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Cryogenic electrolytes

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The emphasis is made on experimentally observed indications of the presence of metastable ion dipoles in solid helium. Similar quasiparticles possessing positive scattering length for injected electrons are assumed to exist in liquid phases of cryogenic liquids. The observed phenomena allowing to detect and monitor the behavior of dipole gas in superfluid helium (referred to as cryogenic electrolyte) are discussed. Most interesting among these phenomena are: special features of the dielectric behavior of ion dipole gas, details of the temperature dependence of the ion dipole gas osmotic pressure at the boundary of the liquid ^{3}He–^{4}He solution stratification, relaxation phenomena of collective origin in cryogenic electrolytes, and liquid helium phonon spectrum transformation due to strong interaction between phonons and heavy dipole quasiparticles.

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1. Introduction

The cloud of charged particles produced by laser irradiation of a cesium (rubidium) substrate at the bottom of a pressure chamber filled with helium and carrying zero total charge was shown in Refs. 1–3 to exert a surplus pressure on the lattice of solid helium. Most prominently this effect is observed when an ion column with the size of the order of the laser beam radius is produced in the solid helium matrix and then the external pressure reduced down to and the persistence of $P \le P_c$, where $P_c \sim 25$ bar is the critical crystallization pressure of ^{4}He. Unexpectedly, under these conditions a local "stalactite" (the authors of Refs. 1–3 refer to this structure as "iceberg") visually remains in the crystalline state.

Basic results obtained in Refs. 1–3 are related to optical properties of alkali metals imbedded into solid helium. Traditionally (since 1970's, e.g., see Refs. 4–6), experiments of that kind dealt with nonthermal excitations in various cryogenic media and, in particular, addressed the effects produced by helium of different densities on the outer electron shells of halogens. For our purposes, extensive experimental data (contained both in original and review papers and gathered in Refs. 1–3) can be classified into the following three rather conventional kinetics frequency ranges: optical (electron transitions involving atomic shells of the particles taking part in the transition), intermediate (metastable excitations, such as excitons, excimers, exciplexes), and relatively slow relaxation. In the last case the evolution is mainly governed by the atomic diffusion parameters of the problem. In semiconductor physics, the relevant analogue is the slow fluorescence where the exciton recombination is suppressed by trapping of excited electrons and holes by the intermediate levels [7] (see also, [8–11]). Within this classification scheme, one particular result reported in Refs. 1–3 concerning the anomalously long (compared with the estimates (1) and (2) below) stalactite lifetime is of special interest. According to remarks made in Refs. 1–3, the stalactites can persist for hours (no special observations to find the upper limit on their lifetime were performed). The authors of Refs. 1–3 believe that the stalactites are only one of the yet few manifestations of a curious and rather general phenomena. Nobody has yet succeeded in combining the prolonged striction effects of electric charges on helium with quenched recombination even within the scenarios outlined in Refs. 7–11. One can only assume that the final part of inevitable mutual approach of oppositely charged particles stops at some intermediate stage where a system of stable ion multipoles (the simplest one being a dipole consisting of a single-electron bubble and a "snowball") without further transition of the electron into the atomic shells of positively charged helium atoms. Schematically, the ion dipole is shown in Fig. 1. Just as single charges, loose dipoles can produce (although to a less extent) a compressing effect on the helium lattice right until their possible recombination.

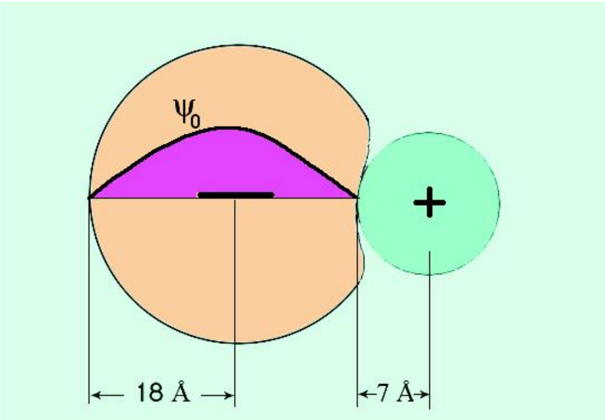

Fig. 1. Schematic diagram of the ion dipole (ψ_0 is the electron ground state wave function). Typical sizes of the single electron bubble and the snowball indicated in the figure are taken from Ref. 12.

