Superfluid helium and cryogenic noble gases as stopping media for ion catchers
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Helium has two stable isotopes $^4\text{He}$ and $^3\text{He}$. Though $^3\text{He}$ has the same chemical nature as $^4\text{He}$, the lighter mass of $^3\text{He}$ and the fact that $^3\text{He}$ is a fermion result in markedly different behavior. Helium was first liquefied by Kamerlingh Onnes in 1908 for which he won a Nobel Prize in 1913. This work deals with the properties of $^4\text{He}$, and unless specifically stated, it should be assumed that $^4\text{He}$ is the isotope being discussed and that parameters are given at saturated vapor pressure.

When liquid helium is cooled to 2.172 K, it undergoes a phase transition. Because of the characteristic profile of the heat capacity curve (see Figure 3.1), the temperature at which the transition takes place is called the lambda temperature $T_\lambda$. There is no specific volume change or latent heat involved in the lambda transition. In 1938, P. Kapitza [63] and independently J. F. Allen and A. D. Misener [1] reported the zero viscosity of liquid helium below $T_\lambda$. Unlike most fluids, liquid helium below $T_\lambda$ behaves as a non-Newtonian fluid. It was Kapitza who introduced the term “superfluid” helium to describe the unusual behavior of helium below $T_\lambda$. The quantum-mechanical nature of the superfluid helium introduces properties which cannot be explained with a classical treatment.

To account for this unique behavior, F. London [68] suggested that as a single $^4\text{He}$ atom is a boson, a large collection of $^4\text{He}$ atoms can form a Bose-condensate-like state. Although London was right, experimentally it was very difficult to prove. In the meantime another theory known as the two-fluid model was introduced by L. Tisza [115] and L. Landau [66]. The 2-fluid model postulates that the superfluid helium is composed of two inseparable inter-penetrating fluids, one component being referred to as the normal fluid and the other component as the superfluid. Due to the simplicity and excellent agreement with experimental results, the two-
Figure 3.1: Specific heat capacity $c_p$ of liquid helium at saturated vapor pressure as a function of temperature $T$ [14].

Figure 3.2: Relative density of the normal and superfluid components in superfluid helium as a function of temperature $T$ [14].
**Figure 3.3:** Entropy $S$ of superfluid helium as a function of temperature $T$ [14].

**Figure 3.4:** Second sound velocity $C_{ss}$ as a function of temperature $T$ [14].
According to the two-fluid model the density of the superfluid component is given by

$$\rho_s = \rho \left(1 - \frac{T}{T_\lambda}\right)^{5.6},$$

(3.1)

$$\rho = \rho_s + \rho_n,$$

(3.2)

where $\rho$ is the effective density of the superfluid helium, $\rho_s$ is the density of the superfluid component and $\rho_n$ is the density of the normal component. The effective superfluid density $\rho = \rho_s$ at $T=0$ K and $\rho = \rho_n$ at $T=T_\lambda$ (see Figure 3.2). The superfluid component is thought to have no entropy and no viscosity. One of the most interesting characteristics of superfluid helium is the ability to transmit more than one type of sound. In addition to ordinary or first sound which is a density variation brought on by a local pressure gradient, there is a second sound which is the propagation of a thermal wave as a result of fluctuations in the local entropy. The first sound results in a compression shock wave, whereas the second sound results in a thermal shock wave. The characteristic feature of these sound waves can be explained on the basis of the two-fluid model. The velocity of the second sound is given by the Tisza-Landau thermodynamic expression [31]

$$C_{ss}^2 = \left[\left(\frac{\rho_n}{\rho}\right)^{-1} - 1\right] \frac{T S^2}{C_p},$$

(3.3)

where $T$ is the temperature, $S$ the entropy (see Figure 3.3) and $C_p$ the specific heat capacity (see Figure 3.1) of the superfluid helium. Figure 3.4 shows the second sound velocity $C_{ss}$ as a function of temperature. A detailed discussion of the two-fluid model is outside the scope of this work, however there are excellent references on the subject [67, 69, 97].

