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Multielectron Bubbles in Liquid Helium.

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Abstract. – We have investigated multielectron bubbles in liquid ^{4}He, which develop when the charged helium surface undergoes an electrohydrodynamic instability. Well-defined bubbles with charges ranging from 10^5 to 10^7 electrons are easily generated. Oscillations and fissioning of the bubbles are reported, and a new upper limit on their lifetime is set.

Electrons on the surface of liquid helium represent a nearly ideal 2-dimensional Coulomb system. They are localized about 100 Å above the surface in a potential well consisting of the image potential (plus holding potential) and a potential barrier of 1 eV due to the Pauli principle, that prevents them from entering the liquid helium [1]. The interparticle spacing of the electrons on helium is in general larger than 2000 Å. At a threshold density of about $2.4 \cdot 10^9$ electrons/cm^2 the surface becomes hydrodynamically unstable [2, 3]. When the instability develops, the surface deforms spontaneously: dimples with a size of the order of the capillary length (~ 1 mm) appear, which increase in depth, until charged macroscopic gas bubbles split off the dimple tip [4]. Under the influence of the applied electric holding field ($\geqslant 3$ kV/cm) the bubbles are dragged away from the surface and move towards the positive electrode immersed in the liquid, where they discharge and collapse (1).

Although they are somewhat difficult to observe, the multielectron bubbles appear as rather interesting objects. They are stabilized by the surface tension σ and the hydrostatic pressure p in the liquid counterbalanced by the repulsive Coulomb forces of the electrons. The equilibrium bubble radius R_C («Coulomb radius») is determined by minimizing the energy

$$E = 4\pi R^2 \sigma + \frac{4}{3}\pi R^3 p + \frac{Z^2 e^2}{2\varepsilon R}$$

(accounting for surface energy, external pressure and electrostatic energy, respectively; ε: dielectric constant of helium) [7, 8], which yields a range between 10 μm and 100 μm for

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(1) A related phenomenon has been observed by DAHM *et al.* [5] for positively charged droplets being pulled from the liquid into the vapour phase. Their description is quite analogous to that of the multielectron bubbles investigated here [6].

typical charges Z of 10^5 to 10^7 electrons with no external pressure applied. Due to their mutual Coulomb repulsion the electrons are confined to a thin «skin» close to the bubble surface [9], and thus constitute on a local scale a 2D Coulomb system like the electrons on the free liquid surface, however with a considerably higher density (10^{10} electrons/cm^2 and more). They therefore might provide an example where quantum corrections to the otherwise classical behaviour of the 2D electron system on helium should become important.

In this letter we report on some initial experiments with such bubbles, concentrating on properties like charge, velocity and stability against decay into smaller multielectron or also single electron bubbles (sometimes called «negative ions» [3]). Our results show, that it should be possible to trap the bubbles in adequate field configurations, so that they are accessible for more detailed investigations.

All our measurements were performed in ^{4}He above the λ-point. The electron source was a hot tungsten filament mounted inside a small glass tube for better localization of the site of bubble production. In order to charge the surface and reach the instability threshold the negative potential of the source was increased to several kV. The electron bubbles were then dragged to the anode consisting of a wire tip at zero potential about $(2 \div 3)$ mm below the liquid-helium surface. A schematic drawing of the set-up is shown in fig. 1.

We observed the bubbles optically by means of small-angle light scattering using a He-Ne laser. Pictures were recorded by a sensitive video camera and stored on tape. This enabled us to inspect the bubble tracks more closely in the «single-frame» mode of the video recorder, providing a time resolution of 40 ms.