The present work provides description and estimates for the processes associated with the natural tendency of oppositely charged ions to form bound complexes. Obviously, the contact stability of the structure consisting of a snowball and a single-electron bubble cannot be proved phenomenologically (generally, finding the equilibrium properties of the dipole is a very hard nonlinear problem), and one should just rely on the direct evidence [1–3] indicating the existence of these dipoles. The paper concerns with the observable consequences of possible existence of a stable solution of these dipoles in liquid helium. Considered are some proposals for diagnostics of a superfluid dipole solution allowing to establish their existence and treat the situation as a whole as an example of a cryogenic electrolyte. Of special interest are the consequences of the coupling arising between the massive dipoles and the liquid helium phonons (for details, see the section "Some properties of cryogenic electrolytes").

2. Typical processes involved in formation of ion dipoles

To get a quantitative estimate for the characteristic relaxation time τ_{He} of excited electron-ion cloud in cryogenic media it is useful to consider the processes occurring during formation of cosmic ray tracks in cryogenic gas chamber (the problem well known since the pioneering work of Wilson [8]). Analysis [9–11] reveals that at qualitative level kinetics governing positive and negative charges approaching each other under the conditions of [8] is described by Smoluchovsky equation involving correlation functions of different complexity levels. At the final (which is simultaneously the slowest one) stage this equation assumes the form of Eq. (1) for the pair correlation function $c(r,t)$ complemented with a suitable boundary condition which ensures at the last stages the recombination of a pair of charges into neutral molecule

$$\frac{\partial c}{\partial t} = \nabla\left[D\left(\nabla c + \frac{c}{T}\nabla U \right)\right],\ U(r) = e^2/(\varepsilon r),\ c(R_i,t) = 0, \tag{1}$$

where $c(r,t)$ is the current probability of finding a particle at a distance r from the attracting central body of radius R_i in the Coulomb field in the presence of diffusion coefficient D and ε is the media dielectric constant. In that case the problem features two parameters. One of them is the characteristic distance r_c, the other one — the typical charges mutual approach time τ

$$e^2/(\varepsilon r_c T) \simeq 1, \quad \tau \simeq r_c^2/D\ . \tag{2}$$

The details of the solution of Eq. (1) depend on the relationship between the initial relative position r_0 of charged particles and the scale r_c: either $r_0 < r_c$ or $r_0 > r_c$. In both cases the time τ (2) remains the typical time scale in the relevant scenario. For the situations involving the iceberg-effect [1–3], i.e., under the conditions $\varepsilon \simeq 1.08$, $T \simeq 1.5$ K the typical length scale is $r_c \simeq 10^{-3}$ cm. Bearing also in mind the available data on D in helium (at pressure $P_c \simeq 26.6$ bar (crystallization pressure), $D \sim 10^{-8}$ cm^2/s [12]) one obtains for τ_{He}

$$\tau_{\mathrm{He}} \sim r_c^2/D_{\mathrm{He}} \le 10^2\ \mathrm{s}. \tag{2a}$$

This estimate is considerably shorter than the observed [1–3] lifetime of the iceberg-effects whose upper boundary has actually not been established (exceeds several hours).

The approach involving Eqs. (1), (2) assumes the ions recombination when they approach each other at a distance $r \sim R_i$ (which is why the boundary condition $c(R_i,t) = 0$ (1) is imposed). This recombination produces a rather weak but still optically detectable luminescence (phosphorescence) [7]. The authors of Refs. 1–3, although employing time-resolved techniques and reporting observation of fluorescence with the duration of several hundreds microseconds for the kinetics with intermediate frequencies, do not mention anything on this phenomena at the time scale of about τ_{He}. Thus, two phenomena are clearly observed: anomalous lifetime of the macro-stalactite and the absence of any signs of fluorescence from the pace they occupy. If this is true, one can talk of the suppression of the relaxation process of the system of charged particles to the equilibrium state at the level of loose dipoles consisting of a single-electron bubble and a snowball or similar more complex multipole structures. At the same time, there are no visible reasons which could prevent preservation of such metastable formations also in liquid phase where the "merging" process can be identified with the association phenomena typical of oppositely charged ions in the electrolyte of arbitrary nature.

