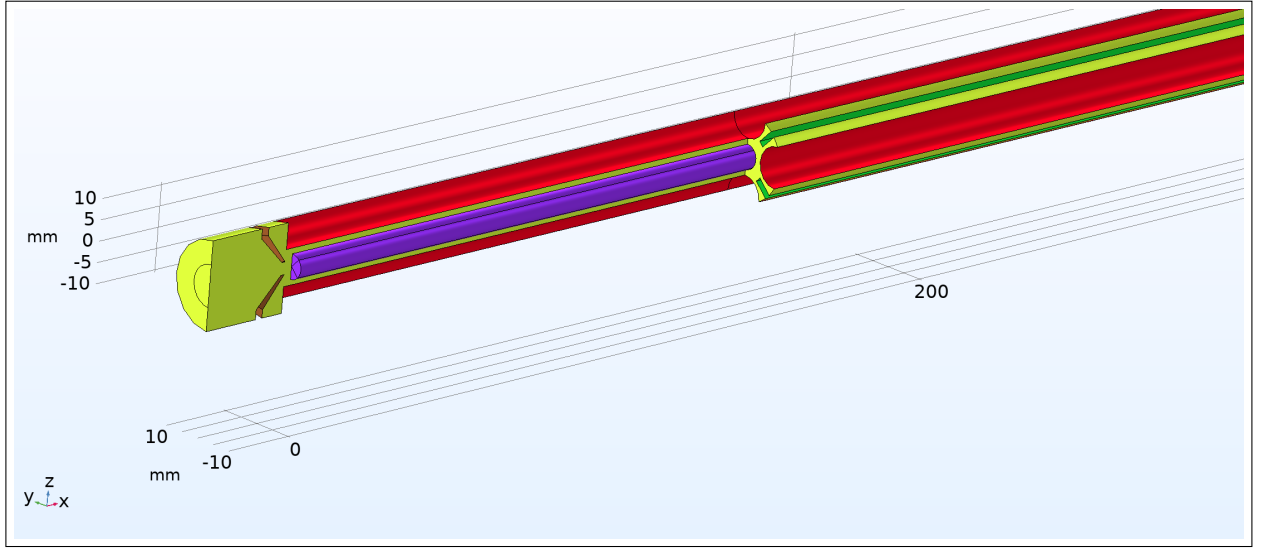
The most important prediction an ion simulation model of the RFQ negative ion cooler should achieve is the residence time. The influence of ion species, buffer gas pressure, RF-amplitude and guiding voltage had to be investigated. Experiments performed with $^{63}\text{Cu}^-$ ions by Martchini et al. 2017 [26] provide data for comparison. Apart from that, also statements about the average energy the ions are cooled to, as well as the necessary time and penetration length to reach that temperature are of great interest. Only a SIMION model generated by Johanna Pitters [15] has made predictions about these conditions. It is based on a HS elastic collision model that has been compared and deemed superior with respect to a viscous damping model on the example of $^{36}\text{Cl}^-$ ions. COMSOL offers a *Particle Tracing Module* to compute the trajectories of particles subjected to various forces, particle-boundary interactions and particle-domain interactions. The contained *Charged Particle Tracing interface* concentrates on the computation of ions [32]. Simulations have been performed using both mentioned ions, in order to compare the results with former experiments and existing computational models.

3.6.1 Particle geometry

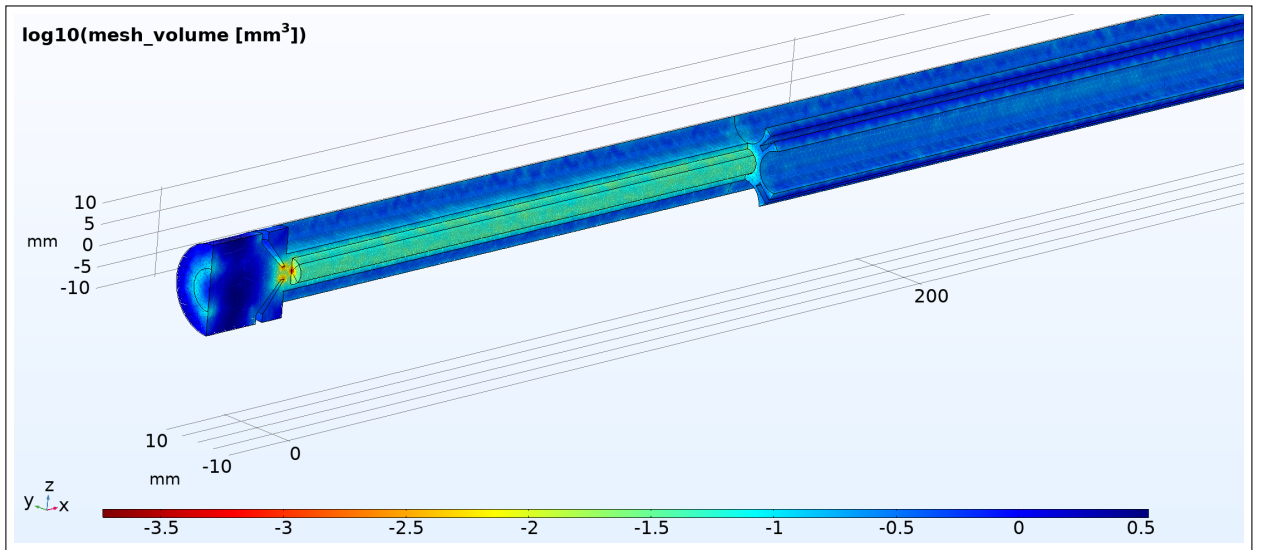
As explained in subsection 3.5.1, simulations of present electric fields as well as particle simulations have been combined into one model. However, in order to significantly decrease computation time to roughly a third, only a small part of the modeling domain, highlighted in yellow in Subfigure 3.10(a), has been used to simulate ion trajectories through the cooler. Figure 3.15 shows the modeling domain used for ion simulations as well as its mesh quality. Neighbouring components that have not been included are still indicated by their highlighted color in Subfigure 3.15(a) (RF-electrodes (red), guiding electrodes (green), apertures (brown), high mesh quality subdomain (purple)). The maximal radius for the depicted domain has been chosen at the centers of the RF-electrodes. Doing so has guaranteed that particles reaching any boundary of the particle domain are terminated in any case, even when choosing to use the full geometry from Subfigure 3.10(a). This does not contradict the experimental situation as any ion at this point is lost anyway. COMSOL allows for the import of former simulated electrical study results in the form of initial conditions for proceeding studies. As these have been simulated using the complete geometry, the change of geometry for ion simulations has no influence on the impact of electric fields.

The subdomain given in purple does not represent any real structure within the cooler. Rather it serves as a control region for which a mesh of finer quality with respect to its surrounding can be assigned. This assures optimal mesh quality at the beam axis where particles will be simulated, while accepting a coarser mesh on the regions outside. Boundaries appearing between the mesh control region (purple) and the rest of the particle domain (yellow) do not represent

actual surfaces and are therefore not treated as such in simulations. Subfigure 3.15(b) shows a plot of the volume distribution of generated mesh elements. Note the step wise decline in mesh quality between the near axis region and the rest of the domain.



(a)



(b)

Figure 3.15: Modeling domain used for particle simulations. To cut down computation times, only a subdomain of the complete geometry given in Subfigure 3.10(a) has been used.

(a) is a representation of the modeling geometry. As the geometry is symmetric, an extended perspective has been chosen to allow for a detailed view. Parts of the geometry have been cut open to allow for a good representation of all included components and subdomains. Boundaries of omitted components are highlighted in color (RF-electrodes (red), guiding electrodes (green), apertures (brown), particle geometry (yellow) and high quality subdomain (purple)). The purple subdomain serves the purpose of manually assigning a mesh of higher quality near the beam axis to improve accuracy of results. It does not represent any real structure within the cooler and is not handled as such within simulations.

(b) shows the volume distribution in of the generated mesh elements (mm^3) for the particle geometry on a logarithmic scale.

3.6.2 Underlying equations in COMSOL

3.6.2.1 Particle propagation in electromagnetic fields

The non relativistic Lagrangian for a charged particle with mass m_p , velocity v and charge $Q = Z \cdot e$ (Z representing the charge number and e the elementary charge) moving in an electromagnetic field described by the electric scalar potential V and the magnetic vector potential A is given by

$$L = \frac{m_p v^2}{2} + QA - QV \quad (3.18)$$

The electric field E and the magnetic flux density B can be described by [51]

$$E = -\frac{dA}{dt} - \nabla V \quad (3.19)$$

and

$$B = \nabla \times A \quad (3.20)$$

respectively. The equations of motion can be obtained by the Lagrange equation

$$\frac{d}{dt} \left(\frac{dL}{dv} \right) = \frac{dL}{dq} \quad (3.21)$$

where q is the particle position vector. Solving Equation 3.21 by inserting Equation 3.19 and Equation 3.20, one obtains

$$\frac{d(m_p v)}{dt} = -ZeE + Ze(v \times B) = F \quad (3.22)$$

where F represents the force acting on an individual particle. Equation 3.22 is the equation of motion solved by the charged particle tracing interface when using Newtonian formulation. An option for relativistic correction is implemented within the COMSOL framework [32]. However, as particle energies within the RFQ negative ion cooler are on the order of eV, this feature has not been necessary.

3.6.2.2 Time stepping

Naturally, simulations of particle trajectories require to update position and velocity components at discrete time steps rather than continuously. The choice of the time interval is a crucial one and an individual field of research. Choosing it too large, influences affecting the particle motion can be underestimated and cause wrong results. For example, a poor choice of a time step equal or similar to the time period of a sinusoidal electrical field would cause simulation results

of an ion motion through a constant electric field, as all other phases are skipped. Also when simulating particle scattering, time steps should be sufficiently small such that the probability of two interactions within one interval becomes negligible. Otherwise simulation results would correspond to an underestimated interaction cross section.

When choosing very small time steps, results usually become accurate and reliable. However, this often leads to a significant increase of computation time and resources. Proper time stepping algorithms are therefore necessary to ensure for time steps to be sufficiently small to obtain accurate results, but at the same time large enough to keep computation times as low as possible. The time dependent solver implemented within the charged particle tracing interface offers three different time stepping methods.

- The *backwards differentiation formula (BDF)* solver is an implicit solver that uses backward differentiation formulas with variable order. This means that a high order will be used if possible, while regressing to lower orders if necessary to obtain stability. BDF-methods have been used for a long time and are known for their stability [52].
- The *Runge-Kutta solver* is an explicit time stepping method most suitable for non stiff problems. This solver is most efficient and reliable if the particle's position, velocity or other dependent variables are not expected to change abruptly over short time intervals [32, 52].
- The *Generalized- α* solver has properties similar to a second order BDF solver. It introduces a so called α -parameter that controls the degree of damping for higher frequencies and adapts during computation. This makes this solver much more accurate when comparing to (second order) BDF-methods, but also less stable [32, 52, 53]. The COMSOL solver detects whether a dependent variable is of first or second order and applies the correct formulas accordingly [52].

Particle motion inside the RFQ cooler undergoes severe changes perpendicular to the beam axis due to the periodic RF-field with a frequency of several MHz. Therefore the application of the Runge-Kutta time stepping method has not been considered. In such a case, it is recommended to use the generalized- α method, as long as solutions are stable and converge, before switching to the BDF-method [32, 52]. All particle simulations performed within the scope of this master thesis have used the Generalized- α time stepping method without encountering problems with stability or convergence. While it is possible to implement an individual time stepping method, it is strongly advised not to use this feature.

There are several options to force the solver to evaluate at specific time steps (intermediate, strict, manual) in addition to, or instead of the time steps generated by the used method [32]. However, as the scattering of particles inside the cooler is randomized, this option has not been necessary. The time stepping for all mentioned simulations has been set to „free“ to allow for highest stability and accuracy.

3.6.2.3 Elastic collisions

COMSOL's default solver node for modeling elastic collisions uses the HS model. The basic mathematics of this collision model is described in subsection 2.3.1. The following summary therefore reduces to COMSOL's numerical approach of random number sampling for collision detection.

