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Negative ions in liquid helium

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The structure of negative ions in liquid ^{4}He is analyzed. The possibility of cluster or bubble formation around impurity ions of both signs is discussed. It is shown that in superfluid helium, bubbles form around negative alkaline earth metal ions and clusters form around halogen ions. The nature of "fast" and "exotic" negative ions is also discussed. It is assumed that "fast" ions are negative ions of helium excimer molecules localized inside bubbles. "Exotic" ions are stable negative impurity ions, which are always present in small amounts in gas discharge plasmas. Bubbles or clusters with radii smaller the radius of electron bubbles develop around these ions.
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I. INTRODUCTION

Positive helium ions and electrons in liquid helium exhibit very low mobilities. This is because a spherical region of solid helium with a radius of about 0.7 nm develops around an ion owing to a polarization interaction with the atoms of the liquid, while an electron is localized in a bubble, whose radius is close to 2 nm, owing to a strong exchange interaction.[1–3] The mobility has also been measured for a number of positive impurity ions in superfluid ^{4}He.[4–6] A qualitative difference between the mobilities of alkali and alkaline-earth metal ions was found (see Table I): the mobilities of alkali metal ions were smaller than the He^+ mobility and decreased with atomic number, but the mobilities of the alkaline-earth metal ions (other than Be^+) were higher than the He^+ mobility and increased with atomic number. These differences cannot be interpreted in terms of the simple electrostatic model proposed by Atkins,[7] in which the structure of a complex depends only on the ionic charge. Cole and Bachman[8] gave a qualitative explanation for the observed effects. In their analysis, the radius of the ionic complex depends strongly on the extent of the wave function of the lone valence electron, which creates a repulsive interaction with the surrounding helium. In the case of the alkali metal ions, the van der Waals interaction of the ion core with helium atoms plays an appreciable role. This interaction results in an increase in the radius of the solid ion complex and in a dependence of this radius on atomic number. In the case of alkaline-earth metal ions, the valence electrons have extended orbits and the formation of empty voids around the ions is possible. This effect is related to the strong exchange interaction of the valence electrons with the shell electrons of the atoms of the surrounding liquid.

The structure and transport properties of electrons and positive ions in low temperature atomic liquids have been well studied. Much less is known about the properties of negative ions. The mobility of O_2^- in Ar, Kr,[9] and Xe (Refs. 9, 10) has been studied in a few papers. Berezhnov *et al.*[11] discussed the possibility of the bubble formation around H^- ions in liquid hydrogen. Experimental data on the mobility of electrons in liquid hydrogen and deuterium at the saturation curve[12,13] are in a good agreement with the electron bubble model in the region of relatively high temperatures from 22 to 32 K for liquid hydrogen and in deuterium at all temperatures. However, in liquid hydrogen at low temperatures from 17 to 22 K, Levchenko and Mezhov-Deglin[13] measured an anomalously high mobility of negative charge carriers, 1.5 times higher than the mobility observed earlier by Sakai, Böttcher, and Schmidt.[12] Levchenko and Mezhov-Deglin interpret this difference in terms of the experimental conditions. In the work of Sakai *et al.*[12] excess electrons were injected into the liquid by photoemission from the cathode. The energy of these electrons is $\sim$1 eV, too small to dissociate molecular hydrogen. Levchenko and Mezhov-Deglin[13] created excess electrons by β-decay of tritium atoms. The energy of these electrons is $\sim$10 keV, large enough not only for ionization but also for multiple dissociation of molecular hydrogen. Therefore, in the latter experiment a significant concentration of hydrogen atoms was generated near the track of a β-particle in the liquid. These atoms, as opposed to hydrogen molecules, are able to form stable negative ions. The anomalous mobility of negative charges at low temperatures in β-irradiated liquid hydrogen corresponds to the mobility of H^- ions. Levchenko and Mezhov-Deglin assumed that at low temperatures, clusters are formed around the negative ions of atomic hydrogen which move as a single entity in the liquid. However, calculations by Berezhnov *et al.*[11] have shown that bubble creation around negative H^- ions is energetically more favorable. The mobility of the negative ion bubble is higher, as observed in the experiment.[13]