Helium atoms are nonpolar, stable and spherically symmetric. They have a very low electric polarizability ($2.04 \times 10^{-25}$ cm$^3$). Charged particles introduced into superfluid helium will interact with the helium atoms to form various complexes. This chapter gives an overview of the properties of electrons and positive ions in superfluid helium which are relevant to this work.

### 3.1 Positive ions in superfluid helium

Charged particles in superfluid helium will introduce a polarization interaction with the ambient helium. An ion produces a strong field in its vicinity and the potential energy experienced by the polarizable medium results in an electrostrictive increase in the local density. A solid layer of helium atoms forms around positive ions because the pressure produced by the electrostrictive interaction of positive ion and the ambient medium is larger than the melting pressure for helium.
3.1 Positive ions in superfluid helium

This quasimacroscopic entity of a positive ion and the helium solidification region around is called a “snowball” (see Figure 3.5). Atkins [7] put forward a simple model to explain this phenomenon. In this model, the ion complex is taken to be a solid helium sphere surrounding the positive ion core. This structure arises because of the induced dipole attraction between the core ion and a helium atom. In Atkins’ model the snowball radius $R_{sb}$ is given by

$$R_{sb} \simeq \frac{V_s}{(V_l - V_s)} \left( \frac{2\sigma_{ls}}{(P_m - P_a)} \right),$$

where $P_a$ is the ambient pressure of the liquid helium far from the ion, $P_m$ is the melting pressure of solid helium ($\approx 25$ bar), $V_s (=27.6 \text{ cm}^3 \text{ mole}^{-1})$ and $V_l (=21 \text{ cm}^3 \text{ mole}^{-1})$ are the molar volume of liquid and solid helium, and $\sigma_{ls} (=1.35 \times 10^{-4} \text{ N m}^{-1})$ is the solid-liquid surface tension for helium. In Atkins’ model the snowball radius $R_{sb}$ is a function of the ion charge $q$ and is roughly 0.6 - 0.7 nm for singly charged ions (Figure 3.7). The effective hydrodynamic mass of a spherical shape of radius $R_{sb}$ in a fluid of mass density $\rho$ is given by

$$M_{\text{hydro}} = \frac{2}{3} \pi R_{sb}^3 \rho .$$

This results in a snowball mass of ion mass plus 40 to 60 helium masses. Deviations from Atkins’ model have been observed for different ionic species.

A more rigorous model which takes into account the direct interaction with the central ion’s valance electrons, an interaction associated with the absence or excess of helium near the core ion and a van der Waals interaction can be found in later literature [25, 50]. Nevertheless, Atkins’ model retains its significance as a
first approximation and can be used to estimate the scale of various effects. It is important to note that heavy alkali-earth ions form bubbles in contrast to the other positive ions (see Section 3.3).

### 3.2 Electrons in superfluid helium

The electron in superfluid helium forms a structure which cannot be described by Atkins’ model. The helium atom is a stable quantum system, which cannot accommodate a surplus electron at distances of the order of Bohr’s orbit ($a_0 = 0.529 \times 10^{-10}$ m) and the interaction between the electron and neighboring helium atoms is of a very strong repulsive nature at short range (Pauli repulsion). A long-range attractive interaction arising from the electrostrictive polarization of the atoms is also present. When a free electron is injected into superfluid helium and forced to move in the interatomic spaces, it experiences a repulsive potential of about 1 eV and as a result is energetically favorably localized within a spherical cavity from which the helium atoms are completely excluded, a so-called bubble (Figure 3.6). In the representation of the bubble-helium interaction, we can treat helium as a continuous medium rather than as discrete atoms. The balance between the electron zero point energy, the surface energy of the liquid and the pressure-volume energy determines the bubble size. As a first approximation the bubble
energy can be expressed as \[ E = \frac{h^2}{8mR_b^2} + 4\pi R_b^2 \sigma_s + \frac{4}{3} \pi R_b^3 P_a, \] (3.6)