Considering an ideal background gas with mass m_{gas} at temperature T , the root mean square (RMS)-velocity is given by

$$v_{rms} = \sqrt{\frac{3k_B T}{m_{gas}}} \quad (3.23)$$

with corresponding RMS-velocity components [33] $v_i = (v_{x,rms}, v_{y,rms}, v_{z,rms})$

$$v_{i,rms} = \sqrt{\frac{k_B T}{m_{gas}}} \quad (3.24)$$

The collision frequency $\nu(t)$ for a reaction between a model particle moving through a background gas with local particle density n_{gas} can be expressed in terms of the collisional cross section σ

$$\nu(t) = n_{gas} \int \sigma(|v_{rel}|) \cdot |v_{rel}| \cdot f(v_{gas}) dv_{gas} \quad (3.25)$$

where v_{gas} is the velocity of a background gas particle and $f(v_{gas})$ is a probability distribution function of the background gas particles, usually assumed to be Maxwellian. The variable v_{rel} denotes the relative velocity between a model particle moving with velocity v_{ion} and a background particle, given by

$$v_{rel} = v_{ion} - v_{gas} \quad (3.26)$$

The integral in Equation 3.25 is taken over all velocity space [54]. When using finite time steps t to model collisions, the probability $P(t)$ of a collision occurring in the interval $[0, t]$ yields

$$P(t) = 1 - \exp\left(-\int_0^t \nu(t') dt'\right) \quad (3.27)$$

The COMSOL collisions node uses a stochastic approach to model particle collisions with atoms or molecules in a background gas. The following conditions apply when using the collision detection option *“At time steps taken by the solver”*, which has been the case for all particle simulations described within this master's thesis.

At every time step taken by the solver, a random gas background particle is sampled and initialized with random velocity components using the implemented *randomnormal()* function centered at $v_{i,rms}$ described in Equation 3.24. This creates uncorrelated Gaussian pseudorandom numbers for each velocity component. The velocity distribution function $f(v_{gas})$ in Equation 3.25 can then be replaced with a Dirac Delta function at sampled velocity components, yielding a new expression for the collision frequency [54]:

$$\nu = n_{gas} \cdot \sigma(|v_{rel}|) \cdot |v_{rel}| \quad (3.28)$$

This simplification requires two criteria for the used time step t to be valid:

1. t must be small enough such that the probability of two or more collisions with background gas molecules in a single time step is negligibly small.
2. t must be small enough so that the particle velocity can be treated as a constant value over the time step.

The second criteria is of great importance, as the collision cross section usually depends either directly on the particle velocity, or indirectly via the particle kinetic energy [54]. If these criteria hold, one deals with time steps significantly smaller than the mean free time between two collisions ($t \ll 1/\nu$). Under such conditions, the collision frequency can be assumed constant at each time step, which reduces Equation 3.27 to [32]:

$$P(t) = 1 - \exp(-\nu \cdot t) \quad (3.29)$$

In order to determine, whether a collision is to take place or not, a random number $U \in [0, 1]$ is sampled and compared to the expression in Equation 3.29. For $U < P(t)$, a collision takes place and is treated according to Equation 2.12. The impact parameter b , as well as a rotation angle around the center of mass frame are sampled from a uniform distribution using COMSOL's implemented *random()* function. This is consistent with the theory described in subsection 2.3.1.

3.6.3 General settings

The main interest lies on the residence times of ions in the cooler. Ion injection for simulations has been placed right within the opening of the INJLO lens. A cylindrical beam node has been applied, with a uniform ion density and constant kinetic energy. The initial energy has been chosen such that it reaches 50 eV at the beginning of the cooler tube after electrostatic deceleration. The beam radius has been set to the maximum value at which no beam losses occur at the injection aperture in order to gain the maximum amount of data. As the model used here does not include any space charge effects, the choice of an injected ion current is not necessary, as it has no influence on the outcome.

To ensure proper time steps for the use of Equation 3.28 and Equation 3.29, a maximum time step constraint has been built into the time dependent solver. The maximum value has been chosen to never surpass 1% of the present mean time between two collisions for a single ion, given by the inverse of the collision frequency ν . However, simulations showed that the time dependent solver automatically chooses time steps within the same range.

Simulations have been performed with 1001 particles of randomly sampled initial positions, higher numbers would have required excessive computation times. For the same reason, data storage of particle properties (position and velocity coordinates) has not been possible for every used time step. Instead, only the particle properties after completion have been stored to read out residence times. In order to still access data during the progression through the cooler tube, the COMSOL feature „*global variable probes*“ has been used. Such probes allow the storage of a single global variable for each time step [19]. Such global variables have been implemented to record parameters of interest (kinetic energy, progressed distance, on-axis distance...) averaged over all active particles within the simulation. A stop condition has been implemented, ending the simulation as soon as 99% of initial particles have been terminated. This has been a reaction to single particles residing in the simulation, almost doubling storage as well as computation time. Including all features described, a single simulation run takes between several hours up to a day, depending on the parameters influencing the residence time (buffer gas pressure, guiding field strength, collisional cross section).

3.6.4 Comparison of $^{63}\text{Cu}^-$ simulations with experimental data

3.6.4.1 Settings and boundary conditions

Cooler settings used for simulations of $^{63}\text{Cu}^-$ ions are based on operation parameters in experiments, published by Martschini et al. 2017 [26] and performed on an independent test bench. Amongst other, they measured and documented the residence times of $^{63}\text{Cu}^-$ ions through the ILIAMS RFQ negative ion cooler, using He as buffer gas.

The operational conditions are summarized in Table 3.5. Here, the injection energy refers to the kinetic energy of the ions when entering the cooler tube. The buffer gas properties as well as the displayed inlet gauge pressure are given in Table 3.6. The corresponding mass flux into the cooler has been calculated using the experimental data summarized in Table 3.2. Table 3.7 shows the applied voltages for all given electrodes relative to the cooler-HV, which has been treated as ground voltage. Note that the applied voltage for the RF-electrodes represents the amplitude of the periodic signal rather than a constant value.

To account for the focusing during and the defocusing after passing the injection and extraction apertures, a voltage has been applied to the INJLO and EXTLO low lenses according to typical operation parameters taken from [15]. The high lenses (INJHI and EXTHI) are not included in the geometry.

Table 3.5: Operational conditions for performed experiments on $^{63}\text{Cu}^-$ ions described in [26]. The mass value has been taken from [55]. The values of the injection energy, Mathieu parameter and RF-frequency are given in [26].

Ion	Mass	Charge state	Injection energy	Mathieu parameter	RF-frequency
^{63}Cu	62.93 u	-1	50 eV	0.42	1.97 MHz

Table 3.6: Properties of He (taken from [55]) for the main part of the experiments performed on $^{63}\text{Cu}^-$ ions. The inlet pressure refers to the displayed, uncorrected inlet pressure on the inlet-gauge and is taken from [26]. The corresponding mass flux has been calculated using the conversion parameters given in Table 3.2.

Buffer gas	Mass	Inlet pressure	Inlet mass flux
He	4.00 [u]	0.30 [mbar]	$4.34 \cdot 10^{-8}$ [kg/s]

Table 3.7: Voltage settings of the electrodes included in the ion simulations. All values are given relative to the cooler-HV, which is considered as ground voltage. Typical values for the low lenses (INJLO and EXTLO) have been taken from [15], others are from [26]. INJHI and EXTHI values are not given as they are not included in the COMSOL simulations. The RF-voltage corresponds to the amplitude of the periodic voltage applied. The voltage for the guiding electrodes corresponds to a guiding field strength of 2.2 V/m (as used in [26]) when applying Equation 3.14.

Electrode	INJLO	EXTLO	Guiding	RF-amplitude	Apertures	Cooler tube
Applied voltage [V]	940	400	-38.22	200	0	0

As mentioned in Section 2.3, the HS-model for elastic collisions used by COMSOL has been extended by the implementation of an energy dependent cross section, thus applying the VHS collisional model (see subsection 2.3.2) [33].

The required VHS reference parameters for the presented simulation results are given in Table 3.8 (including values for the optional buffer gases N_2 and Ar). While such values are well documented for natural gases (see [33]), reference parameters describing the gas dynamics behavior of metals and especially their negative ions are hard to find. Values for Cu shown in Table 3.8 have been calculated using density functional theory (DFT) on Cu-Cu atomic pairs created during the laser induced plume expansion of a Cu-target by Petrov et al. 2020 [56]. Already included are reference parameters used in $^{36}\text{Cl}^-$ simulations. For these, documented reference parameters of the gaseous Cl_2 have been used [33].

The reference parameters for different isotopes are usually rather similar, while the effects of an additional electron or even atom may be of much greater influence. However, despite great effort, better fitting values have not been found.

Table 3.8: The VHS reference parameters for the three available buffer gases He, N₂ and Ar (taken from [33]), as well as for the simulated ions Cu (taken from [56]) and Cl₂ (taken from [33]). Simulations using ³⁶Cl⁻ have been performed to compare the presented model with former computational approaches ([15]). As no documentation of VHS reference parameters for pure Cl could be found, known parameters for the gaseous Cl₂ have been used.

Listed below are the reference diameter d_{ref} , the reference temperature coefficient ω_{ref} and the reference temperature T_{ref} . The averaged reference parameters for the elastic collision reaction He-Cu and He-Cl₂ are also shown.

	d_{ref} [Å]	ω_{ref} [1]	T_{ref} [K]
He	2.330	0.660	273.15
N ₂	4.170	0.740	273.15
Ar	4.170	0.810	273.15
Cu	5.773	0.737	300.00
Cl ₂	6.980	1.010	273.15
He-Cu	4.052	0.696	286.58
He-Cl ₂	4.655	0.835	273.15

By inserting the reference parameters from Table 3.8 into Equation 2.14, one can derive a reference value for the relative velocity $v_{r,ref}$. The pseudorandom velocity components of the sampled gas particles can then be used to calculate the present relative velocity v_r . Plugging all obtained values into Equation 2.13 yields the energy dependent cross section for the current time step. Figure 3.16 shows the VHS cross section for He-Cu as well as He-Cl₂ scattering according to the parameters from Table 3.8. Also included is the constant HS collisional cross section for elastic scattering of ³⁶Cl-He, which has been assumed within SIMION models by Johanna Pitters (2015, [15]) and yields about $3.12 \cdot 10^{-15} \text{ cm}^2$.

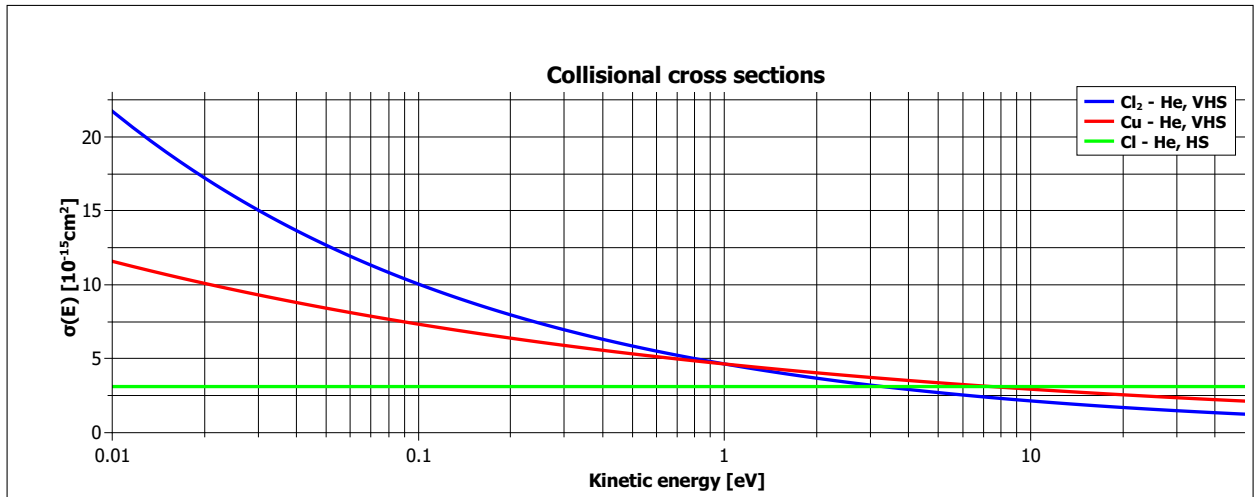


Figure 3.16: VHS elastic scattering collisional cross section for He-Cl₂ (blue) and He-Cu (red) as functions of their respective kinetic energy. VHS cross sections are based on the VHS reference parameters given in Table 3.8. The constant HS cross section used for SIMION simulations in [15] (green) is shown for comparison. Note the logarithmic scale of the kinetic energy. As can be seen, the VHS cross sections increase strongly with the decrease of particle kinetic energy and velocity.

