Superfluid helium and cryogenic noble gases as stopping media for ion catchers
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The study of atomic nuclei takes a prominent position in the quest to understand quantum-mechanical many-body systems. Studying merely the naturally occurring atomic nuclei imposes a severe limit since these constitute only 5% of the about 7000 combinations of protons and neutrons which are bound by the strong interaction. Experimental facilities where radioactive nuclei are created and studied are thus a necessity. The use and study of radioactive nuclei has had a great impact on other fields of science (ranging from particle physics to materials science) and has yielded an enormous amount of spin-off technologies and applications [3, 81, 117, 119]. For a long time, high-energy beams of only stable isotopes were readily available. In the past 25 years, radioactive ion beam (RIB) facilities allowed for the first time the study of many exotic nuclei with proton or neutron combinations very different than those of stable nuclei. This led to the discovery of many new and unexpected phenomena, such as halo nuclei and the melting of nuclear shells. The construction of next-generation RIB facilities is of the highest priority for the nuclear physics community. Such a next generation facility will produce beams with several orders of magnitude higher intensity, allowing new research with a wide range of nuclear species much further away from the region of stable nuclei. In RIB facilities of the in-flight type such as FAIR (Facility for Antiproton and Ion Research) at GSI, Germany [45], RIBF (Radioactive Ion Beam Factory), RIKEN, Japan [90], NSCL (National Superconducting Cyclotron Laboratory), MSU, United States of America [80] and GANIL (Grand Accélérateur National d’Ions Lourds), France [48], radioactive ions are produced and selected at high energies (100 - 1000 MeV per nucleon), resulting in a radioactive ion beam of high energy and poor beam quality (large emittance and large energy spread). Many precision studies of exotic nuclei far from the valley of stability, such as high-resolution particle spectroscopy or studies in atom or ion traps, need low-energy beams (typically less than a few tens of keV) of high quality (small emittance and energy spread smaller than 1 eV). The same requirements hold for the re-acceleration of the radioactive ions to a precisely defined energy needed e.g. for nuclear reaction studies with radioac-
tive ions. This makes the transformation of a high-energy, low-quality beam into a low-energy, high-quality one (a so-called “cold beam”) an essential part of next generation RIB facilities. The essential requirements for such a transformation are speed and efficiency because the most exotic nuclei have short half-lives (down to milliseconds) and are produced in small quantities. The so-called “ion catcher” method to perform this transformation is being investigated in several laboratories for use in virtually all existing and planned RIB facilities. It is based on the IGISOL method developed in the early 1980’s at the University of Jyväskylä, Finland by J. Ärje, J. Äystö and collaborators (see [9, 34] for reviews on this topic): the high-energy ions are stopped in a chamber filled with helium gas and extracted through an exit-hole. The size of the chambers required by the high energy of the RIB (up to 2 m long with a pressure of up to 2 bar) makes the use of the gas flow to extract the ions from the chamber too slow; guidance by DC electric fields or a combination of DC and RF fields is therefore an essential, but non-trivial task.

The main aim of this research programme is to check the feasibility of cryogenic noble gases and superfluid liquid helium as stopping medium. This thesis will discuss these two approaches separately. Physical processes behind both approaches are discussed in the next two chapters and the experimental techniques, results and discussion are presented in the following chapters.

The fundamental limit of efficiencies of a noble gas ion catcher has been an open question for years. Near and at thermal energies, ions cannot neutralize in collisions with noble gas atoms due to the high ionization potential of the latter. So in case of near zero impurity level, most of the neutralization of ions will happen during the slowing down process. This means that the relative importance of neutralization and ionization cross sections of the ion in the noble gas during slowing down will determine the efficiency limit of a noble gas ion catcher. The relevant physics has been explained since the early days of quantum theory of atomic collisions in the book “The theory of atomic collisions” by N. F. Mott and H. S. W. Massey [76]. However, accurate charge exchange cross sections could not be calculated accurately because of the mathematical complexity involved. The pioneering work by Hughes et al. on electron capture for singly charged particles like protons and muons [58] had been ignored in the nuclear physics community for some three decades and led to a series of misunderstandings. Some recent measurements of the average charge state of low-energy xenon ions in helium are reported by Willmann et al. [130]: the average charge state of xenon ions decreases down to about 0.25 at the lowest measured energy of 10 keV in full agreement with the expectations of the early models in the relevant range of energies.

