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RADIOACTIVE CHARGE CARRIERS IN LIQUID HELIUM CREATED FROM FAST NUCLEAR BEAMS

NORIAKI TAKAHASHI[1]

Osaka Gakuin University, Kishibe-Minami 2-36-1, Suita/Osaka, 564-8511 Japan

Paper devoted to honour the memory of Professor Nikola Cindro

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An ionic positive charge in superfluid helium exists in the form of an aggregate of helium atoms around a core ion and is called a "snowball". Positively charged impurities were created through injection of fast nuclear beams into liquid helium. The transport of positive charges in liquid helium was traced through the measurements of electric currents and of radioactive decays of the impurity ions. Applied electric fields of both directions controlled the movements of impurity ions unambiguously. Production rates of snowballs and of neutrals from the injection of nuclear beams have been estimated. The rates change differently for various ions used, not only with temperature of the liquid helium but also with spatial density of the incident fast beams.

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1. Introduction

Impurity ions and atoms exist in liquid helium as moveable objects and as inhomogeneous localisations resembling colour centres in solids. Especially the superfluid helium with imputiries represents an excellent system to study the transport phenomena in a quantum liquid. Already in the fifties, it was known that the positive ions created from α radioactive sources possess special transport properties, depending on the temperature of the quantum fluid. Reif and Meyer [1] investigated the electric mobility of ^{4}He ions from the ^{210}Po source immersed in superfluid he-

[1] Professor Emeritus, Osaka University

lium. The measured small speed of the positive charge carriers was interpreted as a results of a large number of collisions with rotons in the temperature range of 0.7 K to 2.17 K. The magnitude of the speed suggested the existence of macroscopic aggregates for positive charges. Atkins explained the formation of such an aggregate of helium atoms around a core ion through the electrostriction [2], since the electric polarisability of ^{4}He atom is as small as 0.2×10^{-24} cm^3, but not zero. The motion reflected the viscosity of normal fluid helium above 2.17 K. The studies leading to the finding of the vortices and vortex rings were the most impressive ones of the early stage of the studies on impurity ions in superfluid helium [3]. Detailed mobility studies in recent years allowed more specific insights into the structure of impurity ions in superfluid helium, snowballs or bubbles [4].

The detection of such positive charge carriers was usually accomplished through measurements of the electric charges or the currents of the carriers. Although developed during many decades, these methods were not able to compete in efficiencies with the detection of the individual charge carriers. As one of the attempts to detect individual impurity ions, the method of radioactive snowballs was contrived and tested. β rays from ^{12}B were used to count the impurity ions in superfluid helium. Nuclear spin polarisation of ^{12}B ions in superfluid helium was successfully determined by using this method. The idea and the first result have been reported at the Gaussig Conference [5]. More elaborated experimental results on the freezing-out of nuclear polarisation of the impurity ions in superfluid helium have been presented with the recent improvements of the measuring methods [6, 7].

The loci of the radioactive positive ions have been clearly exhibited in two-dimensional space-time plots and are used to trace their motions. Electric fields of $100-500$ V/cm were effective in driving the charged impurities in superfluid helium [8]. The position and time spectra obtained for the radioactive impurity ions in superfluid helium indicated the different behaviour of the ions and the neutrals created following the injection of high-speed beams into liquid helium.

The versatility of the radioactivity-detection methods has been repeatedly demonstrated in other experiments, too, where radioactive ^{219}Ra ions were extracted from the superfluid helium surface. The efficiency for the extraction of positive ions was obtained and the potential barrier was determined for the first time. Extraction of electrons was hitherto observed but the individual identification of positive ions seemed to be difficult for a long time. The α radioactivity of the ^{219}Ra ions was most powerfully and effectively utilized [9]. One of the ambitious projects to apply this method is the production of ultra-cold zero-energy radioactive nuclear beams. Energetic radioactive nuclear beams produced in nuclear reactions are injected into superfluid helium and are decelerated and stopped. These ions are transported through the quantum liquid in a form of radioactive snowballs and are extracted from the liquid–vapour interface. The zero-energy nuclear beams thus produced will be used, possibly after re-acceleration, for nuclear reaction studies and supply indispensable data for nuclear astrophysics or nuclear structure physics [10].