3.6.4.2 Results

Residence time

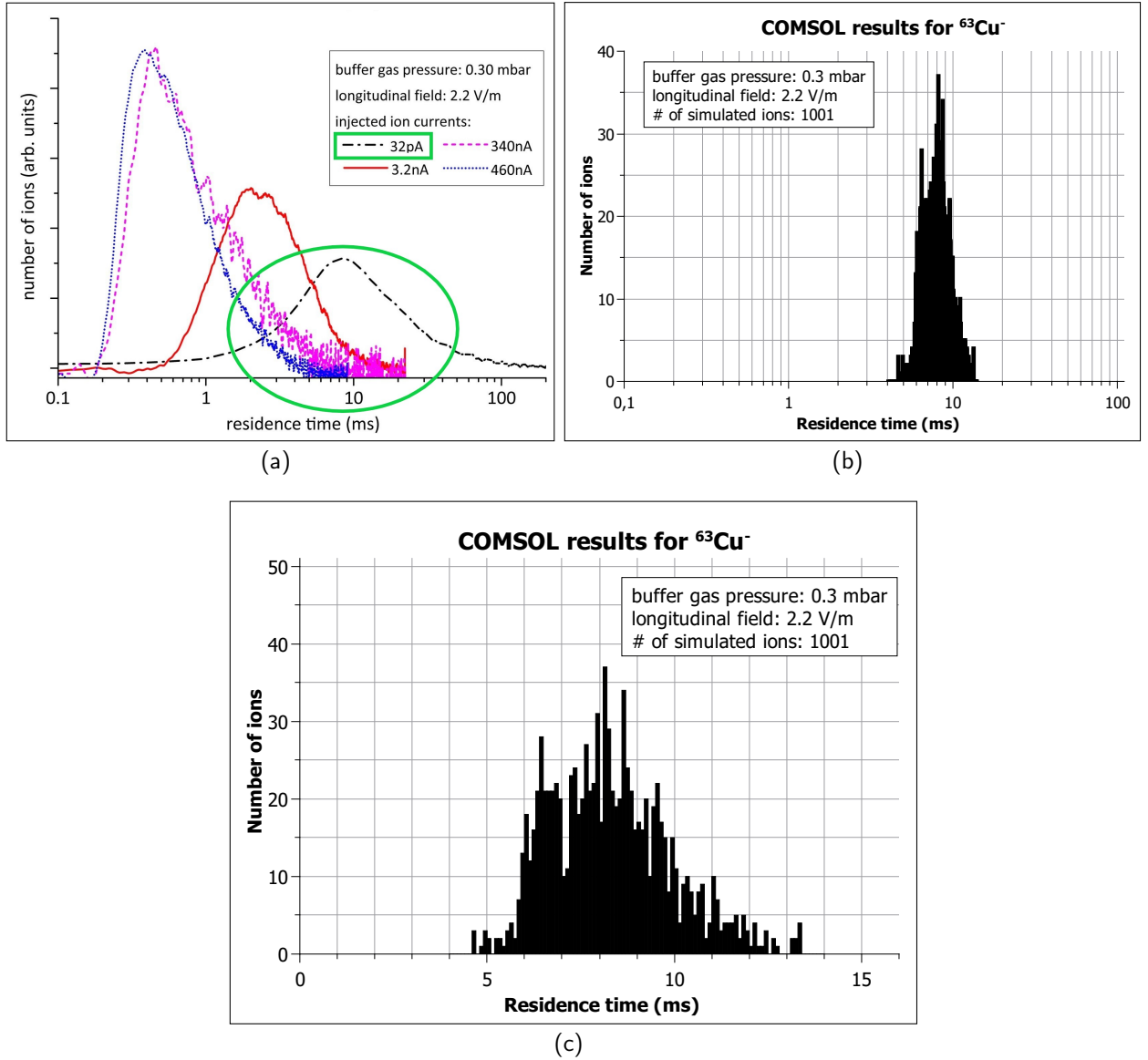


Figure 3.17: Simulation results of the residence time distribution of $^{63}\text{Cu}^-$ ions.

(a) shows experimental results from [26]. Studies have been performed for various injection beam currents. The results indicate the strong influence of space charge effects on the residence times of ions through the RFQ cooler. As the computational model does not account for these, experimental results for 32 pA (framed in green) are most representative for comparison, as space charge effects are expected to become negligible at such low currents. Adapted from [26].

(b) represents the results from COMSOL simulations for $^{63}\text{Cu}^-$ ions passing through the RFQ cooler, using the VHS scattering model based on reference parameters given in Table 3.8. Cooler operational conditions for these simulations were identical to the experiments in (a). The timescale has been chosen logarithmic to allow for better comparison with experimental results from [26].

(c) shows the same computational results on linear timescales. Note the asymmetric Gaussian distribution towards longer residence times.

Subfigure 3.17(a) illustrates the measured ion residence time distributions from performed experiments [26] for various injected beam currents. With decreasing injection currents, effects of space charge are strongly reduced. The best approximation for a cooler transit not influenced by these, as it is the case for the performed COMSOL simulations, are the results for 32 pA (framed in green). Under these conditions, the most probable residence time has been reported as 8.2 ms [26]. Subfigure 3.17(b) and Subfigure 3.17(c) show the results of COMSOL simulations performed with 1001 particles.

The most probable residence time in the simulations lies at about 8.2 ms. The average value is 8.3 ms, with a sigma of 2.0 ms. These characteristic values are in very good agreement with the experiments. However, differences regarding the distribution of residence times are clearly evident. While maximum and minimum values for computations are 4.6 ms and 13.4 ms respectively, the full width half maximum (FWHM) of the experimental distribution is a striking 18.5 ms [26]. Apart from this, the general shape of the simulated distribution agrees well with experimental results. Both show an asymmetric Gaussian shape with a tail towards longer residence times.

Influence of buffer gas pressure

Figure 3.18 shows the influence of the buffer gas pressure on the ion residence time distribution. Subfigure 3.18(a) illustrates experimental results from [26], Subfigure 3.18(b) depicts the output of the corresponding COMSOL simulations applying the same pressure values. To do so, the displayed inlet pressure values from Subfigure 3.18(a) have been converted into mass fluxes using the conversion parameters given in Table 3.2.

Experiments as well as performed simulations both show an expected decrease in residence times for decreasing buffer gas pressure. As a result, also the distributions become narrower. Direct comparison between experimental and computational results are not easily possible, due to the injection current of 3.2 nA in the experiment [26]. These currents cause space charge effects leading to a strong decrease in residence times (see Subfigure 3.17(a)), a feature the current COMSOL model does not cover. However, when observing the relative decline of the most probable residence times relative to the value at highest pressure, the computational model seems to describe experimental values quite well. Such a comparison is made in Table 3.9.

Table 3.9: Comparison of relative decline in residence time t_r with decreasing buffer gas pressure relative to the residence times at 0.3 mbar. Experimental peak values have been estimated roughly based on Subfigure 3.17(a). For the simulation results the most probable as well as the average residence times (including the respective sigma) have been analyzed.

Pressure [mbar]	Experiment [26]		COMSOL simulation			
	Peak t_r (estimated) [ms]	Decline [%]	Peak t_r [ms]	Decline [%]	Average t_r [ms]	Decline [%]
0.30	2.0	0	8.2	0	8.3 ± 1.9	0
0.20	1.1	45	4.7	43	4.6 ± 1.1	45
0.14	0.4	80	2.2	74	2.3 ± 0.6	73

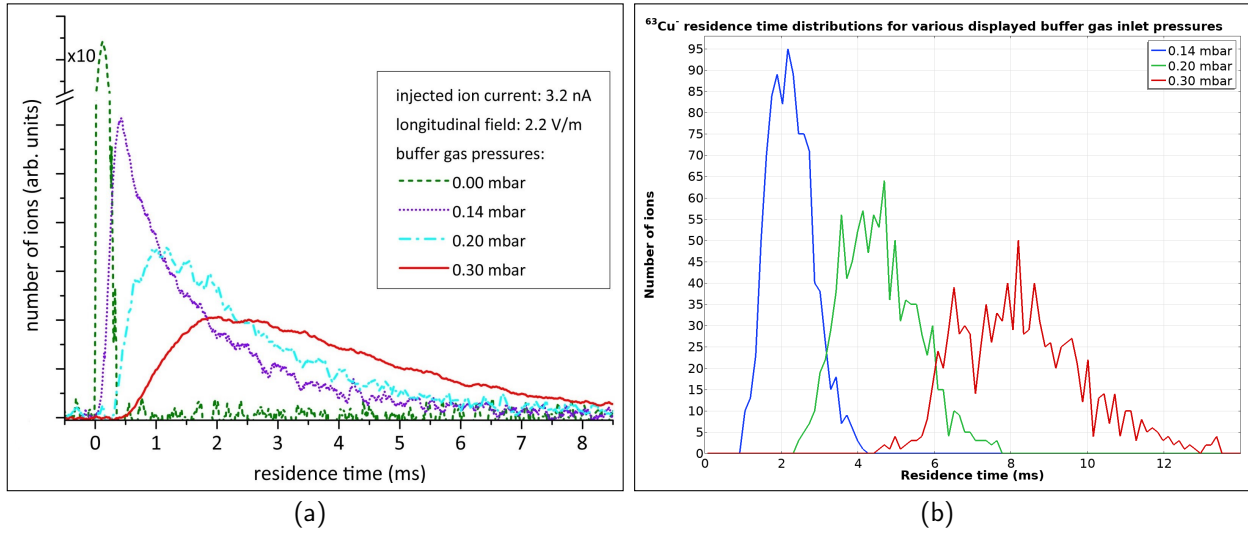


Figure 3.18: Influence of the buffer gas pressure on the residence time distributions of $^{63}\text{Cu}^-$ ions through the RFQ cooler.

(a) shows experimental results for the residence time distributions for the displayed inlet buffer gas pressures at an injected beam current of 3.2 nA (Adapted from [26]).

(b) illustrates the COMSOL results for the displayed inlet pressures 0.14 mbar, 0.20 mbar and 0.30 mbar. Pressure values have been chosen to match experimental conditions in [26] and converted into mass flux values according to conversion parameters given in Table 3.2 Cooler operational conditions for these simulations were identical to the experiments in (a).

Table 3.9 shows the relative decrease in residence time when going to lower buffer gas pressures. Although absolute values of the simulation output are quite different from experimental results, the relative decline agrees very well. This indicates a high accuracy of the COMSOL predictions regarding low current ion beams, where space charge effects do not arise to a great extent. Future implementation of a realistic space charge contribution into the existing framework should therefore be able to also describe typical experiments at beam currents of several hundred nA.

Influence of the guiding field strength

Figure 3.19 shows the experimentally observed and computationally predicted influence of the guiding field strength on the ion residence times distribution. Subfigure 3.19(a) illustrates experimental observations by Martschini et al. 2017 [26] at a beam current of 3.2 nA and a displayed inlet pressure of 0.30 mbar. For these experiments, the ion beam has been chopped to allow for the measurement of residence times. The terms rising and falling edge refer to the measurement of the beginning or the end of the chopped ion beam respectively. As illustrated for the falling edge, a strong increase in the guiding field strength merely narrows the peak. The peak residence time is unchanged, suggesting space charge effects to be the dominant influence on the motion of ions along the beam axis. As ions corresponding to the rising edge are injected into an „empty“ cooler, the initial absence of space charge effects has been proposed to be the reason for the increase in residence time [26].

3. Methods and Results

Subfigure 3.19(b) shows COMSOL results under same conditions as in [26], with an additional result for 6.5 V/m to cover the wide margin of experimentally applied guiding field strengths. Equation 3.14 has been used to choose the voltage to apply on the guiding electrodes in simulations.

