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500 kHz or 8.5 GHz? And all the ranges in between.





Production of Zero-Energy Radioactive Nuclear Beams through Extraction from the Liquid-Vapour Interface of Superfluid Helium

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Abstract. A new approach has been investigated to create an ultra-cold radioactive beam from high-energy ions. A ²²³Ra alpha-decay recoil source has been used to produce radioactive ions in superfluid helium. The alpha spectra demonstrate that the recoiling ²¹⁹Rn ions have been extracted out of liquid helium. This first observation of the extraction of heavy positive ions across the superfluid helium surface has been possible thanks to the high sensitivity of radioactive ion detection. An efficiency of 36 % has been obtained for the ion extraction out of liquid helium.

INTRODUCTION

Precision spectroscopic and reaction studies with exotic nuclei involve low energy beams, typically of a few tens of keV and of energy spread of the order of 1 eV. The next generation Radioactive Ion Beam (RIB) facilities considered of high priority around the world aim at such ion beams. Exotic nuclei emerge as a high-energy beam from nuclear reactions with unavoidably large emittance and energy spread.

In this contribution the first results are presented on a new approach for creating an ultra cold ions. Superfluid liquid helium is used here as a stopping medium for energetic beam. After thermalization in superfluid helium, positive ions form owing to electrostriction "snowballs"; clusters of helium atoms around positive ions [1, 2, 3]. The formation and the fast transport of snowballs in liquid helium have been demonstrated earlier at Osaka [4, 5, 6, 7]. We concentrate on the extraction of snowballs/ions from the liquid helium into the vapour phase from where they can be injected into vacuum for further handling and/or post-acceleration. The results in combination with those from Osaka provide a new approach for producing ultracold radioactive ion beams for nuclear physics research. The method developed in this work also has applications in the studies of impurity ions and atoms in superfluid helium. 03 August 2023 14:19:42

EXPERIMENTS

The experimental setup is shown schematically in Fig. 1. A ²²³Ra ($T_{1/2}$ =11.4 d) alpha source was placed at the bottom of an experimental cell with an inner diameter of 62 mm and a height of 105 mm. The decay chain consists of ²¹⁹Rn ($T_{1/2}$ =3.96 s), ²¹⁵Po (1.78 ms), ²¹¹Pb (36.1 min), ²¹¹Bi (2.17 min), ²⁰⁷Tl (4.77 min), and ²⁰⁷Pb (stable). The ²²³Ra alpha-decay products, ²¹⁹Rn, recoiling out of the source with an energy of approximately 100 keV, are stopped within 1 μ m of liquid helium from the source and provide the source of thermalized positive ions. A surface-barrier silicon detector was mounted at the top of the cell to detect the alpha-decay of nuclei. Four ring electrodes were installed in between to provide an electric field to guide the snowballs/ions from the source onto a thin aluminium foil in front of the detector. The experimental cell was placed inside a helium cryostat and the lowest temperature attained was 1.2 K. The temperature of the experimental cell was measured with a 68 Ω Matsushita carbon resistor . The alpha energies were calibrated at a temperature of 1.22 K in the empty cell and with the aluminium foil by using the known alpha lines from the ²²³Ra decay chain. The absolute strength of the ²²³Ra source was also measured; typically 10⁴ Bq.

The ²²³Ra source was covered by 5 mm of liquid helium. This prevented alphas from the source to reach the detector. Without electric field no alphas were observed. Alphas appeared after correct voltages were applied to the electrodes. Based on the ion trajectory calculations with the SIMION code [8], the applied voltages were optimized by maximizing the count rate of ²¹⁹Rn alpha decay on the aluminium foil. The charge of the snowballs/ions was confirmed to be positive from the fact that no alphas were observed when lower voltages were applied on the source and bottom electrode than on the electrode 4, thus creating an electrostatic barrier for positive charges. Alpha spectra measured at different temperatures are shown in Fig. 2. The identification of the peaks is based on the measured energy and the calculated energy loss between the place of decay and the detector by using the SRIM code [9]. The peaks marked as Rn, Po and Bi are due to ²¹⁹Rn, ²¹⁵Po and ²¹¹Bi alpha decays on the aluminuim foil in front of the detector; the



FIGURE 1. Experimental setup. The inner diameter of the experimental cell is 62 mm and the height is 105 mm. The 223Ra source is covered by superfluid helium. The typical voltages on the electrodes are shown, except those on the source and bottom electrode which were varied during the measurement.