It is appropriate here to address the local mechanisms of preserving helium in solid state under the action of the system of ions (dipoles) whose finite density is necessary for observing the iceberg-effect. One of the possibilities mentioned by authors of Refs. 1–3 is related to the existence of a higher density domain in the vicinity of each ion. In liquid helium this domain has a finite radius R_{sn} usually assumed to have been introduced by Atkins [13] (correct description of the solid phase nucleation center involving electrostriction effects can be found in the textbook [14] which mentions Refs. 8–11). In the range of $P_\infty \le P_c$ the Atkins radius has a finite value of the order of $R_{sn} \simeq 6$ Å. Under supercritical with respect to pressure conditions $P_\infty \ge P_c$ the presence of a finite (although rather low) density of nucleation centers in the form of snowball-ions is sufficient for maintaining of bulk helium plasma in the solid phase [14].

Another (less obvious) scenario of pressure increase within a spatial domain acted upon by laser radiation has a thermodynamic origin. Appearance of finite density of impurities (ions and dipoles) should result in local pressure enhancement δP of the osmotic nature [15]:

$$\delta P \simeq cT, \tag{3}$$

where c in the volume density of ions in the irradiated domain. Here the role of the membrane separating the solid solution containing ions from the liquid phase is played by the image force at the liquid–solid helium interface which is repulsive from the solid phase side and proportional to the square of the charge it acts upon.

3. Some properties of cryogenic electrolytes

Let us now consider several effects allowing to detect the presence of ion dipoles in liquid helium and study the details of their behavior; in liquid helium their finite equilibrium density could be rather useful for the physics of liquid helium as a whole introducing a new type of quasiparticles in the problem of calculating osmotic pressure and dielectric constant as well as providing a possibility of manipulating a new type of impurities in superfluid ^{3}He, etc.

First of all, we should discuss the mechanism of formation of finite dipole density in liquid helium. One of the possible ways — liquid irradiation by a weak radioactive source — has long been used to work with ions in helium. Among recent related references one should mention a series of works [16–18] on the study of stability of charged liquid helium surface. The source integrated in the substrate of the cell used to study the properties of helium films allowed to maintain the charged liquid helium surface in a state close to the critical one. If necessary, the supercritical conditions resulting in the loss of stability of the vapor–liquid interface could also be produced. The critical surface charge density n_c is easily estimated to be $n_c \sim 10^{10}$ cm^{-2} with average distance between charges of the order of 10^{-5} cm. This estimate allows to understand the processes occurring in bulk helium. In the absence of charge selection, the system considered in Refs. 16–18 can maintain within the bulk helium the charged pairs density not less than 10^{14-15} cm^{-3}.

Second, the dielectric constant ε is affected. Indeed, appearance of ionic dipoles with anomalously high dipole moment $p \simeq 20$ D in nonpolar liquid dielectric with small atomic polarizability $\alpha_0 \simeq 5\cdot 10^{-25}$ cm^3 can dramatically change the properties of ε (its value is raised, and it becomes temperature and field dependent).

Basic equations describing the volt-polar properties of the system of ion dipoles are [19]

$$\frac{\tilde{\varepsilon}_\parallel - 1}{4\pi} = \frac{< P >}{E_\parallel}, \tag{4}$$

$$< P > = Np < \cos\theta >, \ < \cos\theta > = L(\lambda) = \cot\lambda - 1/\lambda,$$
$$\lambda = pE_\parallel / T, \ \ p = e(R_- + R_{sn}).$$

Here N is the number of dipoles with strength p in the unit solvent volume, R, R_{sn} are the bubble and snowball radii, respectively, T is the temperature, $L(\lambda)$ is the so-called Langevene function, $\tilde{\varepsilon}_\parallel$ is the volt-polar (depending on the dimensionless filed strength λ) dielectric constant of liquid dielectric consisting of polar molecules (the subscript $\parallel$ is used to emphasize the existence of additional ion concentration dependent dielectric constant $\varepsilon_\odot$ [20] in the system of polar molecules). In the weak field limit

$$\tilde{\varepsilon}_\parallel |_{\lambda \to 0} \to \varepsilon_\parallel,$$

with the average dipole polarizability $\alpha_p = p^2 / 3T$.