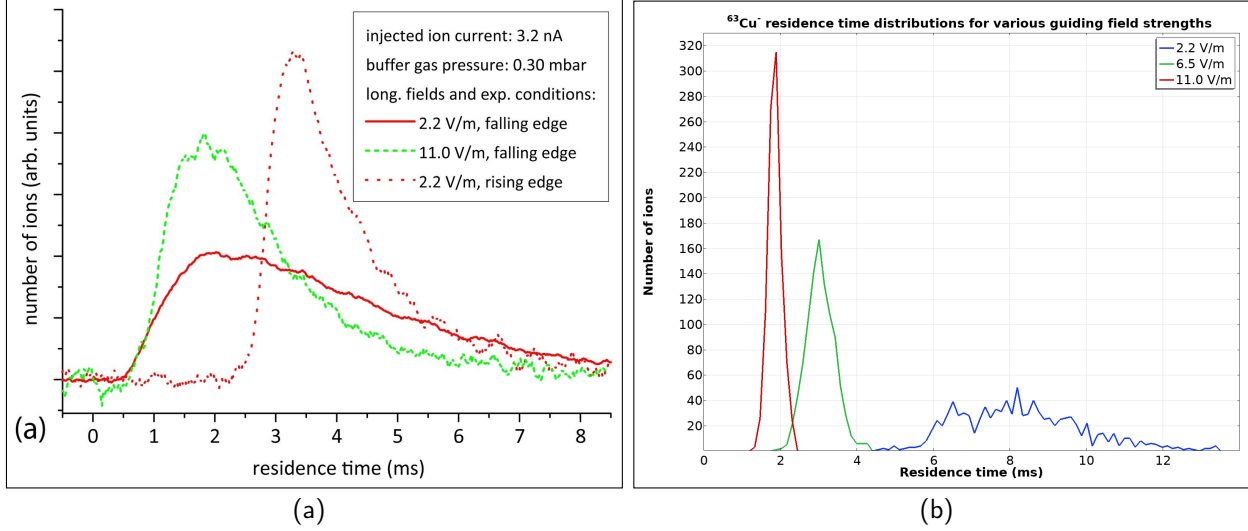


Figure 3.19: Influence of the guiding field strength on the residence time distributions of $^{63}\text{Cu}^-$ ions through the RFQ cooler. (a) shows experimental results for the residence time distributions for the displayed guiding field strengths at an injected beam current of 3.2 nA and a buffer gas pressure of 0.30 mbar (adapted from [26]). The falling and rising edge refer to the measurement of the beginning and ending of a chopped ion beam respectively. (b) illustrates the COMSOL results for guiding field strengths of 2.2 V/m, 6.5 V/m and 11.0 V/m. Guiding field strengths have been chosen to match experimental conditions in [26]. An additional simulation applying 6.5 V/m has been performed. Cooler operational conditions for these simulations were identical to the experiments in (a).

As the model does not include the simulation of space charge effects, residence times differ strongly from experimental results. The rate of decrease in average residence time in the model is almost equal to the rate of increase of the guiding field strength. Regarding the COMSOL results, a guiding field strength five times as strong is necessary to imitate space charge effects present at a beam current of 3.2 nA. The peak and average residence time values of the simulations are summarized in Table 3.10.

Table 3.10: COMSOL Peak and average values (with corresponding sigma) of the ion residence time t_r in the RFQ cooler for different applied guiding field strengths.

Guiding field strength [V/m]	2.2	6.5	11.0
Peak t_r [ms]	8.2	3.0	1.9
Average t_r [ms]	8.3 ± 2.0	3.1 ± 0.5	1.9 ± 0.4

3.6.5 Comparison of $^{36}\text{Cl}^-$ simulations with former computational models

A former work by Johanna Pitters (2015, [15]) investigated the RFQ ion cooler while still at an independent test bench. In the frame of her work, simulations using the software SIMION ([17]) were performed, including computations on the behavior of $^{36}\text{Cl}^-$ isotopes within the RFQ cooler using a HS scattering model. This allows for comparison between the presented COMSOL model and previous assumptions regarding the elastic collision theory as well as the pressure distribution inside the RFQ cooler.

A crucial problem at that time was the lack of knowledge about the pressure distribution inside the RFQ cooler. For the SIMION model, a uniform pressure equal to the displayed value on the inlet gauge was assumed [15]. The gas dynamics simulations described in Section 3.4 based on experimental results summarized in Section 3.3 strongly disagree with this assumption, as buffer gas pressure values inside the cooler are only in the range of several Pa for typical inlet pressures. Simulations also showed that the pressure distribution is not uniform, but rather step wise declining from the cooler center towards its openings. A strong deviation between simulation results obtained with the SIMION and the generated COMSOL results have therefore been expected. Especially because the VHS collision model tends to be far more realistic than the HS-model [33].

3.6.5.1 Settings and boundary conditions

COMSOL simulations on $^{36}\text{Cl}^-$ ions have been performed using similar settings as in [15]. Operational conditions are summarized in Table 3.11, the applied electrode voltages are given in Table 3.12.

Table 3.11: Operational conditions used for SIMION simulations of $^{36}\text{Cl}^-$ ions as described in [15].

Ion	Mass	Charge state	Injection energy	Mathieu parameter	RF-frequency
^{36}Cl	36 [u]	-1	50 [eV]	0.42	2.62 [MHz]

Table 3.12: Electrode voltages as used for SIMION simulations of $^{36}\text{Cl}^-$ ions through the RFQ cooler. Values are given with respect to the cooler-HV, which is considered as ground voltage. Entries marked with * have not been included in SIMION simulations performed in [15]. For COMSOL simulations they have been considered in order to account for proper focusing during the injection process. The voltage specified for the RF-electrode represents the amplitude of the periodic signal. The applied guiding voltage corresponds to a guiding field strength of about 2.88 V/m.

Electrode	INJLO*	EXTLO*	Guiding	RF-amplitude	Apertures	Cooler tube
Applied voltage [V]	940	400	-50	200	0	0

SIMION simulations have been performed with a uniform buffer gas pressure of 0.01 mbar, 0.05 mbar and 0.10 mbar. Typical operational inlet pressure values in the range of 0.20 mbar - 0.30 mbar have been omitted [15]. To match these conditions, two COMSOL simulations at displayed inlet pressures of 0.1246 mbar and 0.4423 mbar have been performed. According to the gas dynamics simulations in Section 3.4, these inlet values correspond to average buffer gas pressures inside the cooler of 0.01 mbar and 0.05 mbar, respectively. This allows for direct comparison between HS (SIMION model) and VHS (current COMSOL model) elastic collision model. An average buffer gas pressure inside the cooler of 0.10 mbar, also performed with SIMION, would correspond to a displayed inlet pressure of 0.84 mbar in the current COMSOL model. This is far beyond typical operation conditions at ILIAMS. Simulations in this region have therefore been omitted. To cover the whole operational range, presented results also include COMSOL simulations at a displayed pressure of 0.10 mbar, 0.20 mbar and 0.30 mbar respectively. All simulations have been done with He.

3.6.5.2 Results

Thermal conditions and cooling parameters

Typical injection energies into the RFQ cooler are in the range of some tenths of eV. After injection, the ions rapidly lose their kinetic energy through collisions with the background buffer gas until they reach a stable equilibrium state. In the absence of electric fields, this thermal energy would equal that of the buffer gas. For an ideal gas, this energy is at about 39 meV at 300 K. However, SIMION simulations already showed that the thermal energy for ions inside the RFQ cooler is increased due to their constant energy gain from the RF trapping field [15]. The average time after injection necessary to reach thermal equilibrium is called *cooling time*. The average distance into the RFQ cooler covered during this time span is the *cooling length*. Both are important parameters for the cooling behavior of the RFQ cooler.

Figure 3.20 illustrates this process for performed COMSOL simulations at different displayed inlet pressures and a guiding field strength of 2.88 V/m. Shown are the average kinetic energies (Subfigure 3.20(a)) and progression distances (Subfigure 3.20(b)) for $^{36}\text{Cl}^-$ ions during their first 300 μs inside the RFQ cooler. A great distance through the cooler is already covered during the first few μs until a level of thermal energy is reached. After this cooling length/time the rate of progression is steady and changes with buffer gas pressure.

To obtain the thermal energies occupied at the respective inlet pressures, the average over the whole range of residence time excluding the first 500 μs has been used. Cooling time and length are then given by the first time thermal conditions are reached. The corresponding numerical values for all performed simulations are summarized and compared to SIMION results by Johanna Pitters 2015 [15] in Table 3.13. SIMION results are marked with * and included for simulations, where pressure conditions are most comparable.

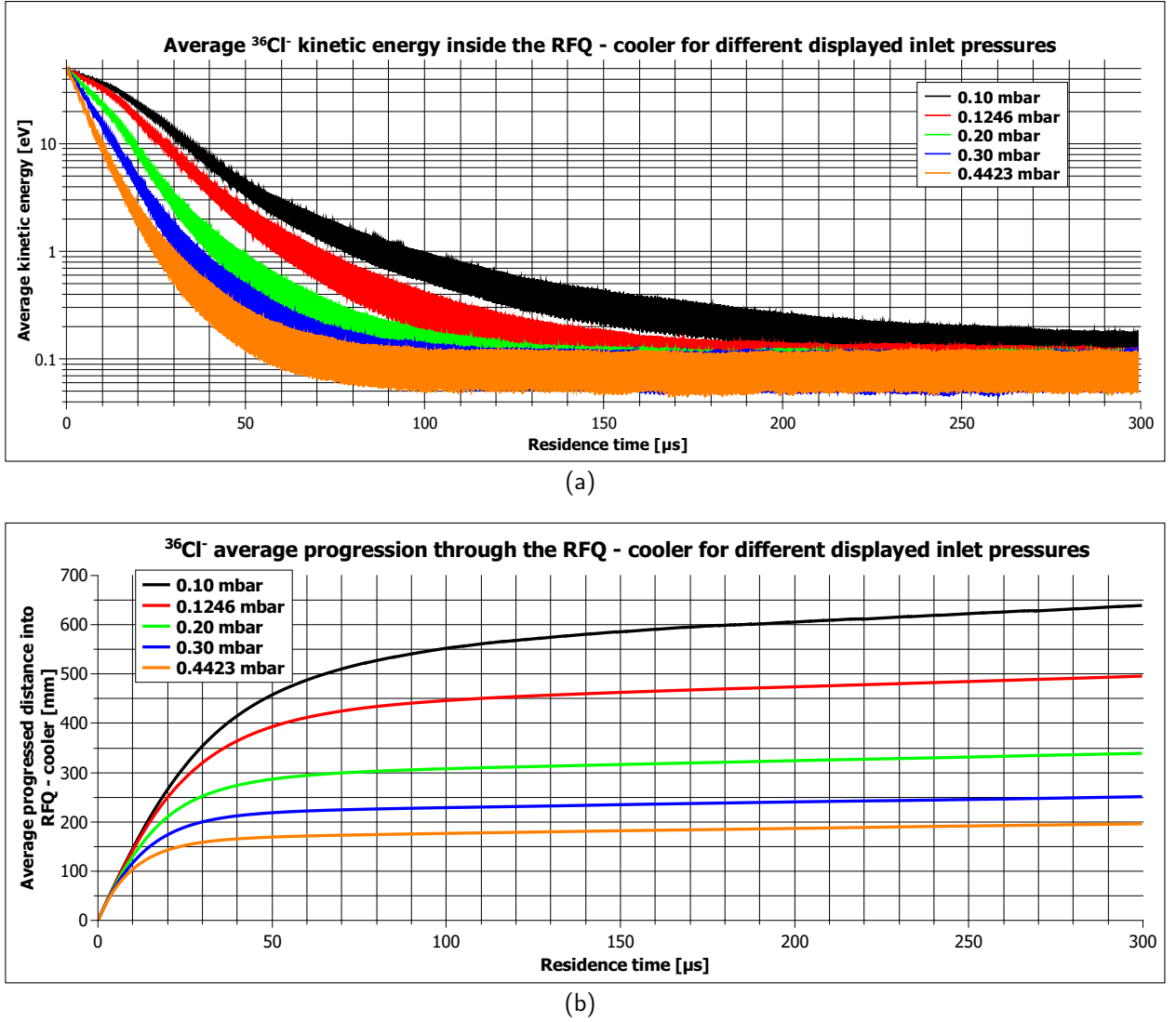


Figure 3.20: Behavior of $^{36}\text{Cl}^-$ ions inside the RFQ cooler during their first 300 μs for different displayed inlet pressures. (a) shows the average kinetic energies. The broad distribution of the average energy is due to the periodic nature of the RF trapping field. Note the logarithmic energy scale. (b) displays the average progression distance into the cooler as a function of the residence time.

While the SIMION model states a constant thermal energy of 80 meV [15], COMSOL results suggest different equilibrium energies depending on the buffer gas pressure. These are illustrated in Subfigure 3.21(a) and included in Table 3.13. Displayed inlet pressures above 0.20 mbar yield a thermal equilibrium energy of about 83 meV. This is in very good agreement to the SIMION simulations. However, when decreasing the inlet pressure the mean time between collisions increases. Therefore, ions are able to gain more energy from the RF-field before eventually returning it to a gas particle allowing them to fluctuate around up to 134 meV for a displayed inlet pressure of 0.10 mbar. The on-axis distance of the ions behaves according to the thermal energy. While the SIMION model yields a mean distance of about 500 μm , obtained values from COMSOL simulations given in Table 3.13 and illustrated in Subfigure 3.21(b) yield much lower values on the order of 152 μm .