II. NEGATIVE IMPURITY IONS

The mobility of negative impurity ions in superfluid ^{4}He was measured by Kasimov *et al.*[14] The mobilities of the negative ions of both halogens (Cl^-, F^-, and I^-) and alkaline-earth metals (Ba^- and Ga^-) were found to be lower than the mobilities of electron bubbles as well as of He^+ ions (see Tables I and II). Evidently, only the formation of multiatomic complexes (clusters or bubbles) around the ions can be responsible for these low mobilities.

TABLE I. Mobilities of positive impurity ions in liquid ^{4}He at T = 1.3 K (Ref. 5).

Ion	He^+	K^+	Rb^+	Cs^+	Be^+	Ca^+	Sr^+	Ba^+
μ, cm^2/(V·s)	0.88	0.85	0.78	0.78	0.81	0.98	1.01	1.12

The properties of negative impurity ions in liquid helium have been studied.[11,15–19] It was found that the binding energy E of the outer-shell electron in a negative ion (electron affinity) increases by about 1 eV in a liquid dielectric and a spherical cavity is formed around the ion. The radius of this cavity depends on the characteristics of the negative ion, as well as on the thermodynamic parameters of the liquid. The negative ion *in vacuo* is formed by a long-range polarization attraction and a short-range exchange repulsion between the outer-shell electron and the ion core. The following simplest model potential can be used as a potential for the interaction of an electron with its atom:

$$V_i(r) = \begin{cases} -\dfrac{\alpha e^2}{2r^4}, & r > R_c, \\ \infty, & r \le R_c, \end{cases} \tag{1}$$

where α is the atomic polarizability, e is the electron charge, and R_c is the radius of the solid atomic core. It is caused by the exchange interaction of the outer-shell electron with electrons of internal atomic shells. Table III lists the solid core radii R_c obtained by solving the Schrödinger equation for an electron in the potential of Eq. (1) with known polarizability α and electron affinity E. The asymptotic behavior of a wave function away from a repulsion center has the form $\Psi(r) = r^{-1}\exp(-r/\lambda)$. The characteristic size of the region of the spatial electron localization depends on the attachment energy, $\lambda \cong \sqrt{\hbar^2/2mE}$. The electron affinity E is usually much lower than the ionization potential I; because of this, λ is much greater than the size of the corresponding atom or molecule. Most of the time a weakly bound electron is away from the ion core and interacts with the atoms of the surrounding liquid similarly to a free electron. The exchange interaction results in the formation of a spherical cavity of radius R around the ion. The electron potential energy undergoes a jump of approximately 1 eV at the boundary of a cavity. In our calculations we use a model potential for the interaction of the outer-shell electron of a negative ion with the atoms of the liquid V_l proposed by Stampfli.[20] The binding energy E_e of the outer-shell electron of a negative ion placed in a cavity of the liquid was found by solving the Schrödinger equation with the resulting potential $V(r) = V_i(r) + V_l(r)$, and the equilibrium radius of the cavity R was found subject to the condition that the free energy

$$F(R) = -E_e(R) + 4\pi\sigma R^2 + (4\pi/3)pR^3 \tag{2}$$

TABLE II. Mobilities of negative impurity ions in liquid ^{4}He at T = 1.3 K (Ref. 14).

Ion	*e*-bubble	Cl^-	F^-	I^-	Ba^-	Ga^-
μ, cm^2/(V·s)	0.54	0.46	0.47	0.45	0.47	0.41

TABLE III. Atomic core polarizability α, electron affinity E, core ionization potential I, solid core radius R_c, van der Waals constant C_6 for the interaction of the core with helium atoms, and radius R of the cavity around the negative ion.