Basic data are still scarce in planning experiments in which radioactive snowballs are used as essential entities in superfluid helium, e. g., the studies on the freezing-out of nuclear polarisation for spectroscopic purposes or on the production of ultracold radioactive nuclear beams for various nuclear reaction studies. In this paper, the detection of nuclear radiations from the impurity core ions is described and compared with the traditional electric measurements. The present experiment has been carried out to fill the gaps where no relevant data exist for the optimisation of production rates of snowballs.

2. *Experiment*

2.1. *Nuclear radiation measurements*

Beams of 150 MeV ^{7}Li ions were obtained from the 230 cm cyclotron at the Research Center for Nuclear Physics (RCNP), Osaka University. The fast beams were shaped into pulses of 1.2 s duration with a repetition rate of 0.3 Hz and were focussed onto a $(CD_2)_n$ target. The reaction products, ^{8}Li ($T_{1/2}$ = 838 ms), were injected at 0° into superfluid helium at temperatures of 2.1 K and 1.7 K. The experimental apparatus is shown in Fig. 1. The cryostat was contained in a vacuum chamber (G). The superfluid helium chamber consists of a fibre-reinforced plastics cylinder (H) and of a metal bottom plate (D). The entrance window (A) was made of aluminium with thickness of 50 μm. The temperature of the liquid helium was

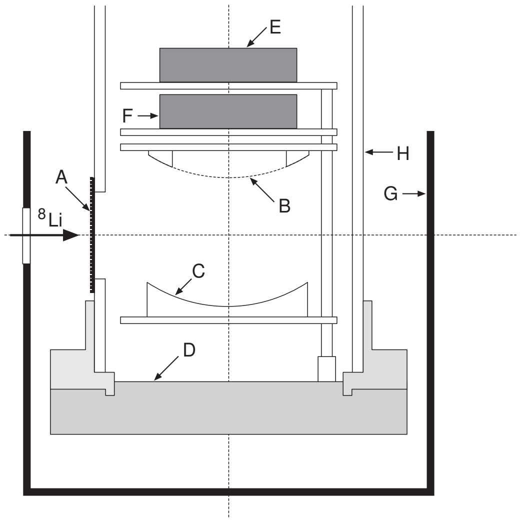

Fig. 1. Schematic configuration of the cryostat, indicating the entrance window A, the grid B and the bottom electrode C. The surface-barrier detectors E and F are for the measurements using the radiation-detection method. A metal bottom plate D and a fibre-reinforced plastic cylinder H constitute the superfluid helium chamber and G is the vacuum chamber. The conventional current and charge measurements were carried out by deploying a collector electrode and a concentric grid at the centre of the superfluid helium chamber.

monitored by the helium vapour pressure in the chamber, with a germanium and a carbon resistors placed inside for temperature measurement.

The ^{8}Li ions with kinetic energy exceeding a specified value, determined from the range and the energy loss in the entrance window and in liquid helium, were allowed to penetrate through liquid helium up to the middle of the liquid helium chamber, generating large amounts of helium ions and electrons by ionisation. Most of the injected ^{8}Li ions were neutralised during the stopping process. But a few of the ^{8}Li ions, through the electrostriction on the He atoms surrounding the ions after deceleration, form snowballs around themselves, thus escaping neutralisation. The snowballs were displaced by approximately 25 mm towards the Si surface barrier detector (F) in the superfluid helium chamber through an electric field applied perpendicularly to the original beam direction between the grid (B) and the bottom electrode (C). This vertical displacement of ^{8}Li was necessary to avoid the neutron background which was produced mainly along the incident beam axis and deteriorated the α counting. To suppress the detection of background β rays, the coincidence of β and α counts in two surface barrier detectors (E and F) was required. The lower detector (F) was a thin transmission counter to stop the low energy α particles from the instantaneous decay of ^{8}Be. The time interval between the incoming ^{8}Li pulses and the application of the vertical electric field served as delay time in determining the transport time and also the trapping time. The intensity of ^{8}Li snowballs was quite low, a couple of tens of counts per minute. But it was possible to get good counting rates for α particles coming from the ^{8}Li decay into $^8\mathrm{Be} \rightarrow \alpha + \alpha$.