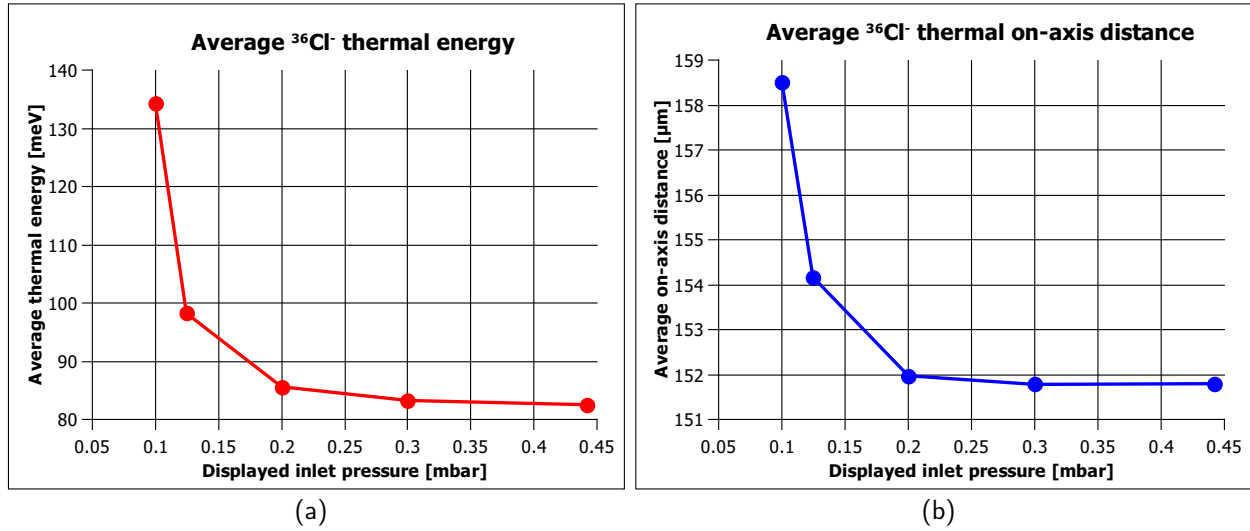


Figure 3.21: Influence of the displayed inlet pressure on the thermal equilibrium conditions of $^{36}\text{Cl}^-$ ions inside the RFQ cooler. (a) displays the average kinetic energies (in meV) at which the ions are in thermal equilibrium with the buffer gas (He) as a function of the displayed inlet pressure. (b) shows the average on-axis distance (in μm) of the $^{36}\text{Cl}^-$ ions at thermal equilibrium as a function of the displayed inlet pressure. Standard errors are about 0.1% in magnitude and therefore not included.

Comparison with SIMION results from [15] in Table 3.13 are given at displayed inlet pressure values which represent an average pressure inside the cooler equal to the uniform pressure used for SIMION simulations. For simulations at 0.1246 mbar, the COMSOL cooling length and time are about 60% - 70% lower than for the SIMION model. However, at a slightly higher pressure of 0.4423 mbar, the COMSOL model predicts a higher cooling length and time.

Table 3.13: Summary of relevant values regarding the cooling conditions of $^{36}\text{Cl}^-$ ions passing the RFQ cooler for all simulated displayed inlet pressures. Injection energy and guiding field strength have been 50 eV and 2.88 V/m respectively. Given are the average kinetic energy and distance to the beam axis after reaching thermal equilibrium, as well as the average cooling time and length necessary for an ion to reach equilibrium. Given uncertainties correspond to the standard error.

Values marked with * refer to SIMION results and have been taken from [15]. These values are compared with COMSOL results for which the resulting average pressure inside the cooler equals the uniform pressure assumed for the SIMION model. SIMION simulations performed at a uniform pressure of 0.10 mbar correspond to a displayed inlet pressure value of 0.84 mbar in the case of the current COMSOL model. No COMSOL simulation has been performed at this pressure as it strongly exceeds usual operation conditions.

Displayed inlet pressure [mbar]	Cooling length [mm]		Cooling time [μs]		Thermal energy [meV]	Thermal on-axis distance [μm]
0.10	625	-	280	-	134.23 ± 0.05	158.52 ± 0.26
0.1246	475	$>800^*$	200	$>120^*$	98.22 ± 0.14	154.17 ± 0.02
0.20	325	-	180	-	85.61 ± 0.07	151.97 ± 0.01
0.30	225	-	100	-	83.26 ± 0.05	151.80 ± 0.01
0.4423	175	186^*	90	47^*	82.52 ± 0.04	151.78 ± 0.01
0.84	-	92^*	-	22^*	-	-

The cooling behavior through RFQ the cooler is further visualized in Figure 3.22. It shows the progression distribution of simultaneously injected $^{36}\text{Cl}^-$ ions for various residence times at a displayed inlet pressure of 0.20 mbar. After only 10 μs , the average progression distance is already at 128 mm, with a narrow distribution due to the high kinetic energies. The cooling length for this scenario is about 325 mm, with a cooling time of 180 μs (see Table 3.13). At this time, the distribution is much broader, as random collisions with the buffer gas can force ions towards the cooler entrance and exit respectively. After reaching thermal energy, the progression rate is approximately constant at about 115 mm/ms. Although the average progression after 1 ms is already at about 435 mm and thus almost half the cooler length, the overall residence time for this case yields 4.42 ms.

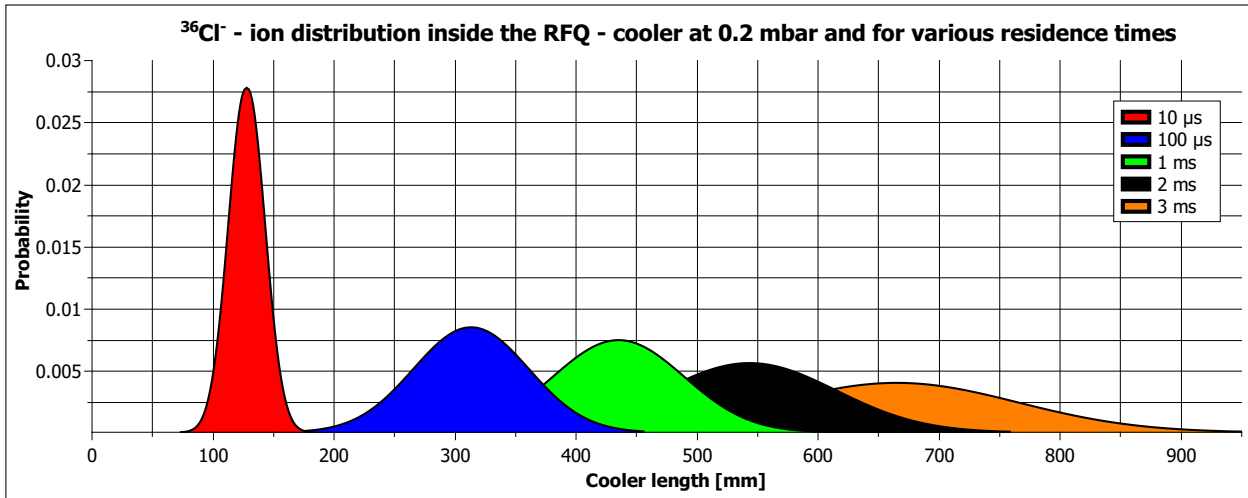


Figure 3.22: Evolution of a $^{36}\text{Cl}^-$ ion beam simultaneously injected into the RFQ cooler at a displayed inlet pressure of 0.20 mbar. Shown are the distributions of the progression distance into the cooler at various residence times. Gaussian functions have been assumed and are based on the average values of progression distance and its standard deviation.

COMSOL results for cooling lengths and times are in the same order of magnitude as SIMION results at comparable buffer gas pressures inside the cooler. On the first sight, this is rather surprising, as the VHS collisional cross section used for COMSOL simulations is much higher the constant HS cross section used in the SIMION model. One may expect shorter cooling times when using the VHS-model. However, the non-uniform buffer gas distribution used for COMSOL simulations (see Figure 3.8) includes a region of low gas density right after injection, allowing ions to penetrate deeper into the cooler.

The two models also disagree regarding the average on-axis distance. While SIMION simulations obtained a mean distance of 500 μm , the current COMSOL model only predicts about 150 μm at usual operational inlet pressures. Most likely, this is a consequence of the high value for the VHS collisional cross section at thermal energy with respect to the constant HS cross section used for the SIMION model (see Figure 3.16).

Residence times

Table 3.14 shows the computed residence times of the SIMION model [15] as well as the results from the COMSOL model developed in the frame of this master's thesis. Pressure values for the SIMION model refer to a uniform pressure assumed inside the cooler. In the case of the COMSOL model, pressure is given as the displayed inlet pressure. The corresponding average buffer gas pressure inside the cooler has been interpolated from gas dynamics simulation data given in Table 3.3. Figure 3.23 shows the distribution of the computed residence times for all simulated displayed inlet pressures.

Table 3.14: Computed residence times of $^{36}\text{Cl}^-$ ions passing the RFQ negative ion cooler. Results of former SIMION computations performed by Johanna Pitters (2015 [15]) are compared with those of the COMSOL model developed in the scope of this master's thesis. The uniformly assumed buffer gas pressure for the SIMION model as well as the resulting residence times have been taken from [15]. COMSOL simulations have been performed for five different displayed inlet pressures corresponding to typical experimental settings. The corresponding average pressure inside the RFQ cooler are based on gas dynamics simulation results given in Table 3.3. t_{COMSOL} represents the average of resulting residence times. Given uncertainties represent the corresponding sigma.

SIMION [15]		COMSOL		
Uniform pressure [mbar]	t_{SIMION} [ms]	Displayed inlet pressure [mbar]	Corresponding average pressure [mbar]	t_{COMSOL} [ms]
-	-	0.10	0.007	0.89 ± 0.37
0.01	1.2 ± 0.2	0.1246	0.010	1.77 ± 0.47
-	-	0.20	0.020	4.42 ± 0.88
-	-	0.30	0.032	7.9 ± 1.5
0.05	5.7 ± 0.8	0.4423	0.050	12.6 ± 2.4
0.10	11.0 ± 2.0	0.84	0.100	-

The increase in residence times with increasing displayed inlet pressure as well as the average residence times are similar to the results of $^{63}\text{Cu}^-$ ions given in subsection 3.6.4.2. However, when considering that the VHS collisional cross section of $^{36}\text{Cl}^-$ surpasses that of $^{63}\text{Cu}^-$ (see Figure 3.16) for kinetic energies below 1 eV, one would expect the final residence times for $^{36}\text{Cl}^-$ ions to be higher. Figure 3.24 compares both simulations at the same buffer gas pressure of 0.30 mbar by their average kinetic energy during the first 250 μs and their average progressed distance into the cooler for a residence time of 5 ms. Subfigure 3.24(a) shows that the energy decline for $^{36}\text{Cl}^-$ ions is significantly faster than for $^{63}\text{Cu}^-$, as one would expect regarding their respective VHS cross sections. As a direct consequence, progression into the cooler is faster for $^{63}\text{Cu}^-$ at first, as illustrated in Subfigure 3.24(b). However, after about 3 ms $^{36}\text{Cl}^-$ progresses faster, eventually traversing at lower total residence times. It is strongly suggested that the reason lies within the different velocities of the two isotopes at same thermal energy. These are about 346 m/s and 457 m/s for $^{63}\text{Cu}^-$ and $^{36}\text{Cl}^-$ respectively. Furthermore, the guiding field strength in the case of $^{36}\text{Cl}^-$ simulations has been about 30% higher.