What happens to thermalized ions is determined by the presence of impurities and the ionization of the noble gas by the energetic ions and possibly by an accelerator beam or radioactive decay radiation or both [6]. Impurities take part in the neutralization process via three-body recombination involving a free electron and form molecules or adducts with the ions, see e.g. Reference [65]. It is important to note that the ionizing radiation also plays a role in re-ionizing those ions which
have been neutralized. So one of the main factors that could improve the efficiency of the gas cells is a low impurity level. Over the past 25 years, a lot of technical development has focused on removing impurities from and preventing ionization of the noble gas. Sub-ppb impurity levels have been achieved in noble gas catchers that are built according to ultra-high vacuum standards, that are bakeable and filled with ultra-pure noble gases, see e.g. References [65, 93]. Constructing large ultra-pure gas catchers, although possible, is far from trivial [93]. There is, however, an alternative approach to reach ultra-pure conditions: freezing out the impurities. An added benefit of cryogenic gas catchers comes from the fact that for a constant gas density, the mass flow out of a gas cell is proportional to the square-root of the gas temperature. This means that a cryogenic noble gas cell allows easier differential pumping for the same gas density, or, reversing this argument, allows a higher density for a constant gas load on the extraction system. The latter means that higher energy ions can be stopped or that, for the same ion energy, the gas cell can be made shorter.

Chapter 4 is dedicated to describe the setup, the methods and the principles used for the experimental study and the data analysis.

Chapter 2 gives an overview of the physics involved in the processes and a literature survey on the available experimental data within the context of cryogenic noble gases. Off-line experiments using recoil ions from a radioactive $^{223}$Ra source are performed to study the feasibility of using a cryogenic noble gas stopping medium for high-energy ion beams. Based on the off-line result, an on-line experimental study on the extraction of thermalized $^{219}$Rn recoils from an ionized stopping medium was conducted. Results from both on-line and off-line experiments and a discussion of the observed properties are reported in chapter 5.

The much larger density (factor 800) of liquid helium relative to room temperature helium gas at 1 bar allows the use of a very small stopping chamber. This makes the extraction of the ions very fast, thereby also increasing the efficiency of ion extraction because of a reduction in neutralization and radioactive decay which is important for short-lived nuclei. Also, the guiding by electric fields can be very simple. The fact that nuclear polarization is preserved in superfluid helium [100, 111, 112, 106, 109, 110] could allow the extraction of polarized beams. At the envisaged temperatures, the low vapour pressure above the superfluid helium surface removes the need for pumping large volumes of helium gas. This new method, if proven successful, could be implemented in many existing and planned laboratories around the world. Experimental work to produce a cold radioactive ion beam using superfluid helium was started at the Department of Physics of the University of Jyväskylä about 7 years ago. $^{219}$Rn ions created in the alpha decay of $^{223}$Ra and recoiling out of the source were stopped in superfluid helium and extracted into the vapour phase as positive ions by means of electric fields. This was the first ever observation of the extraction of positive ions from the surface of liquid helium [56, 57, 108].

Different processes, most importantly the dependence of the survival of snow-
balls on applied electric fields and the dependence of their extraction across the superfluid-vapor interface on temperature are studied in the framework of this project. Chapter 3 give an essence of the physics tools necessary to understand the physical processes involved. The experiments described in chapter 6 aim towards a better understanding of the ion extraction at the superfluid-vapor interface. Further, as a new idea, the possibility to enhance the ion extraction efficiency by second sound assisted superfluid surface evaporation is also investigated.

Finally chapter 7 gives concluding remarks and discusses some future research directions.