2.2. Measurements of electric current and charge

A low-intensity beam of 135 MeV ^{14}N ions was also obtained from the 230 cm cyclotron. The beam was shaped into pulses of 100 ms duration with a repetition time of 200-1 000 ms and was focussed within a diameter of less than 4 mm. The energetic ions were injected into liquid helium at temperatures of 1.7 K to 4.2 K through the 50 mm diameter entrance window made from aluminium. A cylindrical grid of 30 mm diameter and a collection electrode of 2 mm diameter were placed concentrically in the centre of the chamber. The snowballs produced from the ^{14}N beam were displaced by approximately 25 mm through a horizontal electric field between the window, the grid and the electrode. The electric currents to the collection electrode and the grid, typically of the order of picoamperes, were measured with an electrometer and integrated with a current integrator.

The electric potential of the entrance window was – 1.0 kV and that of the grid was – 0.65 kV. The grid potential was applied during the off-beam period following the end of the on-beam period with a certain delay time T_d. It was important to apply an electric potential when the impurity ions are brought to stop in liquid helium. Otherwise, the production rate of snowballs becomes quite low, presumably due to the recombination of ions with ubiquitous electrons liberated during the stopping process of the fast ions. For different delay times, the current on the collector electrode was observed and integrated. This configuration enabled us to keep the snowballs produced in the region between the entrance window and the grid during the delay time. The applied grid potential drove these snowballs to the collector where they could be measured as collector current.

It is often necessary to know certain characteristics of the probability for production of charge carriers in liquid helium. To determine the dependence on beam intensities, the following measurements were carried out. The fast nuclear beams were injected into liquid helium with various intensities but essentially with the same focussing. The ratio of the current on the beam stopper and the collector electrode was determined. The measurements were carried out at various temperature from 1.7 K to 4.2 K below and above the lambda transition point. The injection of the energetic ions did not disturb the superfluidity regime, unless the beam intensities were high, at least in the subnanoamperes range as dealt here. The heat conductivity of the superfluid helium was sufficiently large.

3. *Results and discussions*

A measured delay time spectrum for the snowballs of ^{14}N is shown in Fig. 2. A solid line is drawn for $T_{1/2} = 700$ ms as fitted to the measured points. The temperature range is between 1.85 and 1.88 K, with liquid helium is in the superfluid state. The current on the beam stopper, i_{fc}, is simply the intensity of the incident ^{14}N beam. On the other hand, the current measured at the collector electrode, i_{ce}, consisted of the summed contribution from the ^{14}N ions and the generated He ions. The snowballs observed here have non-radioactive core ions of either ^{14}N or ^{4}He.

It is necessary to assure that the presence of He snowballs did not mask the detection of ^{14}N snowballs. The production ratio of the helium snowballs in liquid helium at this temperature range is known to be unusually low, of the order of 10^{-6}. This low production rate may be due to the high ionisation potential of He ions, and this could be a key to answer the question, whether the core ions

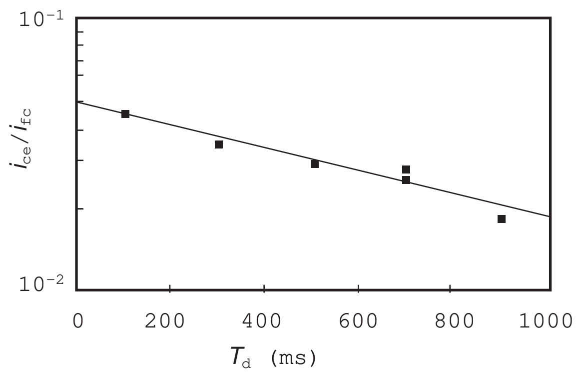

Fig. 2. Decrease of the current on the collector electrode i_{ce} normalized to the incident beam current i_{fc} as a function of delay time for ^{14}N.

in He snowballs are actually $^4He^+$ or $^4He_2^+$. The free $^4He_2^+$ ion is unstable. For many other ions, the production ratio of snowballs is generally of the order of 10% and depends slightly on the temperature of liquid helium. Moreover, the density of the incoming fast ions may influence the production rate of the charge carriers. It amounts to about 10% for ^{14}N. From the large difference in the formation ratio, it is concluded that we are observing the trend in the behaviour of ^{14}N snowballs at the inner electrode through a simple current measurement.