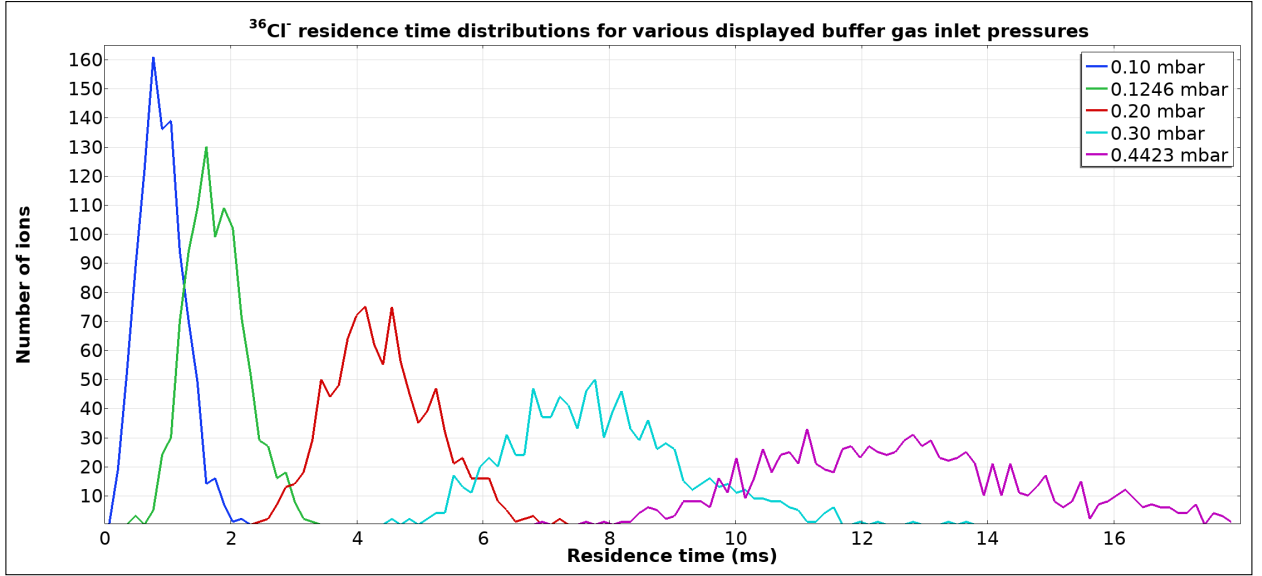


Figure 3.23: COMSOL residence time distributions for $^{36}\text{Cl}^-$ ions passing through the RFQ cooler at 50 eV injection energy and 2.88 V/m guiding field strength. The results for simulations at various displayed inlet pressures are shown.

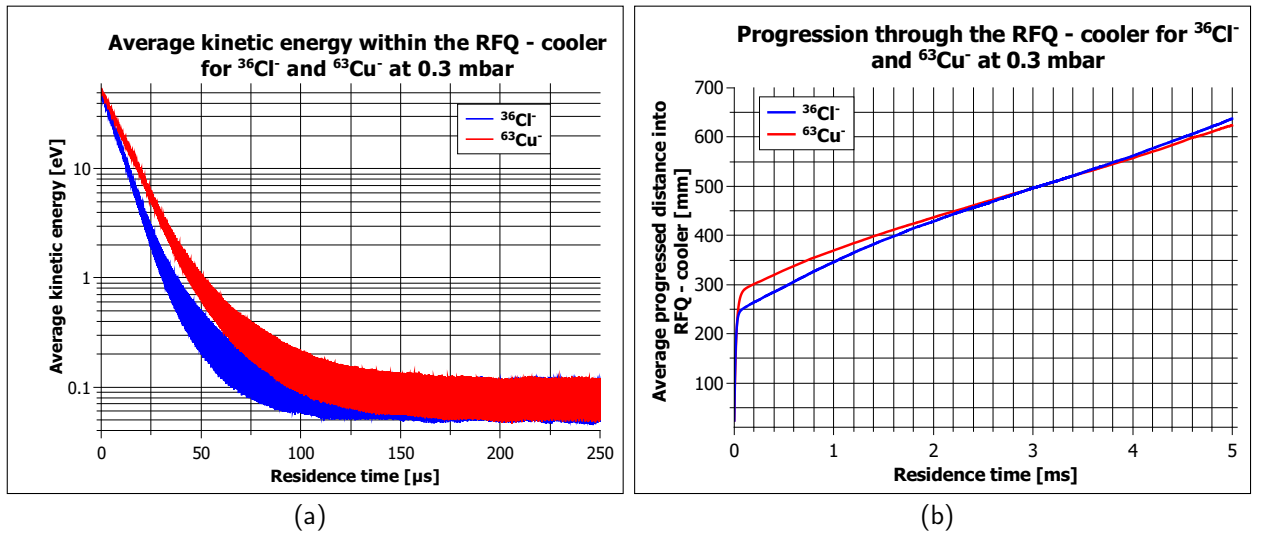


Figure 3.24: Comparison of the behavior of $^{36}\text{Cl}^-$ (blue) and $^{63}\text{Cu}^-$ (red) ions within the cooler at equal buffer gas pressure of 0.30 mbar. (a) shows the average energy for both ions during the first 250 μs . Note the logarithmic energy scale. (b) displays the average progression distance into the cooler within the first 5 ms. The progression through the cooler is not constant due to the varying buffer gas pressure inside the cooler. Lower pressure allows for a slightly higher thermal energy of the ions. Additionally the VHS collisional cross section decreases with increasing ion energy.

Simulations with buffer gas pressures of 0.1246 mbar and 0.4423 have been chosen such that the average pressure inside the cooler matches 0.01 mbar and 0.05 mbar, for which SIMION simulations have been performed. This makes them the most convenient for comparison between the HS and VHS elastic collision models. At a displayed inlet pressure of 0.1246 mbar, the COMSOL

results for the average residence time is about 25% below the SIMION result and in reasonable agreement. However, for a higher pressure of 0.4423 mbar, the COMSOL results predict an average residence time of 12.6 ms, comparing to a SIMION result of 5.7 ms. Even for a uniform pressure of 0.10 mbar inside the cooler, corresponding to a displayed inlet pressure of 0.84 mbar in the case of the COMSOL model, the predicted SIMION residence time is at only 11.0 ms. The predicted residence times for the COMSOL model at typical displayed inlet pressures are therefore higher than SIMION predictions, even for inlet pressures that surpass these typical conditions by a high margin.

There are several reasons for the diverging results. Although inlet pressures for COMSOL simulations have been chosen to roughly match the pressure assumptions of the SIMION model, the latter used a uniform distribution inside the cooler. While the average pressure inside the cooler has been comparable for the discussed cases, the step wise buffer gas distribution used for COMSOL simulations would lead to much higher cooling lengths, provided all other settings are equal. This effect would be intensified by the low VHS collisional cross section during the cooling process down to about 3 eV compared to the HS-model. On the other hand, the VHS cross section is about four times as high as for the HS-model in the range of thermal ion energies, increasing residence time inside the latter part of the cooler with respect to the SIMION model.

3.7 Space charge calculation

As mentioned in Section 2.5 and visible in Subfigure 3.17(a), the influence of space charge effects is not negligible. Already at injection currents in the range of nA, residence times decrease drastically, while beam currents exiting the ion source are typically in the range of a few μA . A computational model capable of predicting such effects would be of great interest.

Only speculations about the way and the structure of a space charge field inside the RFQ cooler have been possible. Predictions have been made after examining COMSOL simulation results, especially regarding the progression of ions after injection into the cooler (see Subfigure 3.20(b)). The Bragg-like stopping through the buffer gas creates regions of different ion densities. A very thinly occupied deceleration passage is followed by a high density region where ions reach thermal energies. Such a gradient is expected to create a potential barrier with a height dependent on the injected beam current. Ions that are energetic enough to pass this barrier would experience a force accelerating them towards the cooler exit. As simulations with different guiding field strengths already show, such a static field can cause residence times to increase significantly (see Figure 3.19). At the same time, ions of lower kinetic energy can be stopped at the barrier and pushed away from its center.

Experimental results by Martschini et al. 2017 [26] can be used to compare the output of such a model. An attempt has been made to realize this, using the COMSOL *Charged Particle Tracing* interface [32] in combination with the *Optimization Module* [44].

3.7.1 Computational approach

The most practical method to simulate an occurring space charge would be to model the interaction between the single ions using Equation 2.20. This would require summing up the forces of all other particles acting on a single particle. As this needs to be done for each particle, computation costs increase quadratically. For an experiment performed at 0.30 mbar buffer gas pressure and 3.2 nA beam current, the number of ions inside the cooler is about $4 \cdot 10^7$ particles. Even when approximating such a scenario by only using 1000 ions, computation time becomes far too long and available RAM would be exceeded easily.

Therefore a step wise approach had been attempted. COMSOL is able to track the time that a modeled ion spends within every mesh element. One can assign a certain fraction $f_{current}$ of an injection current I_{beam} to each model particle corresponding to

$$f_{current} = \frac{I_{beam}}{N} \quad (3.30)$$

where N denotes the number of particles included in the simulation. By summing up all distinct occupation times t_{occ} , a total space charge Q_{SC} can be assigned to each mesh element using

$$Q_{SC} = T_{occ} \cdot f_{current} \quad (3.31)$$

$$T_{occ} = \sum^N t_{occ}$$

The related charge density can then be obtained via dividing Q_{SC} by the volume of the corresponding mesh element. The COMSOL *Electrostatics interface* offers a node to generate the resulting electrostatic field. The modeling approach for presented simulations can be summarized as:

1. Calculate the initial electrostatic field.
2. Calculate the RF-field used for ion simulations.
3. Perform ion simulations at a certain beam current. Use the mesh tracking feature of COMSOL to gather a space charge density.
4. Generate a new static electric field, now including a space charge field based on the information from 3.
5. Repeat step 3 and 4 until the changes in the static field and the resulting residence times are sufficiently small.

The utilized modeling geometries have not been changed and are described in subsection 3.5.1 and subsection 3.6.1.

3.7.2 Settings and boundary conditions

In addition to former particle simulations, a *Space Charge Density* node performing the space charge accumulation during simulation had to be applied. Due to the additional operations, the RAM requirements increased strongly. Therefore, the mesh quality had to be reduced from $2.6 \cdot 10^6$ to $7 \cdot 10^5$ and from $1.3 \cdot 10^6$ to $5.9 \cdot 10^5$ mesh elements for the whole geometry and the particle geometry respectively.

Unfortunately this has caused high gradients in the generated space charge between mesh elements, strongly distorting remaining ion simulations. A workaround has been developed to overcome this problem:

- Via two projection operators, the cross sectional charge density is integrated onto the beam axis. The ion density along the cooler is therefore expressed in one dimension along the beam axis.
- The resulting distribution is scaled by division through the line density ρ_l (see Section 2.5).
- Using the COMSOL *Optimization module*, this distribution is fitted with a suitable function (usually Gaussian, skewed Gaussian or combinations).
- The radial ion density distribution is approximated using Equation 2.21 and Equation 2.22. In combination with the total ion distribution along the beam axis, the space charge distribution within the cooler can be inserted as a coordinate dependent function. The function is passed to the *Electrostatics module* generating the corresponding static space charge field.

As an example the result of such a fitting process for a projected and scaled ion line density is given in Figure 3.25. Early simulations had the attempt to directly approximate the space charge distribution at beam currents in the range of some nA. However, the resulting potential barrier after the initial run was too high to overcome for any ions of the following simulation. Therefore an initial beam current of 100 pA has been chosen. After reaching a stable solution for the space charge distribution, the intention had been to scale it and use as initial run at a higher beam current. In addition to the step wise approach regarding the space charge itself, also the beam current had to be handled similarly. A number of 2001 simulated ions has been used for the initial simulation as well as the first two iterations. Due to extremely high computation times, 1001 ions have been used for the following iterations.

Simulations have been performed with $^{63}\text{Cu}^-$ ions, He as buffer gas and at a displayed inlet pressure of 0.30 mbar. Electrode voltages have been chosen the same as in former simulations and can be seen in Table 3.7. The initial beam current has been chosen as 100 pA, as the accumulated space charge potential barrier after the first iteration has been low enough for ions to surpass.