FIGURE 2. Alpha spectra with voltages on the electrodes at 1.22, 1.50 and 1.60 K. The peaks marked as "Rn", "Po" and "Bi" are from ²¹⁹Rn, ²¹⁵Po and ²¹¹Bi nuclei on the aluminium foil in front of the detector. The peaks "Rn"" and "Po"" are from ²¹⁹Rn and ²¹⁵Po nuclei at the surface of liquid helium.

peaks Rn^{*} and Po^{*} are due to ²¹⁹Rn and ²¹⁵Po decaying at the surface of liquid helium. Other peaks from the surface are masked by the intense peaks from the decays on the aluminuim foil or the background. The vapour pressure above the liquid helium rises drastically with increasing temperature. The observed alpha spectra demonstrate that the ²¹⁹Rn ions have been extracted out of the liquid helium and collected on the aluminium foil in front of the detector by the static electric field. The extraction of positive ions from the superfluid helium surface has been observed for the first time, thanks to the high sensitivity of radioactivity detection.

RESULTS AND DISCUSSION

The overall transport efficiency is the ratio of the numbers of ions observed on the aluminium foil to those produced in the source and determined from the intensity of the ²¹⁹Rn peak. The results of a series of measurements on the overall efficiency against temperature and electric field are shown in Fig. 3. In these measurements, only the voltages on the source and bottom electrode were changed, thus basically changing the electric field in the lower part of the cell. The overall efficiency for ²¹⁹Rn is due to four factors: snowball formation, transport in the liquid, ion extraction and transport in the vapour.

The results of the measurements done with +450 V source voltage at 1.22, 1.34 and 1.60 K are given in Table 1. The best statistics was obtained at 1.60 K. At this temperature the overall efficiency of $(7.2\pm0.5)\times10^{-4}$ was obtained, which includes the statistical and systematic errors. In order to obtain the extraction efficiency from the liquid surface we used the SIMION code to deduce the transport efficiency through the vapour phase and was deduced to be $38\pm5\%$. From the alpha peak of ²¹⁹Rn decaying

TABLE 1. Efficiencies (in %) at 1.22, 1.34 and 1.60 K measured with +450 V on the source and bottom electrodes. This corresponds to an electric field strength at the surface of 200 V/cm.

T (K)	1.22	1.34	1.60
Overall	0.029	0.032	0.072
Snowball formation	8.6	5.2	0.8
Snowball transport in liquid	100	100	100
Ion extraction out of liquid	0.9	1.6	23
Ion transport in vapour	38	38	38

on the surface of the liquid helium, the efficiency for a snowball to form and reach the surface, but not to be extracted, was deduced to be $(6.4\pm0.9)\times10^{-3}$. The efficiency for ion extraction out of liquid helium is then $23\pm4\%$. In all our experimental situations, the transport time of snowballs in liquid helium is much smaller than the snowball neutralization time as mentioned below, we therefore conclude that the efficiency of snowball transportation inside the liquid is virtually 100%. This gives an efficiency for snowball formation of about 0.8% at 1.60 K.