A spectrum with the time delayed α particle counting from ^8Li was obtained before [6]. The typical value for the corresponding half-life for the survival of snowballs was 350 ms, corrected for the radioactive halflife of ^8Li nuclei (848 ms). The difference by the factor nearly of two for ^8Li and ^{14}N is not clear at the moment. The electron affinity of the core ions should not be affected too much after formation of snowballs.

In order to have a definitive picture of the transport of snowballs in liquid helium, we tried to find out the components that might have faster speeds than the normally-charged snowballs. But there were no such components within the accuracy of our measurements. Thus the existence of doubly or multiply charged snowballs is unlikely and the snowballs were singly charged.

The present result guarantees that the present electric current method for ^{14}N, as well as the radiation detection method employed here for ^8Li, is effective in determining the lifetime for survival of snowballs in superfluid helium. Snowballs live in superfluid helium quite a long time, unless there are no disturbances [11]. One of such disturbances might be the heat flow into the cryostat. We found, there was a heat flow into the cryostat which created the motion in the liquid helium. Due to such disturbances present in our cryostat, the loss in stored snowballs in the

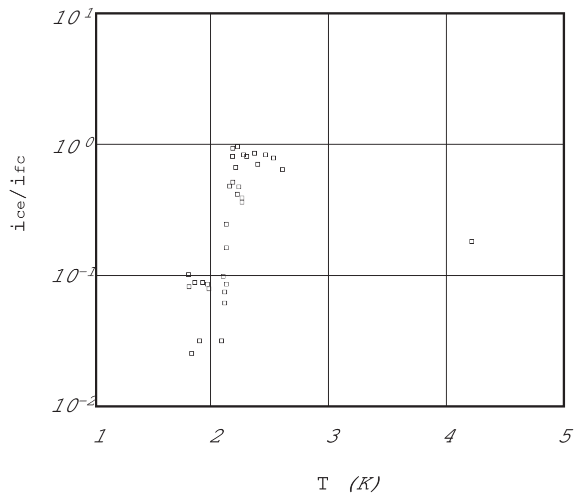

Fig. 3. Ratio of the collector current to the incident beam current, i_{ce}/i_{fc}, versus liquid helium temperature.

region between the window and the grid and in transporting them to the collector electrode had to be considered. These data may lead to the necessary corrections when the absolute efficiency of the production ratios are to be determined.

The ratio of the integrated current at the collector electrode to that at the Faraday cup is plotted against temperature in Fig. 3. The measurements span from the lowest temperature obtained at 1.7 K up to the boiling point at 4.2 K across the lambda point.

It is interesting to notice that an abrupt change in the production ratio takes place when crossing the lambda point at 2.17 K. Above this temperature, the behaviour of the ratio reflects the change of viscosity against temperature. As the temperature is raised, the viscosity increases and the transportation of snowballs to the collector electrode becomes more and more inefficient. This tendency is seen in the change of the ratio from 1.0 at 2.18 K to 0.2 at 4.2 K. Below the lambda point, the ratio is approximately 0.09 and this value does not change much, though the measured temperature range is small.

The efficiency of collecting ^{14}N snowballs in superfluid helium is shown in Fig. 4. The ratio of the two currents at the collector electrode i_{ce} to that at the Faraday cup i_{fc} is strongly dependent on the intensity of the incident beam. The measurements were started with an extremely weak ^{14}N beam, where the efficiency amounted to almost 30%. The recombination takes place on the ^{14}N ions when they come almost to rest, encountering the ubiquitous electrons liberated from the helium atoms of the liquid. It is understandable that the probability of production of ^{14}N snowballs decreases as the beam intensity of the incident beam increases. The beam focussing was kept always the same when increasing intensity.