Velocity of the bubbles. The bubble velocity v was determined by chopping the laser beam at a frequency f of 370 Hz. The bubble tracks then appear as a sequence of dashes at a distance v/f as illustrated in fig. 2. The bubbles were found to approach a constant velocity quickly after leaving the surface. The velocities are typically of the order of 10 cm/s (bulk temperature $T = 3.5$ K, electric field $E = 4500$ V/cm). This implies a time scale on the order

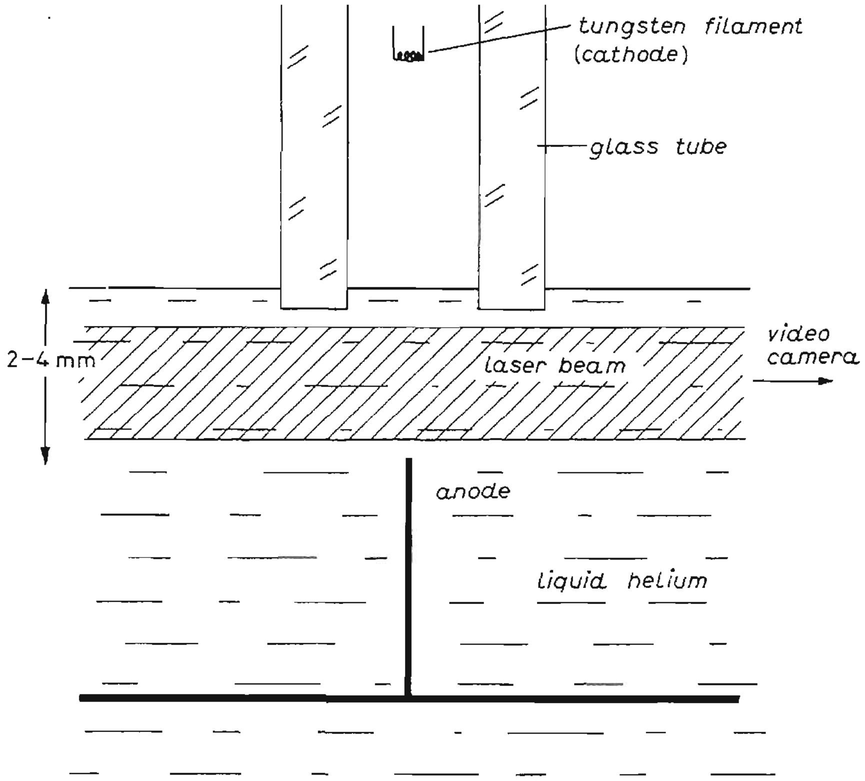


Fig. 1. – Schematic set-up of the experiment.

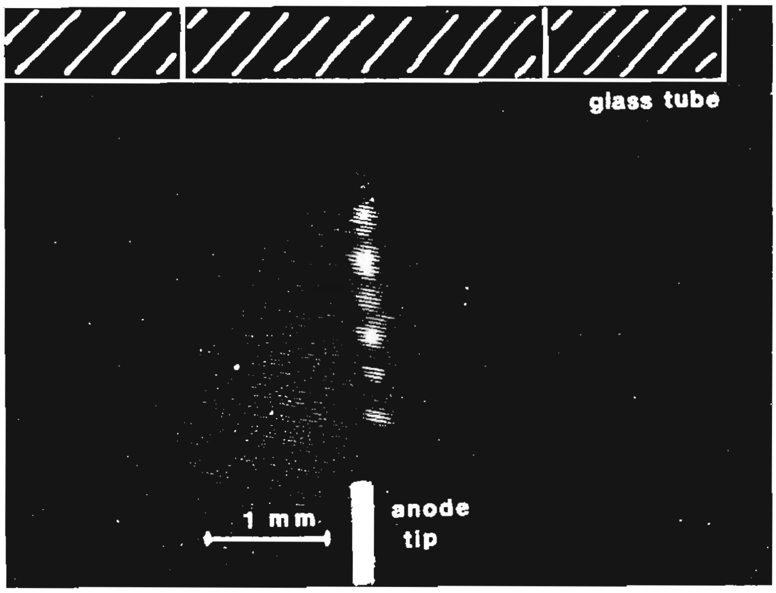


Fig. 2. – Track of a single multielectron bubble moving towards the anode tip. The irradiating laser is chopped at a frequency of 370 Hz to measure the velocity of the bubble. The size of the spots is not the size of the bubble but only a measure for the intensity of the scattered light.

of 10 ms for the observation of an individual bubble in a visual field of a few mm. The rather high velocities are well beyond the applicability of Stokes' law. Thus it was not possible to determine from these measurements directly the diameter or the charge of the bubbles. Therefore, additional experiments were required to characterize the bubbles.