where \( R_b \) is the bubble radius, \( m \) is the electron mass, \( \sigma_s \sim 3.7 \times 10^{-4} \text{ N m}^{-1} \) is the surface energy per unit area and \( h \) is Planck’s constant. At zero pressure the radius at which the energy has a minimum is

\[ R_{b \text{min}} = \left( \frac{h^2}{32m\pi\sigma_s} \right)^{1/4}. \] (3.7)

This radius is 1.9 nm at 0 K and increases slightly as the temperature goes up. It is important to note that the polarization-induced electrostrictive forces which lead to the snowball formation in the case of positive ions will not play a role in electron bubble formation as \( R_b \gg R_{sb} \). As the electron mass is negligible, the effective mass of an electron bubble is the hydrodynamic mass (Equation 3.5) of 243±5 helium masses [99]. Similar to electrons, negative ions and heavy alkali earth positive ions form bubbles [46].

3.3 Snowball and bubble mobility

A direct implication of snowball or bubble formation by charged particles in superfluid helium is their smaller mobility compared to expectation for the bare positive ions or electrons. The mobility of ions in superfluid helium shows a wide variety of phenomena with strong and striking temperature and electric field dependencies. As the mobility depends mainly on the size of the entity in movement, both snowballs and bubbles show fairly similar mobility profiles. The differences can be explained by differences in size and structure, the positive ion being a high density complex of about 0.6 - 0.7 nm diameter and the electrons being a low density complex of about three times this dimension. At temperatures below about 0.6 K the mobility of snowballs and bubbles is limited mainly by collisions with thermal phonons [95]. At temperatures above about 0.8 K, snowball-roton and bubble-roton collisions start to play a major role in limiting the mobility. A roughly exponential temperature dependence of the mobility reflects the exponential population of roton states. In the “kinetic regime” \((0.8 \lesssim T \lesssim 1.7 \text{ K})\) roton-roton scattering can be neglected, while above about 2 K, roton-roton scattering is so frequent that the rotons appear to the ions as a viscous fluid [10]. In the “kinetic regime” the zero-field \((E \ll 1 \text{ V cm}^{-1})\) mobilities of snowballs and bubbles show a temperature dependence of the form \[ \mu_0 \propto e^{\Delta_+/k_BT}, \] (3.8)

where the value of \( \Delta_+/k_B \) is around 8.65 - 8.8 K and that of \( \Delta_-/k_B \) is around 7.7 - 8.1 K [89] (Figure 3.8). Figure 3.9 gives an overview of the behaviour of the snowball
and bubble drift velocity $v_d$ as function of applied electric field. The drift velocity $v_d$ is proportional to the electric field (i.e. the mobility is constant) up to 200 V cm$^{-1}$ [73]. At higher velocity, the mobility decreases due to the enhanced density of rotons in the disturbed region near the snowball or bubble, giving rise to an increased drag force. At an electric field $E_g$ and a drift velocity $v_g$ a transition from a bare snowball or bubble regime to a snowball or bubble-vortex ring regime occurs: the snowball or the bubble produces vortex rings and is captured by them. A further increase in vortex energy (by an increase in electric field) results in an increase in its radius causing $v_d$ to fall to a minimum value. The snowball is much smaller than the electron bubble and therefore much less tightly bound to its vortex ring, thus the thermally activated escape rate of the snowball from the vortex ring is large. The electron bubble is strongly bound to the vortex ring up to temperatures of at least 1.8 K, whereas the escape probability from the vortex ring for the snowball is already large at a temperature of about 1 K. Because of this, the drift velocities of bubbles and snowballs show a different behaviour for electric fields above $E_g$. As the energy (i.e. electric field) increases further, the vortex will be gradually shed from the snowball; the drift velocity increases and reaches a saturation value $v_r$ equal to $v_g$ within experimental errors (see Figure 3.9). This saturation occurs when the snowball creates vortex rings continuously without being trapped. The discontinuity in drift velocity above $E_g$ becomes less and less deep with an increase in
3.3 Snowball and bubble mobility

Figure 3.9: Drift velocity $v_d$ of positive and negative ions as a function of the electric field $E$ [17]. $v_g$, $E_g$ and $v_r$ are indicated for 1.3 K.