Core	α, a_0^3	E, eV	I, eV	R_c, a_0	C_6, a_0^6	R, a_0
e-bubble	–	–	–	–	–	32.1
Cl	15	3.61	12.97	0.97	9.8	5.71
F	3.8	3.40	17.42	0.51	2.9	5.05
I	24	3.06	10.45	1.13	13.5	6.35
Ba	283	0.14	5.21	4.08	93.3	20.7
Ga	33.6	0.41	6.00	1.52	12.4	19.8
He_2^*	316	0.18	4.77	1.79	67.0	15.4
O_2	10.6	0.46	12.1	0.91	6.6	11.0
O	5.4	1.46	13.6	0.63	3.6	6.8
H	4.5	0.75	13.6	0.60	3.0	8.5

is at a minimum (σ is the surface tension coefficient and p is the pressure in the liquid). Table III summarizes the results of this calculation.

One can see that the sizes of the cavities around the halogen and alkaline-earth metal ions are significantly different. First, let us discuss properties of the complexes formed around negative halogen ions. In terms of our model, the radii (5–6) a_0 of the cavities in which these ions are localized are much smaller than the radii of the solid clusters formed around He^+ ions (14.9 a_0) and alkali metal positive ions (15.8 a_0).[5] This suggests that clusters are formed near negative halogen ions. In this case, the presence or absence of a cavity within a cluster is of little importance for determining the radius of these clusters: as in the case of positive ions, the negative ions at the center of a cluster can be regarded as point ions. To understand the reason for the considerable difference between the mobilities of the He^+ ion, on the one hand, and negative halogen ions, on the other hand, Khrapak[17] has drawn on the reasoning that was used to explain the small differences in the mobilities of positive helium and alkali metal ions.[8] Although the polarization interaction of an ion with helium atoms outside the cluster is equal for all the ions, the additional van der Waals interaction of helium atoms with the ion core depends on its atomic number. An excess pressure results in an increase in the cluster radius and a decrease in the cluster mobility. This effect is even more important in the case of negative ions. The potential energy of the interaction of a helium atom arranged near the surface of a cluster with a point ion placed at the center of the cluster is

$$V_a(r) = -\frac{\alpha_{He} e^2}{2r^4} - \frac{C_6}{r^6}, \quad C_6 \cong \frac{3}{2}\frac{I_{He} I_a}{I_{He} + I_a}\alpha_{He}\alpha_a. \tag{3}$$

Here C_6 is the van der Waals constant for the interaction of helium atoms with the atom in the ion core, which was evaluated using the London formula.[21] Table III summarizes the constants C_6 calculated in this way. With increasing C_6, the cluster radius has to increase and the mobility has to decrease in accord with the small mobility changes that are observed experimentally.[14] However, this effect can decrease the mobility of negative halogen ions by 5–10% relative to the mobility of the He^+ ion, but hardly by a factor of 2.

In the case of the alkaline-earth metal ions Ba^- and Ga^-, which have low electron affinities E in a vacuum, the bubble radius is large enough that creation of a solid shell around the bubbles is unlikely. According to our estimates, the electron binding energies E_e for these ions in liquid helium at $T = 1.3$ K are similar and equal to 1.42 and 1.46 eV, respectively. The difference between the characteristic extent of the wave functions, λ, for these ions is small; this fact is ultimately responsible for the observed similarity of the ion mobilities. At first glance, the fact that the mobility decreases with decreasing bubble radius ($Ba^- \rightarrow Ga^-$) is surprising. However, note that, at $T = 1.3$ K on the saturation curve of liquid ^{4}He, the mobility of the ion complexes depends on scattering by rotons.[1] Bondarev[22] has shown that the density of rotons increases with decreasing distance to the ion complex because of the polarization attraction. In the case of electron bubbles this effect does not play a significant role because of their large radii. In the case of Ba^- and Ga^- ions, the polarization interaction of the helium atoms situated in the vicinity of the bubble surface is important ($\alpha e^2/2R^4 = 1.2$ K for Ba^- and 1.4 K for Ga^-) and leads to a significant increase in the roton concentration near the ion bubbles. This effect may be responsible for difference of the mobilities of Ba^- and Ga^- ions. However, the question of why the mobilities of the alkaline-earth metal ions are less than that of e-bubbles is still open.