Bubble charge. When an electron bubble approaches the anode tip, it produces a sharply peaked displacement current. Integration of this peak gives the bubble charge. Figure 3 shows several examples of the time dependence of the tip current, recorded with a digital storage oscilloscope. The sharp rise is due to the bubbles moving fast through the inhomogeneous part of the electric field close to the tip. After a bubble discharges at the tip, the current decays exponentially with a time constant given by the lead capacitance and the input resistance of the electrometer amplifier. Since this time is long compared to the rise time of the peaks, the bubble charge is directly proportional to the peak height.

We observed bubble charges Z between 10^5 and 10^7 electrons. Bubbles with $Z < 10^5$ electrons are not resolved by our apparatus. The value of Z depends on the potential gradient at the helium surface and on the surface tension, which is determined by the temperature of the liquid surface and hence by the vapour pressure (the temperature of the bulk liquid was always kept lower than the surface temperature in order to avoid boiling). Measurements at surface pressures of 1000 and 800 mbar show a very narrow distribution of bubble charges. At lower vapour pressures a transition to chaotic behaviour occurs and bubbles with almost random charge are produced (cf. fig. 3*a*)). One of the reasons may be smaller damping of surface perturbations at lower temperature.

Bubble oscillations. A somewhat closer inspection of the bubble tracks shows that frequently the light intensity along the path is modulated, even if the laser is not chopped (see fig. 4). We interpret these intensity variations as being due to oscillations of the bubbles, excited during the generation process or by turbulence in the liquid. Although in this case a time scale is not provided in the pictures, we can estimate the oscillation frequency to be of the order of 2 kHz, assuming for the bubble in fig. 4 a «typical» velocity of 10 cm/s. This is consistent with calculations by SALOMAA and WILLIAMS, who predict the