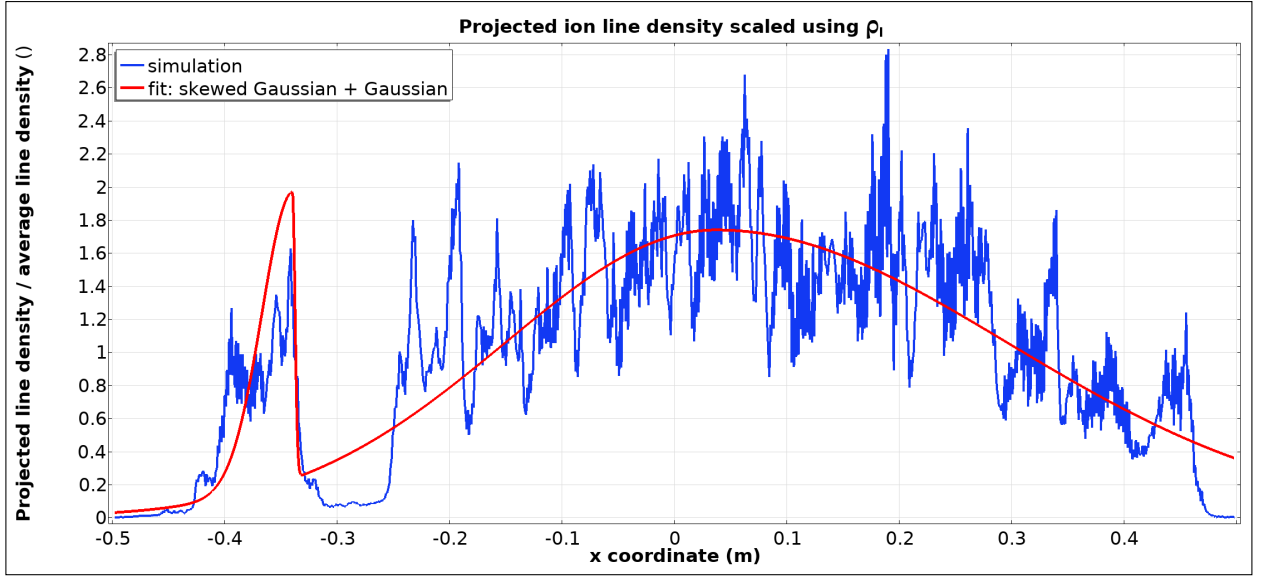


Figure 3.25: Example of a generated fit using the COMSOL *Optimization Module*. The projected ion line density scaled using the average line density ρ_l is given in blue. The corresponding fit, including a skewed Gaussian and a Gaussian function, is shown in red. The x-scale is chosen in m to allow for easy processing. The origin is located at the tube center.

3.7.3 Results

The simulation sequence has been cancelled after the fifth iteration and at a beam current of 200 pA. Although results have indicated a trend towards a stable solution, the last iteration has generated a space charge diverging from that trend.

Figure 3.26 shows the residence time distributions for all performed simulations. Results are given separately for the two simulated beam currents to allow for better differentiation. The corresponding average and peak residence times are summarized in Table 3.15. As can be seen in Subfigure 3.26(a), residence time distributions for the initial simulation and iterations I_1 and I_2 are very similar to each other as well as to the initial run. Although their peak residence times differ, the corresponding average residence times are in very good agreement. Furthermore, their respective space charge configurations have been conform as well. Therefore, results from the second iteration have been considered as sufficient to progress to higher beam currents via scaling. For a setting of 200 pA, three iterations have been performed before ending the sequence.

Table 3.15: Average and peak residence times t_r for all performed iterations of the space charge simulation sequence. The initial simulation not considering any space charge contribution is denoted by I_0 . Given uncertainties represent the corresponding sigma.

Beam current	100 [pA]			200 [pA]		
Iteration	I_0	I_1	I_2	I_3	I_4	I_5
Average t_r [ms]	10.1 ± 3.4	10.1 ± 3.7	10.0 ± 2.3	11.2 ± 3.9	9.5 ± 5.4	8.1 ± 2.6
Peak t_r [ms]	9.0	10.2	9.5	8.6	9.0	8.2

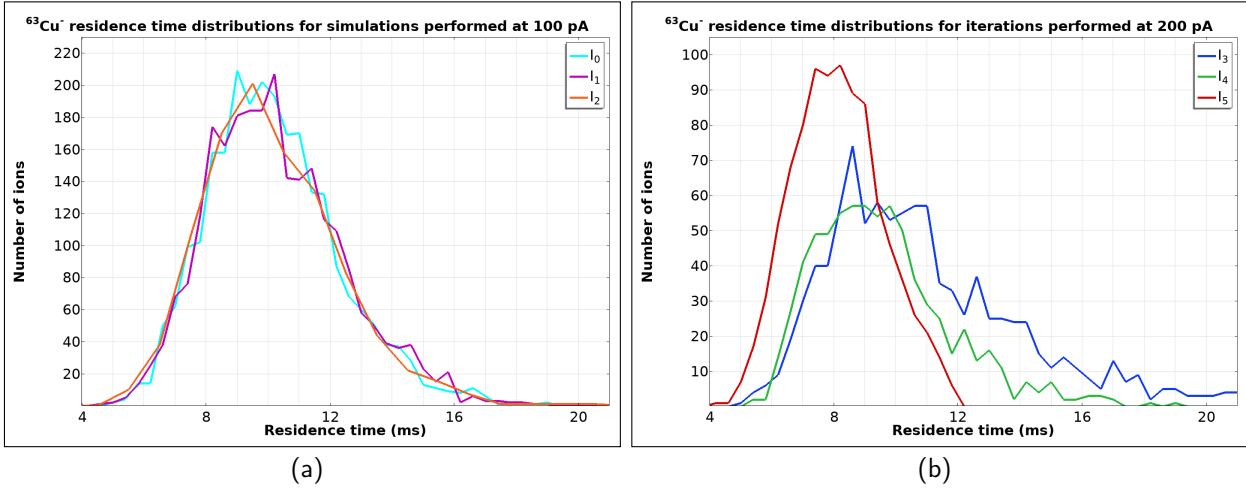


Figure 3.26: Residence time distributions for $^{63}\text{Cu}^-$ ions at a He buffer gas pressure of 0.3 mbar for different iterations i and beam currents.

(a) shows the COMSOL results for the initial simulation as well as two iterations at 100 pA beam current and for a number of 2001 ions each. (b) gives the results for a beam current of 200 pA and a total number of ions of 1001. Different scales are the results of the varying number of simulated ions.

Figure 3.27 and Figure 3.28 show the resulting space charge and total potentials respectively for iterations $I_2 - I_5$. The total static potential is a superposition of the guiding potential and the space charge. Colors have been chosen such that residence time distributions can be connected to their underlying potential, which has been generated by the result of the previous iteration. As the graphics illustrate, results for $I_2 - I_4$ show an expected behavior. Already at I_2 , ions are accumulating at about 250 mm into the cooler, where they reach thermal temperatures. This creates a small potential barrier (blue in Figure 3.27) of about -0.14 V. In the case of I_3 , a fraction of injected ions has lost too much energy before that point to overcome this barrier. They are „collected“ in front of it and begin to bounce. Pushed by the guiding field and hindered by the space charge potential, they create a high peak of space charge potential (green) of about -0.37 V. Furthermore, the border of this potential barrier has shifted towards the injection side. Eventually, a random collision with a buffer gas particle transfers enough energy to overcome the potential barrier. When observing the generated space charge potential of I_3 (green in Figure 3.27) regarding the second half of the cooler, a strong decline can be seen. This indicates that ions covered this stretch in much shorter time. As the former potential (blue) decreases after about 475 mm into the cooler, particles experience a strong push. The total static field strength towards the cooler exit from there on resembles about 120% when compared to only the guiding field. Ions overcoming the potential barrier solely by means of their remaining kinetic energy therefore traverse the cooler faster. As a result, the average and peak residence times decrease compared to I_2 . However, a fraction of ions spends much more time bouncing. As can be seen in Subfigure 3.26(b), this leads to a tail towards longer residence times for I_3 when comparing to Subfigure 3.26(a).

Ions of I_4 now face an even higher barrier (green in Figure 3.27). Although the same procedure applies, gas collisions powerful enough to transfer enough energy are much more infrequent than in the case of I_3 . Particles bounce longer and a much higher (-0.65 V) and broader peak in space charge potential is generated (red). Again, the generated potential barrier has shifted strongly towards the injection side. Due to the strong push from the I_3 potential, ions surpassing the barrier are accelerated towards the cooler exit. Residence times are in the same area for I_4 as for I_3 , as illustrated in Subfigure 3.26(b). However, the average residence time increased while the tail towards longer residence times decreased.

Although the space charge potential barrier acting on ions from I_5 (red in Figure 3.27) is extremely high, only a small fraction lacks the kinetic energy to overcome it due to its short distance to the cooler entrance. Passing ions experience a strong push corresponding to a field strength of about 130% of the static guiding field and therefore traverse the cooler quickly. This can be seen by the low magnitude of the generated space charge potential (black) as well as in the strong shift towards shorter residence times (red in Subfigure 3.26(b)). Ions that did not surpass the potential barrier had no chance to overcome it afterwards, therefore are trapped and bouncing. The simulation has been stopped after remaining particles were all located before the barrier. As a result, no tail towards longer residence times appears in Subfigure 3.26(b).

Simulations up to I_4 have been able to verify the trend towards shorter residence times in the presence of space charge effects. They have confirmed speculations of a space charge potential barrier that emerges after injected ions thermalize. A growth in barrier size as well as a shift in peak position towards the cooler entrance has been observed. However, I_5 has not been able to continue this trend, let alone to yield similar results to indicate an approximation towards a stable solution. The generated potential (black in Figure 3.27) is much lower compared to all previous results. Furthermore, the created barrier potential is rather short (-0.18 V) and located already at about 150 mm into the cooler unit. An additional iteration based on such a space charge configuration yields results similar to I_2 , capturing the simulation sequence inside a loop. Therefore, the sequence has been stopped after I_5 . It can be expected that a stable solution for a space charge potential lies in between the solutions of I_4 and I_5 . Effort has been made to artificially continue the sequence by using an average distribution of I_3 , I_4 and I_5 . However, the corresponding space charge distribution has been almost equal to that of I_5 .

It has to be noted, that resulting space charge potentials illustrated in Figure 3.27 do not represent realistic charge configurations inside the RFQ cooler. Such well defined and high barriers would flatten out immediately after accumulation. Rather, the sequence can be understood as the first steps of the formation of a stationary space charge distribution, as it would appear after a beam enters an „empty“ RFQ cooler.

Unfortunately simulations have not been able to approximate such a flattened charge configuration. However, results still show that the expected formation of a space charge peak around

cooling distance is likely. Future simulations, eventually performed on more powerful hardware or with access to higher computational resources, may even be able to use the same model extended by the flattening effect. A significant increase in mesh density while keeping computation times reasonably low could enable a successful approximation of emerging space charge distributions.

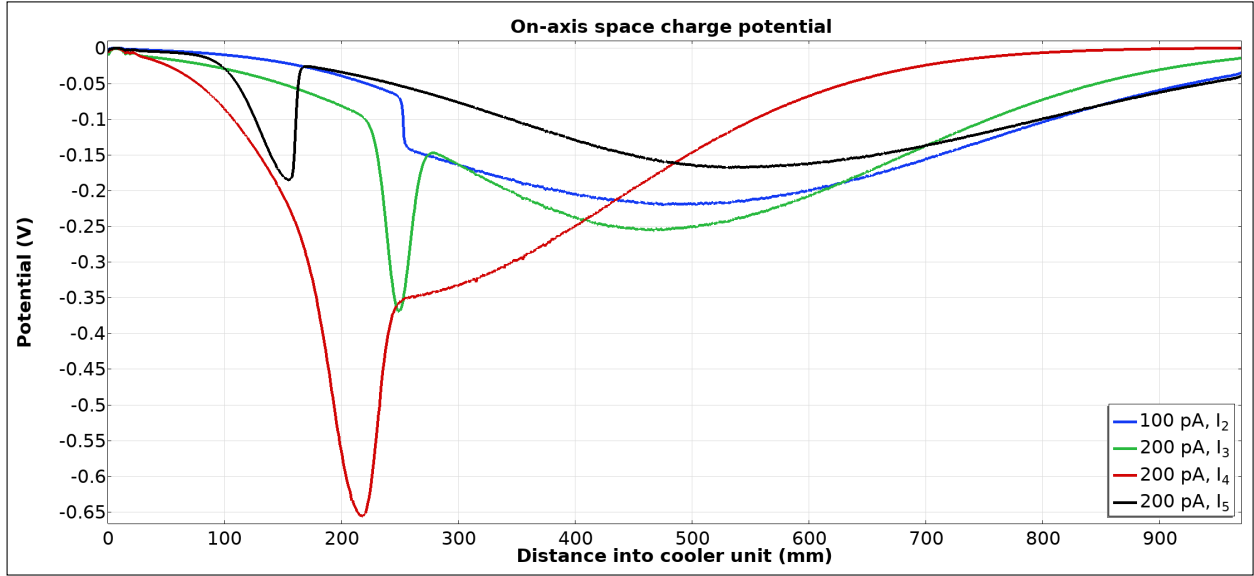


Figure 3.27: Resulting on-axis space charge potentials for iterations $I_2 - I_5$. Due to their similarity, results of the initial run as well as the first iteration have been omitted. Note that these results depict the space charge potential generated by each iteration. The result from the second iteration (blue) is already scaled for a beam current of 200 pA.