We experienced that the application of an electric field at the production point,

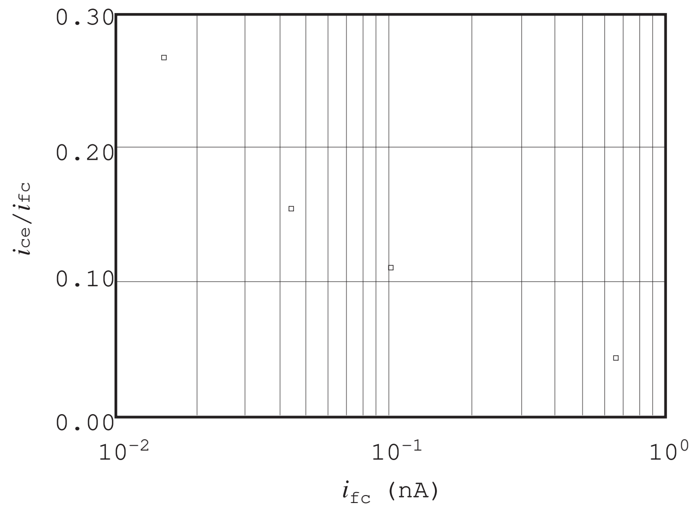

Fig. 4. ^{14}N *snowball current as a function of incident beam current at 1.87 – 1.89* K *superfluid helium temperature.*

i. e., at the end of the range of the incoming ions, is important in order to have a reasonably large snowball production rate. The value of the necessary electric field may vary from ion to ion and it seems that the minimum lies typically around 100 V/cm. The experience came from the experiments on freezing-out of ^{12}B polarisation in snowballs and also from the extraction experiment with ^{219}Ra snowballs. In this experiment, we supplied a sufficiently strong electric field. Lower efficiency for production of snowballs in superfluid helium can generally be attributed to the higher mobility of electric charges leading to recombination.

A different configuration might be used to measure the production probability of snowballs. In Fig. 5, a typical two-dimensional space-time plot for ^{8}Li is given. The experimental data were obtained through the β counting with the apparatus described in Ref. [8] and were newly reanalysed.

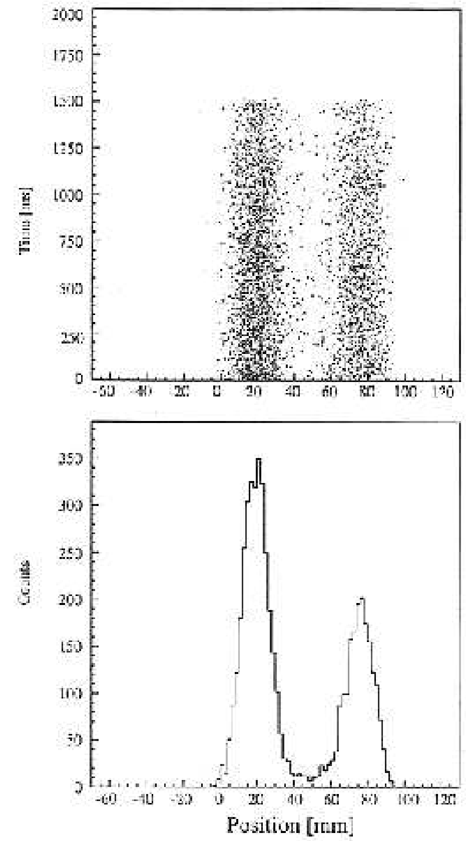

Fig. 5. A two-dimensional space-time plot in the upper part shows the separation of neutrals and charged (snowballs) for ^{8}Li. *The left branch corresponds to neutrals* ^{8}Li *and the right branch corresponds to snowballs* ^{8}Li, *driven with the electric field after injection of* ^{8}Li *ions into superfluid helium at 1.4* K. *The lower spectrum was obtained through projection on the position axis.*

In this case, the separation was carried out with a low electric field at the early stage directly following the introduction of the ^{8}Li ions into the superfluid helium and the electric field was switched off. The original two-dimensional plot over the position and the time axes was converted into a position spectrum by integrating the counts with respect to the time axis. The scale of the time axis was selected so that the transport time for snowballs is negligibly small, compared to the radioactive half-life for ^{8}Li. Now the separation of two peaks for the neutrals and the charged is clean in the position spectrum and the ratio for the production of snowballs and neutrals is directly obtainable by integrating the counts in the individual peaks. This kind of experiments is more persuading and we recognize the advantage of the radiation detection method over the traditional electric charge measurements. The measured snowball production rates are 30% for ^{12}B, 10% for ^{12}N and 35% for ^{8}Li; the errors of these values are estimated at $\pm 3\,\%$. The values of the production ratio are different for different species of impurity ions. Most significant of this is the case of He ions.