It is interesting to note that in usual liquids the limit $\lambda \ge 1$ is practically unattainable. For dipoles in helium one has ($p \simeq 20$ Deb, $E_\parallel \simeq 1$ CGSE, $T \simeq 1$ K)

$$\lambda = pE_\parallel / T \sim 0.5,$$

i.e., transition to the domain $\lambda \ge 1$ can be easily realized.

Let us also estimate (at the level of polarizabilities) typical parameters related to ion dipoles. Instead of traditional effective polarizability

$$\tilde{\alpha}_p = \alpha_0 + \frac{p^2}{3T}$$

it is useful to have the estimate for relative importance of the contributions due to liquid with density N_0 and polar gas with the density N_1 in the form

$$N_0\alpha_0 \sim N_1 \frac{p^2}{3T} \ \text{ or } \ \frac{N_1}{N_0} = 3\alpha_0 T / p^2. \tag{5}$$

For helium,

$$\alpha_0 \simeq 5\cdot 10^{-25}\text{cm}^3, \ N_0 \simeq 2\cdot 10^{22}\text{cm}^{-3}, \ N_1 / N_0 \sim 10^{-8}. \tag{5a}$$

As already mentioned earlier these densities are quite achievable in today experiments [16–18].

Of special interest is the statistical behavior of the gas consisting of large and heavy ion dipoles (possessing effective radius $R_d \gg a$, a being the interatomic distance, and mass $M_d \gg m_4$, where m_4 is the helium atom mass) in liquid helium with well-defined quasiparticles, namely, phonons forming the helium normal component. The associated mass of a sphere in a normal liquid has a square root singularity at zero frequency $M_d(\omega) \propto \omega^{-1/2}$ due to finite viscosity (described by Navier–Stokes equations) [21,22]. In statistical applications this singularities cannot be removed by known methods of classical nonequilibrium thermodynamics (i.e., by introducing Einstein relations or by employing the more general detailed balance principle allowing to relate viscous dynamics to the equilibrium characteristics of the ensemble of particles which do not depend on kinetics of the system). As a result, the effective mass appearing in the Maxwell distribution for massive particles in the solvent proves to be energy dependent at low frequencies (this difficulty one could still put up with). However, what is even worse, the energy dispersion proves to have very bad structure: it depends on viscosity which should never take place in a consistent theory. The suggested way of lifting the arising conflict is to prohibit motion of massive particles with respect to the superfluid helium normal components. This forcible decision shifts the main efforts in the problem from the statistical behavior of massive impurities (in particular, helium ion dipoles) to the details of phonon dynamics in the presence of a finite density of impurities which is well known in physics of superfluids. The problem is also of interest for helium physics as a whole since normally superfluid helium does not accept any bulk impurities different from ^{3}He atoms.

Phonon vibrations in the systems with embedded impurities have long been thoroughly studied in the general case. Basic results obtained by Lifshits (e.g., see Ref. 23) concern phonon spectra in disordered systems. However, they can be easily extended to phonon dynamics in liquid helium. This is most naturally done in the low-frequency range which is of particular interest to us from the viewpoint of ion dipoles and where the so-called quasi-local vibrations due to impurities can be excited. Following Ref. 24 where the general formalism of Ref. 23 is described in detail, we cite here final results on the phonon spectra of liquid helium perturbed by the presence of finite density c of heavy impurities. In the low density limit

$$c < c_* \simeq [m_4/(M_d - m_4)]^2 = [m_4/(\Delta m)]^2 \ll 1,$$

$$\omega^2 = \omega_0^2(k)\left[1 + c\frac{\varepsilon_*[\omega_0^2(k) - \omega_d^2]}{[\omega_0^2(k) - \omega_d^2]^2 + \delta^2}\right], \quad \varepsilon_* \simeq \omega_D^2. \qquad (6)$$