At 1.22 and 1.34 K, our analyses show that the extraction efficiency of ²¹⁵Po out of the surface is significantly small. Because of its very short half-life, ²¹⁵Po decays essentially in the same place as its ²¹⁹Rn mother nucleus and almost no ²¹⁵Po from the source reaches the surface before its decay. Thus the Po* peak gives the same information as the Rn* peak. This equivalence is used for the measurements at the lower temperatures, where the Rn* peak is masked by the peaks from ²¹¹Bi decay. Fig. 4 shows the deduced extraction efficiencies out of the liquid surface and snowball formation probabilities at 1.22 and 1.34 K against electric field strength on the liquid surface. The data show that the snowball formation probability and extraction efficiency behave in opposite ways with respect to temperature and electric field. Free electrons created by alpha-decay in the vicinity of the source can neutralize positive ions, thus preventing the creation of a snowball. Positive ions and free electrons move faster at lower temperatures (because of an increase in mobility [3, 1]) and stronger electric fields. For lower temperatures and



FIGURE 3. Overall efficiencies at different temperatures as a function of the source and bottom electrode voltage. The voltages applied on the other electrodes were fixed at the values shown in Fig. 1.

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larger electric fields, positive ions and free electrons are separated faster, resulting in an increase in snowball formation efficiency. The fact that a higher temperature eases the release of positive ions from the surface was earlier also observed for negative charges [10, 11]. Understanding the decrease of the extraction efficiency with electric field requires further study. The interplay of the two opposite dependencies described above explains qualitatively the curves shown in Fig. 3: the overall efficiency first rises and then drops with increasing electric field. This is clearly seen at lower temperatures. For higher temperatures, the maximum efficiency is larger and can be reached at higher electric fields.

The time distribution for a snowball/ion to travel from the source to the aluminium foil was studied at a temperature of 1.50 K with an electric field strength of 85 V/cm. The ²²³Ra recoil source can be "switched off" by putting it at a lower voltage than the bottom electrode. Moreover, the ion transport can be blocked by raising the voltage on one of the ring electrodes in order to create an electrostatic barrier. Measurements were done in which the source and transport were pulsed. A fit to the data gives a minimum delay time of less than a few milliseconds and a release time of 90 ± 10 ms. The observed release is due to at least two processes: snowball neutralization at the surface and transport across the surface. This observation shows that ions/snowballs are, indeed, trapped on the surface prior to their extraction. This confirms earlier observations that ions can be trapped at the surface by a lateral holding potential [12].

Positive and negative ions in liquid helium near a free surface experience an attractive potential which tends to inhibit evaporation of ions across the surface. Assuming a Brownian distribution of ions, the extraction efficiency through a free surface is then proportional to $\exp(-E_b/kT)$, where k is Boltzmann constant, E_b the magnitude of the barrier. This barrier has been measured for negative charges (electrons) [10, 11], but, due to a lack of sensitivity, never for positive ions. Our experimental extraction efficiencies allow us to determine this barrier to be $E_b/k=19.4\pm4.5$ K. In this case, the electric field strength at the surface was 200 V/cm.

An attempt was made to optimize two factors: a large electric field to enhance snowball formation and a high temperature to enhance extraction from the surface. For this,



FIGURE 4. Extraction efficiency out of the liquid surface (left panel) and snowball formation efficiency at 1.22 K (squares) and 1.34 K (triangles) plotted against electric field strength on the liquid surface. The experimental conditions are identical with those in Fig. 3. The lines are to guide the eye.

we applied a pulsed second sound wave created from a circular heater around the radioactive source. Second sound is the heat transmitted in a wave-like manner rather than by diffusion and was observed in superfluid liquid helium [13]. It is expected to enhance the extraction through the free liquid surface by surpassing the barrier. The current pulses that excited the heater were 2 to 50 ms wide and were repeated every 50 to 500 ms. The increase of the overall efficiency was 10 to 30% depending on heater power.

CONCLUSION

A new method using liquid helium as stopping medium to transform a high-energy beam into ultra cold ions has been proposed and tested with an alpha-recoil source. Extraction of positive ions across the superfluid helium surface has been observed for the first time. An efficiency of $23\pm4\%$ was obtained at 1.60 K. This promises the method to be used at the next generation RIB facilities. Since a probability for snowball formation in an experiment at Osaka [5] has been found to be $20\pm10\%$ at 1.43 K, we expect a better overall performance for high-energy ions. We propose this highly sensitive method to be applied as a new tool in the study of superfluid helium properties, where the traditional electric current or charge measurements have limited sensitivity.

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