Figure 3.10: The critical velocity $v_r$ for the production of vortex rings by positive ions as a function of normal-to-total density ratio $\rho_n/\rho$ [17].
temperature and disappears completely at about 1.35 K. The velocity $v_r$ (measured as $v_g$ for $T < 1.35$ K) is temperature dependent above 1 K (see Figure 3.10). A detailed study of the phenomena can be found in [17, 16, 20, 84]. In the context of this thesis, we deal with electric fields well below $E_g$, concerning both snowballs and bubbles. The main interest of the current project is the fast transport of positive ions in superfluid helium so it’s worth to note that the snowball model proposed by Atkins has its limitation as far as a prediction for the mobility of positive impurities in superfluid helium is concerned. Mobility measurements of K$^+$, Rb$^+$ and Cs$^+$ ions show a mobility lower than predicted by the snowball model [13, 25, 50]. The heavier alkaline earth ions Ca$^+$, Sr$^+$, Ba$^+$ and Mg$^+$ form bubble-like defects instead of snowballs whereas the Be$^+$ ion form a snowball-like structures [46]. All ion mobilities fall within about 25% of the He$^+$ mobility (see Figure 3.11). For most practical purposes the He$^+$ mobility can thus be used.

### 3.4 Charge recombination factor in superfluid helium

G. Careri and F. Gaeta [21] measured the volume recombination coefficient $\alpha_{SF}$ of ions in superfluid helium in a range of 0.87 K to 2.0 K. In their experiment they sent two ionic beams of opposite charges against each other and calculated the recombination coefficient from the loss of charge suffered by each beam. They used
3.5 Positive ions at the superfluid-vapor interface

The extraction of positive ions (snowballs) from the superfluid helium surface to the vapor phase is not trivial as is that of electron bubbles [104, 105]. The electric potential barrier at the superfluid-gas interface is the bottleneck. The image charge potential model provides a simple way to look at this potential barrier. This model gives the electric potential distribution of a point charge in front of a dielectric interface. When a point charge $Q$ is in the vicinity of a dielectric interface, i.e. of order of a few tens of nanometers, its electric field polarizes the dielectric and a bound charge density is induced at the dielectric interface. No bound charge

\[ \alpha_{SF} \propto \exp(\Delta/k_B T) , \]  

with $\Delta/k_B = 8.3$ K between temperatures 2 K and 1.3 K and $\Delta/k_B = 9.9$ K at temperatures lower than 1.3 K. Figure 3.12 shows the recombination coefficient as a function of inverse temperature.

**Figure 3.12:** Recombination coefficient of ions in superfluid helium $\alpha_{SF}$ as a function of the inverse temperature $1/T$ [21].

$\alpha$ particles from a radioactive $^{210}$Po source to create a 0.2 mm thick densely ionized region in liquid helium from which beams of ions are drawn out by means of an applied electric field. They observed a general behavior of exponential increase in $\alpha_{SF}$ with the reciprocal of the temperature. This increase is represented by

\[ \alpha_{SF} \propto \exp(\Delta/k_B T) , \]  

with $\Delta/k_B = 8.3$ K between temperatures 2 K and 1.3 K and $\Delta/k_B = 9.9$ K at temperatures lower than 1.3 K. Figure 3.12 shows the recombination coefficient as a function of inverse temperature.