III. FAST AND EXOTIC IONS

In addition to the "usual" electron bubbles in superfluid helium, two kinds of negative charge carriers are observed: "fast"[23–27] and "exotic"[24–27] ions. The mobility of fast ions is about 7 times higher than the electron bubble mobility and the mobilities of several different exotic ions lie in between. The ions were produced by different methods: an α source placed in the liquid,[23] a β source and gas discharge placed above the liquid,[24,25] and a gas discharge, alone, above the liquid.[26,27] The mobility of the electron bubbles around 1 K is determined by collisions with rotons and is proportional to square of the bubble radius R. Assuming that voids or clusters are created around fast and exotic ions, the radii of these complexes can be estimated assuming that their mobility is proportional to the square of the radius.[3,25] They lie between 30.4 a_0 (electron bubbles) and 11.8 a_0 (fast ions). At low electric fields, the electron bubbles are in thermal equilibrium with the gas of scatterers (rotons) and the bubble drift velocity is proportional to the electric field. With increasing electric field the bubble energy also increases and the field dependence of the drift velocity becomes weaker. Finally, at some critical electric field, the drift velocity undergoes an abrupt change, known as a giant fall or giant discontinuity.[28] This is a result of the creation of a charge-carrying quantized vortex rings in the superfluid. The same effect is observed with the exotic ions but not with the fast ions.[26] This may be evidence of a difference between fast and exotic ions. Measurements show that the critical velocity v_c for nucleation of vortex rings by exotic ions is larger than for electron bubbles, and that among the different exotic ions, v_c increases as the mobility increases. Theory predicts that the critical velocity of an ion should increase with decreasing of the ion radius.[29] Thus, the measured critical velocities also indicate that the exotic ions are smaller than the normal electron bubbles.

Strangely, the nature of the fast and exotic ions is still unknown. Several models have been proposed for these negative charge carriers,[24,30] but none can interpret all experimental data.[3,31] Below we suggest a new model. Experiments show that fast and exotic ions are different. The two kinds of ions are not observed in conventional experiments: both kinds have been observed in gas discharge experiments and exotic ions are not observed in experiments with radioactive sources. Let us consider their properties separately.

We suggest that fast ions are bubbles created around negative ions of the excimer He_2^* in the triplet state. In contrast to "usual" experiments, where electrons are photoinjected into liquid helium with an energy on the order of 1 eV, radioactive sources or gas discharges both ionize and excite atoms and molecules. Singlet atomic and molecular excitations decay rapidly to the ground electronic state, but the long-living triplet species quickly thermalize and form bubbles around themselves. With a 15 μs time constant triplet He* combines with another helium atom to produce He_2^* in a highly excited vibrational state ($v = 16$). This state relaxes to its ground vibrational state with time constant of about 30 μs.[32] The lowest electronically excited state in liquid helium is the triplet excimer He_2^* ($^a\Sigma_u^+$) which lies at 17.8 eV above the ground state. Due to the very weak spin-orbit coupling in He, this state is metastable with a long intrinsic lifetime of about 15 s.[33–35] Spectroscopic studies show that excimers occur in bubble states. The bubble model was employed by Hickman *et al.* to analyze the spectral shifts of He* (Ref. 36) and He_2^* (Ref. 37) in liquid He. The early studies have been confirmed by the later spectroscopic studies.[38–40] Theoretical estimates of the bubble radius give about $12a_0$ for He* (Ref. 41) and $13a_0$ for He_2^* (Ref. 42).