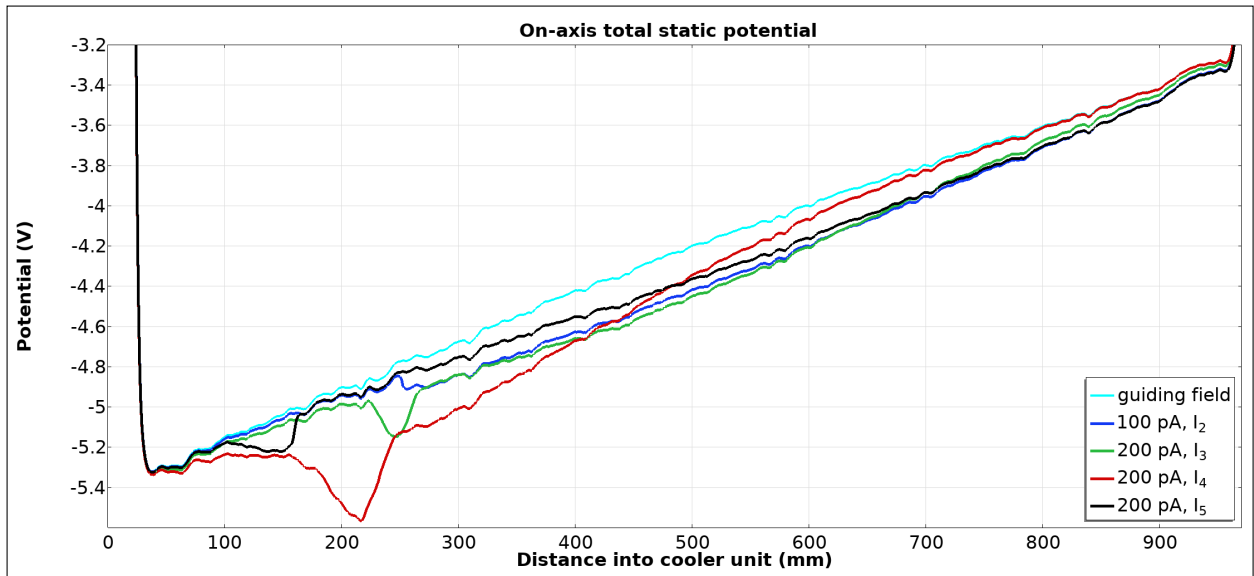


Figure 3.28: On-axis total static potential results for iterations $I_2 - I_5$. The initial run as well as the first iteration have been omitted due to similarity. The static guiding field (cyan) has been added for comparison. The total potential is composed of the guiding potential and the space charge potential as seen in Figure 3.27. Note that these results depict the static potential generated by each iteration. The result from the second iteration (blue) is already scaled for a beam current of 200 pA.

4 | Conclusion and outlook

The aim of this master's thesis has been the development of a COMSOL simulation model of the ILIAMS RFQ negative ion cooler, feasible to explain occurring effects and accurately predict residence times and the behaviour of transiting ions. This required the precise computation of different underlying physical processes, as well as their implementation into a single model. This part has been conducted successfully. Another focus has been on the investigation of occurring space charge effects. A constructed model has been able to suggest a suitable approach, but lacks at least the necessary computational resources for proper implementation.

At the beginning of this work, no information regarding the buffer gas flow inside the RFQ cooler with the exception of an inlet pressure display has been known. A performed experiment has been able to link this value to the actual mass flow into the cooler, providing reliable initial conditions for following gas dynamics simulations. Furthermore, experimental outlet pressure conditions have been obtained to verify performed simulations.

An independent gas dynamics model has been generated and constantly improved by exploiting various symmetric conditions and simplifications. Results indicate a buffer gas pressure distribution inside the RFQ cooler with pressure values of some Pa. While these are an order of magnitude below previous assumptions, they are in very good agreement with operational conditions for similar negative ion cooler devices elsewhere (Liu et al. 2005 [18], Eliades et al. 2011 [48]). In addition, a formerly underestimated effect caused by quad holders as part of the cooler geometry, leading to a step wise pressure distribution has been resolved. Comparison with mentioned experimental data suggests a high accuracy of the model in the case of the mainly used buffer gas He, while N₂ and Ar predictions diverge stronger due to a stronger deviation from the simulated flow regime. Exploitation of the underlying physics of free molecular gas flow has resulted in a mathematical formulation of the buffer gas density inside the RFQ cooler, applicable as soon as the inlet pressure is known. This allows for simple implementation of the high quality results whenever needed.

An independent model has been constructed to combine the computation of applied electric fields with the simulation of ion trajectories through the RFQ cooler. For this purpose, a VHS elastic collision model has been implemented into COMSOL by modifying existing approaches. A personalized meshing procedure has been applied to guarantee high quality of results, while keeping

computation times reasonably low. Electric fields simulations have been able to verify guiding field calculations of former works (Pitters 2015 [15]).

Using the obtained results for the driving electric fields, a series of simulation runs with different operational settings on $^{63}\text{Cu}^-$ ions has been performed to compare computations with available experimental data (Martschini et al. 2019 [26]). Predictions by the COMSOL model regarding the residence time have shown to agree extremely well when compared to experiments at low beam currents where space charge effects are negligible. For higher beam currents in the nA range, the model currently is not able to compute space charge effects. Additional simulations on $^{36}\text{Cl}^-$ ions have shown significant improvements with respect to former SIMION models based on outdated assumptions about the buffer gas conditions (Pitters 2015 [15]).

In the final step of this master's thesis, efforts have been made to construct an iterative approach to reproduce the creation and manifestation of stable space charge distributions within the RFQ cooler. Unfortunately, stationary solutions have not been possible due to the restriction of accessible computation power. Nevertheless, results still offer a basic description of the formation and composition of space charge distributions. This might help to explain their effects on the ion residence time, which have experimentally been shown to drastically decrease at higher beam currents.

The simulation model generated during the course of this master's thesis will be an important tool to predict future experimental outcomes, as well as to estimate optimal operation conditions. Possible future modifications of the RFQ negative ion cooler might include optimizations of the setup regarding the ion transmission, detachment efficiency and residence time. The presented model can serve as a powerful instrument to quickly benchmark the influence of such adaptations beforehand, increasing the efficiency of such intentions by a high margin.

Future works may be able to use this model on more powerful hardware or even clusters. This would enable much higher quality of meshing and therefore increase precision of obtained results, while computation times could be kept reasonably low. Generally, a successful implementation of realistic space charge calculation could allow this model to greatly improve the research on the ILIAMS RFQ negative ion cooler as an important component of the VERA facility.

A | Appendix

Table A.1: Factors and function parameters to convert displayed pressure of the ATMION[®] vacuum-gauge and the InstruTech[®] inlet gauge to real pressure for the buffer gases specimen (He), (N₂) and (Ar), data adapted from [46, 47]. The displayed pressure value has to be inserted in mbar.

Gas	Vacuum gauge conversion factor [1]	Inlet gauge conversion function $f(x) = ax + b$			
		a [1]	Δa [1]	b [mbar]	Δb [mbar]
He	5.0	1.1738	0.0078	0.0014	0.0014
N ₂	1.0	1	0	1	0
Ar	0.7	1.6250	0.0071	-0.0015	0.0010

Table A.2: Numerical results for the mass flow rates in kg/s and (mbar l)/s for the different buffer gases He, N₂ and Ar at four different inlet pressures respectively.

Gas	Displayed inlet pressure [mbar]	Real inlet pressure [mbar]	Mass flux 10 ⁻⁸ [kg/s]	Flux [(mbar l)/s]
He	0.150	0.177	1.90	0.118
	0.202	0.239	2.47	0.154
	0.250	0.295	3.55	0.221
	0.309	0.364	4.52	0.281
N ₂	0.150	0.150	6.64	0.059
	0.200	0.200	9.35	0.083
	0.250	0.250	13.26	0.118
	0.300	0.300	16.93	0.151
Ar	0.150	0.242	13.77	0.086
	0.200	0.324	20.88	0.130
	0.250	0.405	31.39	0.196
	0.300	0.486	41.22	0.257

Table A.3: Parameters for the linear regression function linking the real inlet pressure p_{real} for the respective buffer gas to the corresponding mass flux rate f (in [kg/s]) into the cooler.

The function reads: $f = a \cdot p_{real} + b$

The real inlet pressure p_{real} has to be inserted in mbar.

Gas	a	Δa	b	Δb
He	$1.45 \cdot 10^{-7}$	$0.13 \cdot 10^{-7}$	$-7.87 \cdot 10^{-9}$	$3.43 \cdot 10^{-9}$
N ₂	$6.96 \cdot 10^{-7}$	$0.37 \cdot 10^{-7}$	$-4.12 \cdot 10^{-8}$	$0.85 \cdot 10^{-8}$
Ar	$1.14 \cdot 10^{-6}$	$0.07 \cdot 10^{-6}$	$-1.48 \cdot 10^{-7}$	$0.24 \cdot 10^{-7}$

Table A.4: Fit parameters and their uncertainties for the outlet pressure measured at the centered vacuum gauge as a function of the inlet pressure according to the data shown in Figure 3.5. Four different scenarios, corresponding to all combinations of displayed „dis“ and real „re“ inlet and outlet pressures respectively, are given.

The general function reads: $p_{out} = a \cdot p_{in}^2 + b \cdot p_{in} + c$, where p_{out} as well as p_{in} are to be taken in mbar. Fitting parameters for N₂ are only given once. This is because both pressure gauge devices are calibrated to N₂ and therefore the parameters are the same for all scenarios.

$p_{out,dis}$	Gas	$p_{in,dis}$					
		a	Δa	b	Δb	c	Δc
$p_{out,dis}$	He	$1.518 \cdot 10^{-4}$	$0.059 \cdot 10^{-4}$	$9.300 \cdot 10^{-5}$	$0.210 \cdot 10^{-5}$	$5.2 \cdot 10^{-8}$	$9.4 \cdot 10^{-8}$
	N ₂	$1.120 \cdot 10^{-3}$	$0.013 \cdot 10^{-3}$	$2.491 \cdot 10^{-4}$	$0.049 \cdot 10^{-4}$	$-1.2 \cdot 10^{-7}$	$2.4 \cdot 10^{-7}$
	Ar	$3.600 \cdot 10^{-3}$	$0.130 \cdot 10^{-3}$	$5.190 \cdot 10^{-4}$	$0.420 \cdot 10^{-4}$	$-9.0 \cdot 10^{-7}$	$18.0 \cdot 10^{-7}$
$p_{out,dis}$	Gas	$p_{in,re}$					
		a	Δa	b	Δb	c	Δc
$p_{out,dis}$	He	$1.102 \cdot 10^{-4}$	$0.043 \cdot 10^{-4}$	$7.89 \cdot 10^{-5}$	$0.18 \cdot 10^{-5}$	$-5.8 \cdot 10^{-8}$	$9.5 \cdot 10^{-8}$
	N ₂	-	-	-	-	-	-
	Ar	$1.384 \cdot 10^{-3}$	$0.050 \cdot 10^{-3}$	$3.23 \cdot 10^{-4}$	$0.26 \cdot 10^{-4}$	$-8.0 \cdot 10^{-7}$	$18.0 \cdot 10^{-7}$
$p_{out,re}$	Gas	$p_{in,dis}$					
		a	Δa	b	Δb	c	Δc
$p_{out,re}$	He	$7.590 \cdot 10^{-4}$	$0.290 \cdot 10^{-4}$	$4.65 \cdot 10^{-4}$	$1.04 \cdot 10^{-4}$	$2.6 \cdot 10^{-7}$	$4.7 \cdot 10^{-7}$
	N ₂	-	-	-	-	-	-
	Ar	$2.520 \cdot 10^{-3}$	$0.091 \cdot 10^{-3}$	$3.63 \cdot 10^{-4}$	$0.30 \cdot 10^{-4}$	$-7.0 \cdot 10^{-7}$	$13.0 \cdot 10^{-7}$
$p_{out,re}$	Gas	$p_{in,re}$					
		a	Δa	b	Δb	c	Δc
$p_{out,re}$	He	$5.51 \cdot 10^{-4}$	$0.21 \cdot 10^{-4}$	$3.958 \cdot 10^{-4}$	$0.089 \cdot 10^{-4}$	$1.8 \cdot 10^{-7}$	$4.7 \cdot 10^{-7}$
	N ₂	-	-	-	-	-	-
	Ar	$9.94 \cdot 10^{-4}$	$0.38 \cdot 10^{-4}$	$2.190 \cdot 10^{-4}$	$0.190 \cdot 10^{-4}$	$-5.0 \cdot 10^{-7}$	$13.0 \cdot 10^{-7}$

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