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is created in the bulk of the medium, as the polarizing field is a Coulomb field of a point charge. The sum of Coulomb forces between the point charge and the elements of the induced surface charges acts like the Coulomb force between the point charge and its mirror image $Q'$ [42]. The image charge potential is attractive for a charge on the low-dielectric-constant side and repulsive for the charges on the high-dielectric constant side of the interface, independent of the sign of the charge. Consider a dielectric interface between two media with the permittivities $\epsilon_1$ and $\epsilon_2$ (Figure 3.13). If the point charge $Q$ is placed at a distance $z$ from the interface, the induced image charge $Q'$ is

$$Q' = Q \left( \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + \epsilon_2} \right)$$  \hspace{1cm} (3.10)

and the image potential energy of a point charge $Q$ is

$$\phi_{im}(z) = \frac{1}{4\pi\epsilon(z)} \frac{QQ'}{4z}.$$  \hspace{1cm} (3.11)

This is half of the electric potential energy of a system of two real point charges due to the fact that no work is required to bring the image charge $Q'$ from the infinity [51].

This project concerns positive ions in superfluid helium and the possibility to extract them to the vapor phase. If an electric field $E$ is applied across the interface such that the point charge $Q$ is pushed towards the interface, the total potential energy $\phi_{eff}(z)$ is

$$\phi_{eff}(z) = \phi_{im}(z) + QEz.$$  \hspace{1cm} (3.12)

Figure 3.14 illustrates the electric potential energy of a unit point charge $Q$ at the superfluid helium-vapor interface. It is trapped in a potential well created by the image charge potential in combination with an externally applied electric field normal to the interface. The divergence and discontinuity of Equation 3.11 at $z = 0$ shows that the method of calculating the effective potential using a discrete dielectric interface is unphysical. This divergence is removed by considering a gradual transition of permittivity between superfluid helium and vapor phase [103]. This transition is associated with the gradual change of the superfluid density to the vapor density at the interface. This transition layer is a few atomic diameters thick (Figure 3.15). A mathematical model to represent this transition is a linearly graded transition with sinusoidally rounded corners as used by Stern [103]. A numerical solution for the image charge potential energy of a unit point charge $Q$ at an interface with such a permittivity profile (see Figure 3.15) is given by Stern (see Figure 3.16) [103]. A more realistic superfluid-vapor density transition profile is found in the experimental work of Lurio et al. [70, 71]. No significant variation of interfacial width is observed in a temperature range from 1.1 K to 1.8 K. The width of this interfacial density variation is 0.92(10) nm which is compatible with the 0.68 nm used in Stern’s model. The main difference is the variation profile (see Figure 3.15).
3.5 Positive ions at the superfluid-vapor interface

\[ Q' = \left( \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \right) Q \]

\[ F_{im}(z) = \frac{1}{4\pi \varepsilon_1 (2z)^2} \]

**Figure 3.13:** A point charge \( Q \) and the induced image charge \( Q' \) (at the superfluid helium-vapor interface). \( F_{im} \) is the image charge force experienced by the point charge \( Q \). \( QE \) is the electric force experienced by the point charge \( Q \) due to the applied electric field \( E \).

**Figure 3.14:** Electric potential energy \( \phi \) of a unit point charge \( Q \) as a function of the distance \( z \) from an abrupt superfluid-vapor helium interface. The dotted line shows the image charge potential energy \( \phi_{im} \) of a unit point charge \( Q \) as a function of the distance \( z \), the dashed line shows the electric potential energy of a unit point charge \( Q \) due to the electric field \( E = 300 \, \text{V cm}^{-1} \) applied perpendicular to the interface and the solid line shows the combination of both \( \phi_{eff} \).
Physics processes in superfluid helium ion catchers

3.6 Ion extraction across the superfluid-vapor interface

3.6.1 Extraction of electrons

There has been a lot of experimental and theoretical work on the electron extraction across the superfluid-vapor interface [94, 101]. An interfacial potential energy barrier of $\sim 25 - 40$ K (depending on the applied electric field) and a characteristic trapping time of 1 - 100 s have been measured for electrons [94].