The long-lived metastable negative excimer ion He_2^- was first observed in 1984 (Ref. 43) and now its properties in vacuum are well known. The $^4\Pi_g$ state has a $1\sigma_g^2 1\sigma_u 2\sigma_g 2\pi_u$ electronic configuration, its electron affinity E is 0.18 ± 0.03 eV, and its lifetime is 135 ± 15 μs; only the $v = 0$ vibrational state is responsible for this long-lived state.[44] This ion can exist in liquid helium as result of excitation by α or β particles, or as result of diffusion through the surface from a gas-discharge plasma. However, the properties of He_2^- ions in condensed helium are quite unknown. It is clear enough that, like the excimer He_2^*, the He_2^- ion is localized inside an empty void. The size of the void can be estimated using the model employed in the previous section for impurity negative ions. For this, it is necessary to estimate parameters α, R_c, and C_6 of the interaction potential. The main contribution to the polarizability of the excimer He_2^* is from the excited atom He*, i.e., $\alpha_{He_2^*} \simeq \alpha_{He^*}$. The atomic polarizability can be estimated from its ionization potential, $\alpha \sim a^3 \sim I^{-3}$, where a is the radius of the atom. The excitation energy of the helium atom is 19.82 eV and the ionization potential $I_{He^*} = 24.59 - 19.82 = 4.77$ eV. Thus,

$$\alpha_{He_2^*} \simeq \alpha_{He}\left(\frac{I_{He}}{I_{He^*}}\right)^3 \simeq 204\, a_0^3. \tag{4}$$

The core radius R_c is found by solving the Schrödinger equation with known E and $\alpha_{He_2^*}$. This gives $R_c = 1.79$ a_0. The constant in the van der Waals interaction of the core (He*) with helium atoms can be estimated using the London formula

$$C_6 \simeq \frac{3}{2}\frac{I_{He}I_{He^*}}{I_{He}+I_{He^*}}\alpha_{He}\alpha_{He^*} \simeq 67\ a_0^6. \tag{5}$$

Figure 1 shows the free energy of a spherical bubble around an He_2^- ion as a function of its radius. The equilibrium bubble radius is equal 15.4 a_0, less than half the radius of an *e*-bubble. Note that the wave function of the $^4\Pi_g$ state of He_2^- is not spherically symmetric. As a result, the bubble shape has a characteristic two-lobe peanut structure similar to an excited *e*-bubble in the ^1P state.[45] This effect could reduce the roton scattering cross section of the bubble and explain the observed rise in the fast ion mobility.

The lifetime of the He_2^- ion in condensed helium is still an open question. The experimentally measured drift time[24–27] is about 1 ms, several times the vacuum lifetime of the ion. However, the wave function of the outer electron of the He_2^- ion undergoes significant alterations which can increase the ion lifetime. Another way to explain the difference between the short lifetime and long drift time is the following: during the drift time between electrodes, electrons have several chances to escape from the ionic complex with subsequent capture by another excited molecule. After escaping from an ion with an energy of about 19 eV, the electron is thermalized within approximately 1 ps,[2] after which it can be captured by recombination or it can create an *e*-bubble. If the concentration of the excimer molecules He_2^* in liquid helium is high enough, then electrons will essentially always be localized in He_2^- bubbles. These complex problems lie beyond the scope of this paper.

Exotic ions were observed in superfluid helium only when ionization of helium was produced by gas discharge above the liquid surface. There is good reason to believe that these ions are stable negative impurity ions, which are always present in small amounts in a gas discharge plasma. Although all impurities are frozen out in superfluid helium, atomic and molecular impurities may be present in the discharge plasma as a consequence of etching from the discharge vessel and electrodes. Unfortunately, the plasma composition in the experiments[24–27] is unknown. Here we give some estimates for several of the most probable candidates for the role of the exotic ions, such as O_2^-, O^-, and H^-. All these ions are localized inside bubbles. The free energies of these ions as functions of bubble radius are plotted in Fig. 2. The equilibrium radii of these ions are listed in Table III. The radius of an O_2^- bubble is a third of that of an *e*-bubble and this ion can be responsible for the mobility of one of the fastest exotic ions. The size of an O^- bubble or cavity is much smaller than the radius of cluster usually