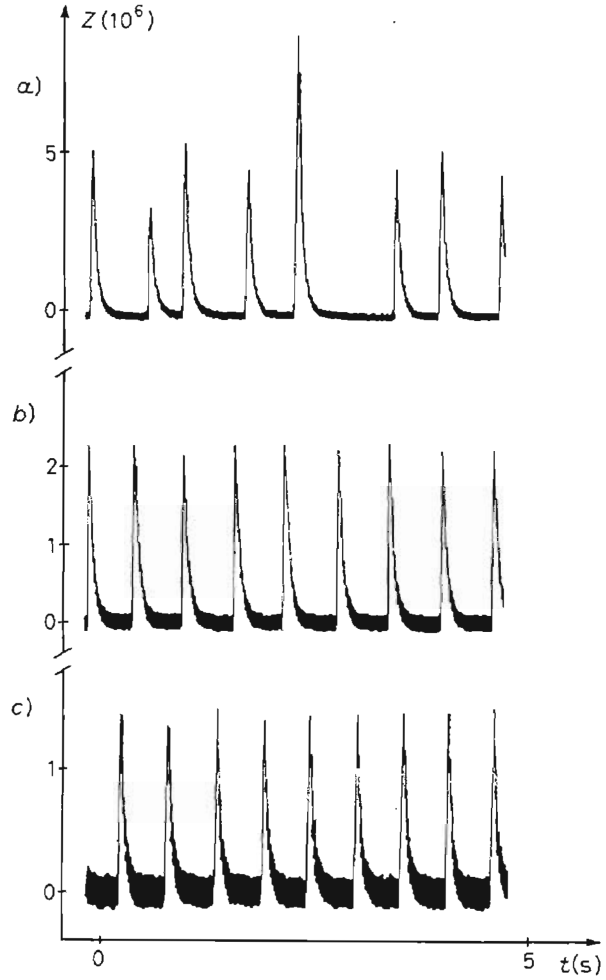


Fig. 3. – Displacement current at the anode recorded at vapour pressures: *a)* $p = 600\,\text{mbar}$, *b)* 800 mbar, and *c)* 1000 mbar. Each peak corresponds to one multielectron bubble; the peak height is proportional to the charge of the bubble (Z: number of electrons in a bubble).

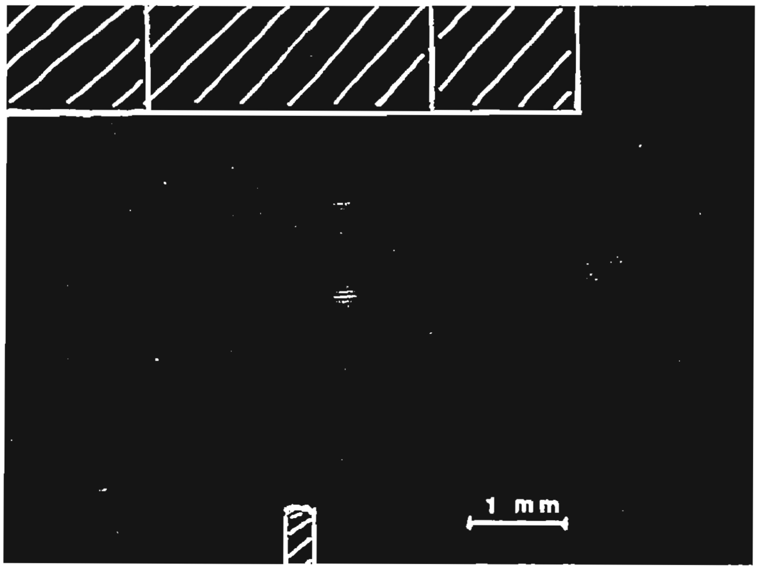


Fig. 4. – Bubble track with continuous laser irradiation; modulations along the track are due to oscillations of the bubble. In the upper part of the track a fairly regular oscillation with a frequency of around 2 kHz can be seen which then develops into a superposition of several oscillation modes, giving larger, irregular spacings.

fundamental frequency of a bubble with Z electrons as [7, 10]

$$\omega_0 = (96\sigma^2 \pi\varepsilon/\rho Z^2 e^2)^{1/2}$$

(σ: surface tension, ε: dielectric constant, ρ: density of liquid helium) giving $\nu \simeq 1.6$ kHz for $Z = 10^7$.

A nonspherical bubble has a nonuniform charge distribution — the local charge density will be higher in regions of higher local surface curvature. Since the time for establishing charge equilibrium is much shorter than typical oscillation periods, the bubbles are expected to become electrohydrodynamically unstable above a certain threshold of the angular-oscillation amplitude, when the Coulomb repulsion between the two regions of high curvature exceeds the surface tension forces. (SHIKIN denotes this as «deformation instability» [8]). We indeed observe fissioning of bubbles, as is demonstrated in fig. 5 (where the laser is chopped again): a fast bubble is seen moving downwards and then breaking up into two smaller and slower bubbles, which move towards the anode tip separately and on curved paths due to their mutual Coulomb repulsion.

For more detailed studies of the electron bubbles one might consider to capture and store them in an electric-quadrupole configuration similar to an r.f. trap used to store ions. Storage in such a device is only possible if the bubble motion is not critically damped, *i.e.* if the bubbles can oscillate back and forth in an electrical-potential minimum. In order to test this, we have modified the electrostatic potential by connecting the former anode tip to the cathode voltage. Figure 6*a*) shows the resulting equipotential lines, and fig. 6*b*) the track of a bubble in this potential. It is clearly visible that the motion contains an oscillatory component. Hence the condition for storage in an electric quadrupole cage appears to be fulfilled for the bubbles in liquid He, even for the normal phase above T_λ.