In the range of $\omega \ll \omega_k$ the general dispersion law reaches the asymptotics

$$\omega = sk, \quad s = s_0/n_0, \quad n_0 = \sqrt{1 + c(\Delta m/m_4)}. \qquad (6a)$$

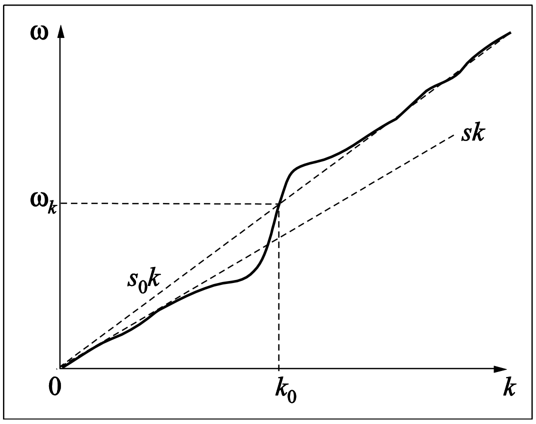

Fig. 2. Phonon spectrum of helium in the limit of low concentration of massive impurities, $c < c_*$. The branch $\omega = s_0 k$ represent unperturbed phonons with the sound velocity s_0. $\omega = sk$ is the asymptotics (6a) for the dispersion law for the case of finite density of heavy impurities. At the point $k = k_0$ found from the condition $\omega_0(k) = \omega_k$, the frequency shift is zero.

Here $\omega_0^2(k)$ is the phonon dispersion law for zero impurities density, k is the phonon wavenumber, ω_D is the Debye frequency, ω_d is the heavy impurity quasi eigenfrequency, and δ is its decay rate. The origin of these parameters in the problem "heavy impurities + phonons" in liquid media is explained in the Appendix by the example of vibrations occurring in the system "heavy ion + vortex line". Behavior of the phonon spectrum in the limit $c < c_*$ is shown in Fig. 2 taken from Ref. 24.

In the opposite limit $c > c_*$ an anti-crossing between the phonon part of the spectrum and the quasi-local eigenfrequency of the heavy impurity occurs. For this scenario, we cite here expressions for the asymptotics "*1*" and "*2*" in Fig. 3 (also taken from Ref. 24) (results similar to straight lines in Fig. 2)

$$\omega_1 = \frac{s_0 k}{\sqrt{1 + c(\Delta m/m)}}, \quad \omega_2 = s_0 k, \qquad (7)$$

where s_0 is the sound velocity in pure helium. Here qualitatively new effect compared to Fig. 2 is the development of the gap ω_0

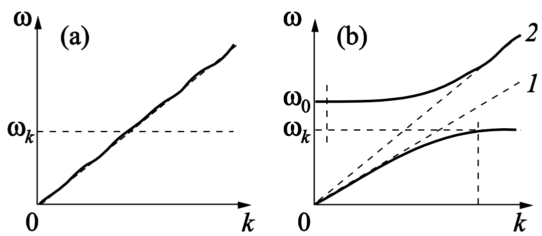

Fig. 3. Phonon spectrum in liquid helium containing heavy impurities in the limit $c > c_*$. Two oscillation branches with their interaction neglected (a). The same branches with their interaction taken into account. A quasi-gap ω_0 (8) separating two branches of the long wavelength vibrations is clearly seen (b).

$$\omega_0 = \omega_d \sqrt{1 + c(\Delta m / m_4)} \,. \qquad (8)$$

The difference between the phonon spectrum presented in Fig. 2 and that plotted in Fig. 3 can be directly observed experimentally.