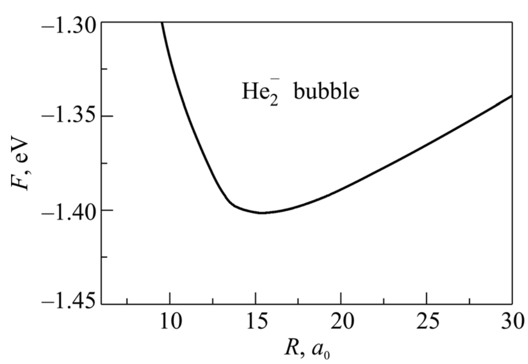

FIG. 1. Free energy of an He_2^- bubble as a function of radius in superfluid helium.

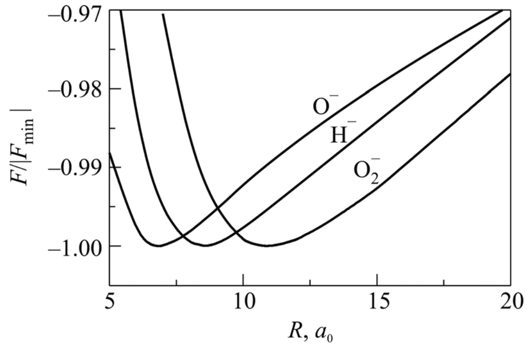

FIG. 2. Free energies F of some negative impurity ions normalized to the minimum value F_{min} as functions of bubble radius in superfluid helium.

formed around positive helium ion. As in the case of the negative halogen ions, this points to the formation of a solid cluster around the O^- ion, with a void inside. The size of this complex has to be close to that of an He^+ cluster (14.9 a_0), and its mobility must exceed two or three times the *e*-bubble mobility. The H^- ion is an intermediate case: the bubble is surrounded by a layer of dense but probably not solid helium. More detailed and comprehensive studies are needed to determine its structure and that of similar exotic ions.

IV. CONCLUSIONS

In this paper we have discussed the properties of various negative impurity ions in liquid helium. With help of a simple model it was found that, owing to the strong exchange interaction of the outer electron of a negative ion, a void is always created around an ion. In the case of ions with a high enough electron affinity, a layer of solid helium can develop around the void, and this complex is a cluster rather than a bubble. It has been shown that complexes formed around negative alkaline-earth metals and halogens have qualitatively different structures (bubbles and clusters, respectively), although the measured values of their mobility are similar. It has been shown that "fast" ions are negative ions of the excimer He_2^* localized in a non-spherical bubble. As for "exotic" ions, we have assumed that they are produced in a gas discharge plasma by etching of the vessel walls and electrodes and are then injected into the liquid.

Additional experimental and theoretical studies are needed for better understanding of properties of impurity negative ions in superfluid helium and other dielectric liquids.

In an experiment by Kasimov, *et al.*[14] the mobility of negative impurity ions injected into liquid helium was lower than the mobility of *e*-bubbles. But the the well known mobility of *e*-bubbles[1,46] was not measured there. Our estimates show that the radius of these charged complexes is about 15 a_0 (halogen negative ion clusters) or 20 a_0 (alkaline-earth metal negative ion bubbles). As a result, the mobility of impurity ions should be several times that of the *e*-bubbles, in conflict with the experiment. Additional experiments with simultaneous measurement of the ion and *e*-bubble mobility, as well as with a wider range of ions, are desirable. Measurements of the critical velocity for nucleation of vortex rings by impurity ions could also give important information about the size of these ionic complexes.

Theoretical studies of the properties of He_2^- ions in liquid helium are of obvious interest. It would be rewarding to perform density-functional calculations of the equilibrium shape of non-spherical bubbles similar to recent calculations for excited *e*-bubbles.[19,42,45] Knowledge of the bubble shape and of the outer electron wave function will make it possible to estimate the lifetime of the He_2^- ion in liquid helium. The kinetics of bubble formation around He_2^- and possible electron trapping by excimer He_2^* are worthy of investigation.

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