Bubble tracks like the one shown in fig. 6*b*) have also been used to obtain a new lower limit for the bubble lifetime τ. Since for this configuration the potential gradients in the centre of the cell were relatively small, the bubbles moved with low velocity and could be observed for more than 100 ms before they left the field of vision. Hence we can state that $\tau > 100$ ms, which implies that multielectron bubbles under our experimental conditions are fairly stable against decay by electron tunnelling into individual single electron bubble states (which have a diameter of about 30 Å and apparently cannot be observed optically). Yet at electron densities distinctly higher than the 10^{10} e/cm^2 of the present ex-

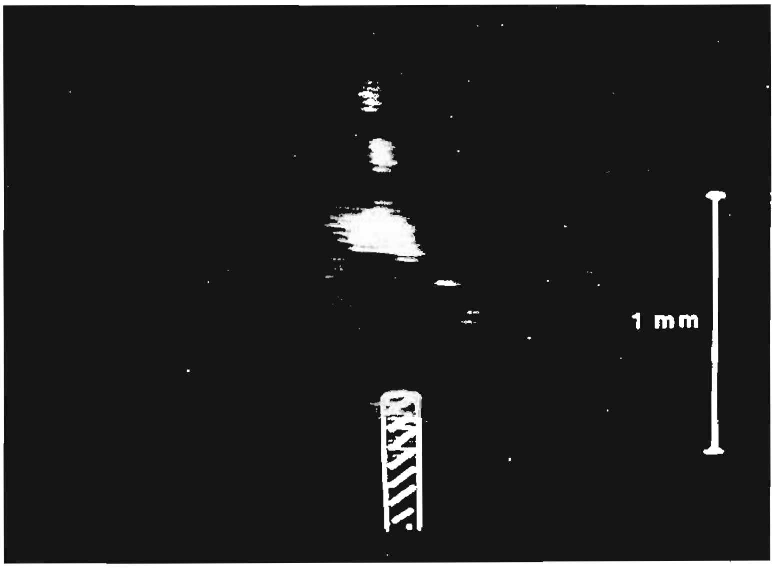
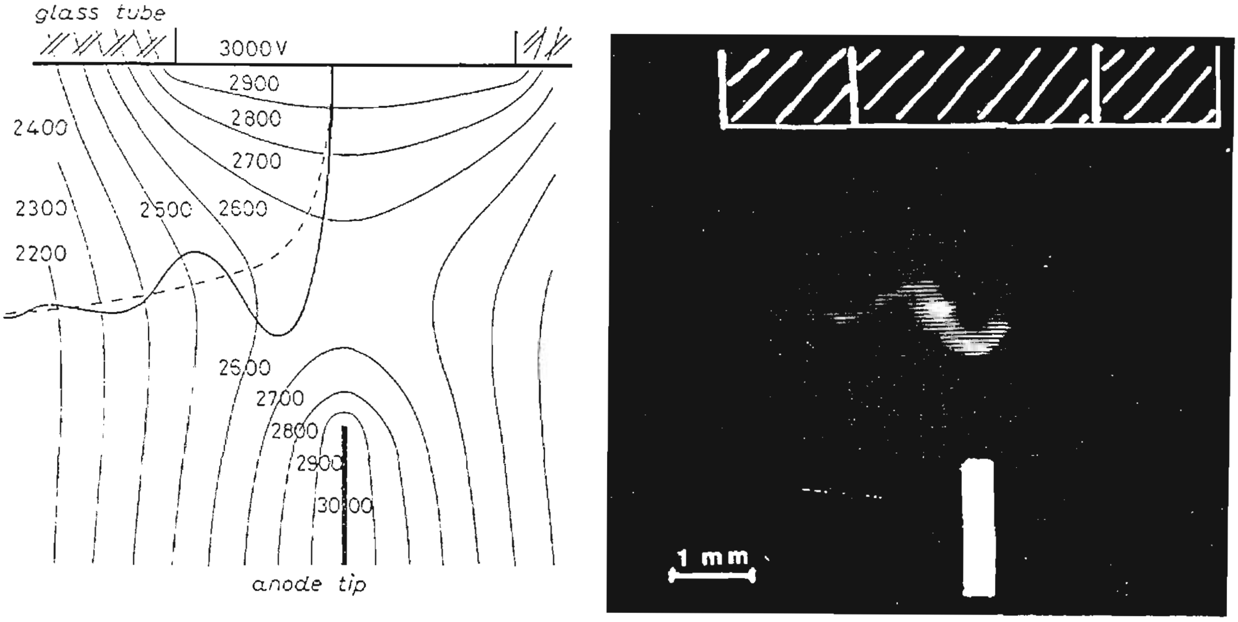