4. Conclusions

We have obtained typical values for the lifetime of snowballs in superfluid helium by the use of nuclear-radiation detection and electric-current measurement. At the same time, we have obtained the probabilities for the production of snowballs as a function of temperature and beam intensity. The lifetime of snowballs in liquid helium is long enough for measurements with many radioactive nuclei of our concern. High probabilities for survival of snowballs are important when we plan experiments involving trapping and transport of radioactive positive ions in superfluid helium.

Acknowledgements

This paper is dedicated to the memory of dear friend, Professor Nikola Cindro, who was a precious physicist and at the same time a well cultivated personality with quite good sense of even humour.

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References

[1] L. Meyer and F. Reif, Phys. Rev. **110** (1958) 279L;
G. Careri, F. Scaramuzzi and J. O. Thomson, Nuovo Cimento **8** (1959) 1758.

[2] K. R. Atkins, Phys. Rev. **116** (1959) 1339.

[3] G. W. Rayfields and F. Reif, Phys. Rev. Lett. **11** (1963) 305;
G. W. Rayfields and F. Reif, Phys. Rev. **136 A** (1964) 1194.

[4] B. Tabbert, H. Günther and G. zu Putlitz, J. Low Temp. Phys. **109** (1997) 653.

[5] N. Takahashi, in *Dynamics of Heavy-Ion Collisions*, ZfK-610, edited by R. Reif and R. Schmidt (1986) p. 176.

[6] N. Takahashi, T. Shimoda, Y. Fujita, T. Itahashi and H. Miyatake, Z. Phys. B **98** (1995) 347.

[7] N. Takahashi, T. Shimoda, H. Miyatake, S. Mitsuoka, Y. Mizoi, H. Kobayashi, M. Sasaki, T. Shirakura, H. Ueno, K. Asahi and S. Morinobu, Hyperfine Interactions **97/98** (1996) 469;
T. Shimoda, H. Miyatake, S. Mitsuoka, Y. Mozoi, H. Kobayashi, M. Sasaki, T. Shirakura, H. Ueno, H. Izumi, K. Asahi, T. Murakami, S. Morinobu and N. Takahashi, Nucl. Phys. A **588** (1995) 235.

[8] N. Takahashi, T. Shigematsu, S. Shimizu, K. Horie, Y. Hirayama, H. Izumi and T. Shimoda, Physica B **284-288** (2000) 89.

[9] N. Takahashi, W.X. Huang, K. Gloos, P. Dendooven, J. P. Pekola and J. Äystö, to be published in Physica B (2003).

[10] W. X. Huang, P. Dendooven, K. Gloos, N. Takahashi, J. P. Pekola and J. Äystö, to be published in Nucl. Instr. Meth. B (2003).

[11] G. A. Williams and R. E. Packard, J. Low Temp. Phys. **33** (1978) 459.

TVORBA RADIOAKTIVNIH NOSITELJA NABOJA IZ BRZIH NUKLEARNIH SNOPOVA U TEKUĆEM HELIJU

Oko iona sredice, u suprafluidnom heliju stvara se nakupina helijevih atoma koja se naziva "(snježna) gruda". Pozitivno nabijene grude tvorili smo ubacivanjem brzih nuklearnih snopova u tekući helij. Gibanje pozitivnih čestica pratili smo mjerenjem električnih struja i detekcijom radioaktivnog raspada ubačenih iona. Primijenili smo električno polje u oba smjera radi jednoznačnog upravljanja gibanja iona. Ocijenili smo vjerojatnosti tvorbe gruda i neutralnih nakupina nastalih ubacivanjem nuklearnih snopova. Za različite ione koje smo rabili, te vjerojatnosti različito ovise ne samo o temperaturi, nego i o prostornoj gustoći upadnih snopova.