Interaction of the electron and superfluid helium constitutes of two factors, the strong short-range repulsion, which arises from the requirement of the Pauli exclusion principle and a long-range attraction due to the polarization potential, represented by the classical image charge potential. It is important to note that the large potential step ($\sim 1$ eV) due to the Pauli repulsion is 45 times larger than the image-charge-induced attractive potential when the electron approaches the interface from the vapor phase. Thus the Stern image charge model is insufficient to explain the interaction of electrons with the superfluid-vapor interface.

A theoretical work by F. Ancilotto and F. Toigo [5] showed that if an electric field is applied such that an electron bubble inside the liquid helium is pushed towards the superfluid-vapor interface, it is stable up to a distance $\sim 2.3$ nm. If it comes closer to the surface due to thermal motion, the bubble bursts and the electron is
ejected into the vapor. They obtained a potential energy barrier of 38 K for the thermal emission. At distances larger than $\sim 2.3$ nm quantum tunneling through the surface layer dominates the extraction probability [5]. Theoretical works suggest that the diffusive nature of the superfluid-vapor interface does not play a big role in electron extraction phenomena [5, 23].

3.6.2 Extraction of snowballs

Contrary to electron bubbles, positive ion extraction is reported to be extremely weak [15, 104, 105]. There is not much information available on snowball-interface interactions in the published literature. Extraction of $^{219}$Rn ions across the superfluid-vapor interface is reported by Huang et al. with an extraction efficiency of tens of percents at 1.6 K [56, 57]. There is no theoretical model available to understand positive ion extraction. In the case of snowballs, the huge mass compared to the electron, makes quantum tunneling an improbable candidate for an effective extraction mechanism. It is important to keep in mind that the structure and sizes of the electron bubbles and snowballs are very different (see section 3). The size of the snowballs is comparable to the width of the interfacial density variation and there is no repulsive force between snowballs and the ambient medium; thus in this case the diffusive nature of the interface may play an important role contrary to the electron bubble case. Stern’s potential barrier model is to be considered as
a first approximation in the case of snowballs, this point is elaborated upon is Section 6.1.3. A similar potential energy barrier calculation with the interfacial density transition profile given by Lurio et al. [70, 71] and inclusion of the surface energy may give a more accurate picture.

As the quantum tunneling effect can be ruled out in the case of snowballs, it’s more realistic to consider a Maxwell-Boltzmann energy distribution of snowballs trapped in a potential well created by the image charge potential in combination with an externally applied electric field normal to the interface. A fraction of ions have energies higher than the potential energy barrier. These ions will cross the interface into the vapor phase. Kramer [64] and Chandrasekhar [22] have shown that the escape rate $P_{th}$ of particles from a potential well in consequence of Brownian motion is

$$P_{th} \propto \exp\left(-\frac{E_b}{k_B T}\right).$$

where $E_b$ is the height of the potential energy barrier and $T$ is the temperature of the system. Measuring the extraction efficiency of positive ions as function of temperature gives the height of the potential energy barrier (see Section 6.1.2).

### 3.7 Conclusion

This chapter gives an overview of positive helium ion and electron properties in superfluid helium. In general, positive ions form “snowballs”, i.e. the density around the ion is increased and electrons form “bubbles”, i.e. a cavity is formed around the object. The mobility and drift velocity of positive ions in superfluid helium is important to understand the experimental results on the snowball efficiencies presented in Chapter 6. The interaction of snowballs with the superfluid-vapor interface is not yet well understood. The barrier height and location of the potential minimum at the superfluid-vapor interface depend strongly on the details of the density profile at the interface. An accurate theoretical description of the extraction of positive ions and the height of the potential energy barrier is not available yet. Thermal excitation across a potential energy barrier is the most probable explanation.