4. Summary

Attempts to explain practically stationary iceberg-effect reported in Refs. 1–3 in the absence of luminescence from this domain led to the hypothesis (which needs further verifications) on the possibility of the existence of stable ion dipoles consisting of single-electron bubbles and snowballs. This hypothesis can be confirmed in several ways. Apart from the special behavior of the dielectric constant and appearance of specific osmotic pressure at the boundary of phase-separated ^{3}He–^{4}He solution "loaded" with finite density of ion dipoles, there is a peculiar transformation of the liquid helium phonon spectrum in the low-frequency range. The point is that any heavy impurity (including helium dipoles) should exhibit a nontrivial behavior of the associated mass $M(\omega)$ in the low-frequency range $M(\omega)_{\omega\to 0} \propto \omega^{-1/2}$ due to viscosity. This system provides a unique example of viscous dynamics and, which is less trivial, can be used to search for possible ways for statistical description of the gas of heavy impurities with rather exotic associated mass $M(\omega)$. In the proposed scenario of this statistics, heavy impurities are firmly bound to the solvent normal component, i.e., in the case of liquid helium, to phonons. Therefore, relative motion between the impurities and the helium normal component (which is responsible for the solvent viscosity) is cancelled thus eliminating any reasons for the effective mass singularity $M(\omega)_{\omega\to 0} \propto \omega^{-1/2}$). The price paid for this cancellation is rather serious: the phonon spectrum of liquid helium is substantially transformed and its perturbations which are proportional to the ion dipole density can be observed experimentally.

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Appendix

Among the known problems illustrating interaction of a heavy particle with the elastic medium possessing acoustic phonon spectrum, we choose to consider dynamics of a heavy ion localized at the vortex ring.

Equations of motion are

$$\rho\Gamma \frac{\partial u_y}{\partial t} = \rho \frac{\Gamma^2 \gamma}{4\pi} \frac{\partial^2 u_x}{\partial z^2} - F_x \delta(z) \,, \qquad (A.1)$$

$$-\rho\Gamma \frac{\partial u_x}{\partial t} = \rho \frac{\Gamma^2 \gamma}{4\pi} \frac{\partial^2 u_y}{\partial z^2} - F_y \delta(z) \,, \qquad (A.2)$$

$$M_i \ddot{\mathbf{r}}_i = \mathbf{F}, \;\; \mathbf{u}_{z=0} = \mathbf{r}, \;\; \gamma = \ln(8R/a) \,, \qquad (A.3)$$

where Γ is the vortex circulation, R is the ring radius, a is the vortex core radius, and M_i is the ion mass. The ring is assumed to have a finite radius to avoid discussions of typical vortex divergencies (in particular, that related to the parameter γ in (A.3)). That radius will be assumed to be sufficiently large to justify replacement of summation with integration over the phonon frequencies in Eq. (A.4).

A critically important point in the main text pinning heavy particles to the normal component of the superfluid liquid is introduced in dynamics (A.1)–(A.3) through the equation $\mathbf{u}_{z=0} = \mathbf{r}$ (A.3) firmly relating the ion position $\mathbf{r}$ to the string vibration amplitude $u(z,t)$ at the point $z = 0$ where the ion is localized at the vortex line.

In the limit $M_i \gg m_4$ (which is quite suitable for the ion dipole dynamics) Fourier transformed equations of motion result in the following dispersion law:

$$\frac{\pi\rho\Gamma}{M_i \omega^3} = \int \frac{dq}{(\Gamma^2\gamma^2 q^4 / 4\pi^2) - \omega^2} \,. \qquad (A.4)$$

Here the sum over discrete phonon frequencies of the ring is replaced with the integral over q.

Solution of this dispersion equation yields

$$\omega_i^2 \simeq \left(\frac{\rho\Gamma^2\gamma}{2\pi R M_i}\right)^{1/2} \left[1 - \frac{i}{4R}\left(\frac{\Gamma\gamma}{4\pi\omega}\right)^{1/2}\right]. \qquad (A.5)$$

For $M_i \simeq 100 m_4$ and $R \simeq 10^{-4}$ cm,

$$\mathrm{Re}\,\omega_i \simeq 10^9 \text{ s}^{-1}, \quad \mathrm{Im}\,\omega_i \simeq 10^{-2}\,\mathrm{Re}\,\omega_i \,. \qquad (A.6)$$

$\mathrm{Re}\,\omega_i$, $\mathrm{Im}\,\omega_i$ explain physical meaning of the eigenfrequency ω_d and its decay rate δ appearing in Eq. (6) of the main text.

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