Fig. 5. – Fissioning of a bubble. The laser is chopped at $f = 370$ Hz.



a)

b)

Fig. 6. – Bubble track oscillating back and forth in a potential minimum; *a)* shows the potential configuration and the expected tracks for overdamped (---) and not critically damped motion (—— actual path), *b)* shows an actual track.

periment—achievable by pressurizing the helium—the electric field at the bubble surface can become so large that this process may become substantial [8, 11].

Since trapping of the bubbles in a quadrupole cage appears feasible, we are planning such an experiment to extend the measurements of τ to much longer times. Provided the lifetime is long enough, it should then be possible to study the eigenmodes of the bubbles, more thoroughly excited for example by an acoustic driving field. Of particular interest in this context is the influence of the bubble surface charge on these modes, which should reflect changes of the 2D electron system, *e.g.* Wigner crystallization. An attractive aspect of the multielectron bubbles then is, that the electron densities achievable there have not been accessible in other experiments so far.

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REFERENCES

[1] See, *e.g.*, WILLIAMS F. I. B., *Surf. Sci.*, **113** (1982) 371; MONARKHA YU. P. and SHIKIN V. B., *Sov. J. Low Temp. Phys.*, 8 (1982) 279 and references therein.

[2] GORKOV L. P. and CHERNIKOVA D. M., *JETP Lett.*, 18 (1973) 68.

[3] LEIDERER P., *Physica B*, **126** (1985) 92, and references cited therein.

[4] VOLODIN A. P., KHAIKIN M. S. and EDELMAN V. S., *JETP Lett.*, 26 (1977) 543; KHAIKIN M. S., *J. Phys. C*, 6 (1978) 1295.

[5] BOYLE F. P. and DAHM A. J., *J. Low Temp. Phys.*, 23 (1976) 477.

[6] WILLIAMS G. A. and SALOMAA M. M., *J. Low Temp. Phys.*, 57 (1984) 539.

[7] SALOMAA M. M. and WILLIAMS G. A., *Phys. Rev. Lett.*, 47 (1981) 1730.

[8] SHIKIN V. B., *JETP Lett.*, 27 (1978) 39.

[9] SALOMAA M. M. and WILLIAMS G. A., *Phys. Scr.*, T4 (1983) 204.

[10] HANNAHS S. T., WILLIAMS G. A. and SALOMAA M. M., in *Proceedings of the XVII International Conference on Low Temperature Physics*, edited by U. ECKERN, A. SCHMID, W. WEBER and H. WÜHL (North Holland, Amsterdam) 1984, p. 297.

[11] ARTEM'EV A. A., KHRAPAK A. G. and YAKUBOV I. T., *Fiz. Nizk. Temp.*, 11 (1985) 1011.