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Extraction of charges from beneath the surface of liquid hydrogen

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Experimental results are presented which confirm the existence of electron bubbles in liquid hydrogen. The effective values of the barrier preventing the electron to escape from the liquid into the gas and the electron bubble radius are estimated to be 350 ± 70 K and 10 Å, respectively. The experiments show that β-radiation gives rise to both the electron bubbles and the negatively and positively charged clusters (snowballs) in liquid hydrogen. Quasi-periodic oscillations of the charged surface and extraction of ~ 10^8 charges during a pulse have been observed at electric fields higher than some critical value ~ 10^3 V/cm.

Introduction

In previous discussions [1–3] of the properties of injected charges in liquid hydrogen it has been assumed that the negative charge is an electron localized in a spherical cavity with a radius of about 10 Å, while the positive charge is a cluster (snowball) consisting of an H_2^+ molecular ion surrounded by a layer of solid hydrogen. However, metastable atomic H^- ions can be created along with electrons and H_2^+ ions in liquid hydrogen subjected to irradiation [4]. Ions of any impurities dissolved in hydrogen can be also produced. The structure of injected charges in liquid hydrogen has not been so far determined experimentally.

In this report we present results of our experiments on charge extraction through the surface of liquid hydrogen at electric fields up to 10^4 V/cm, which confirm the existence of electron bubbles and of positive or negative charge clusters. Some of the results reported here have been already published [5].

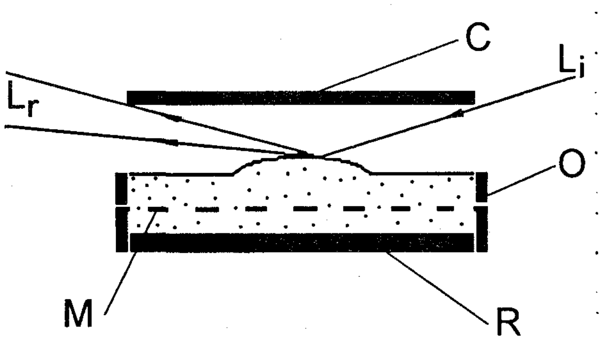

Fig. 1. Scheme of the experiment to observe oscillations of the charged liquid hydrogen surface in the triode cell. *R* — radioactive source; *C* — collector; *M* — metal grid; *O* — guard ring; L_i — incident laser beam; L_r — reflected laser beam.

Experimental procedure

In experiments we studied the field and temperature dependence of the current flowing across the interface from the liquid into the vapor. We also measured the total charge arriving at the collector as the charged surface begins to oscillate. The measurements were performed in a diode or triode placed into an optical container partially filled with liquid hydrogen. As the source of charges we used a radioactive plate immersed in the liquid. This plate ionized an adjacent layer of liquid about 10 μm thick. The distance from the liquid surface to the collector was close to 2 mm. A metal guard ring 3 mm high, insulated from the source, prevented charges from escaping from the surface of the liquid to the container wall. The radioactive plate and the guard ring formed a cup. Hydrogen was condensed into this cup. A change in the curvature of the charged surface was estimated from a broadening of the laser beam reflected from the surface. A scheme of this experiment is shown in Fig. 1.

Results and discussion

Low electric fields (below 1 000 V/cm)

The current of positive charges through the liquid-vapor interface in a diode is negligible (less than $1 \cdot 10^{-15}$ A) at applied electric fields up to 1 000 V/cm. The negative charges can pass across the surface and the value of the current depends on temperature, voltage at the source, and voltage at the guard ring.

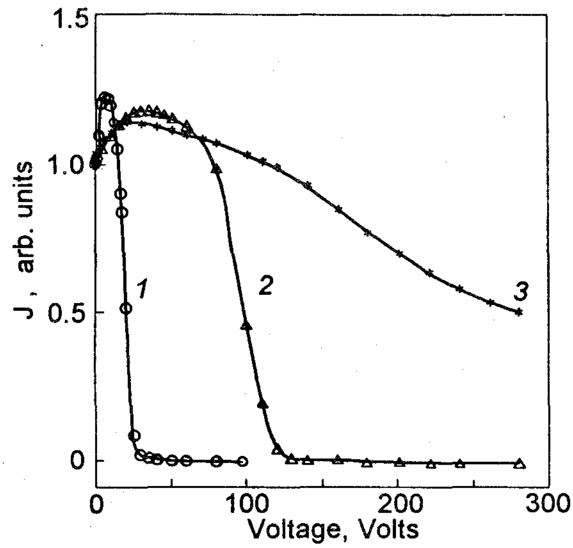

Fig. 2. The current of negative charges in the diode versus the voltage at the guard ring, V. Curves *1* and *2* are for the source-collector voltages of $U = 20$ and 100 V, respectively, at $T = 16$ K; curve *3* is for a diode completely filled with the liquid at voltage $U = 100$ V and temperature $T = 16.5$ K.

Figure 2 shows the current of negative charges versus the guard ring voltage V at a constant value of the potential difference between the source and the collector, U. Curves *1* and *2* were recorded at 16 K at two different values of U. As the potential of the guard ring V approaches that of the source U, the current is observed to drop sharply to zero. In a diode filled completely with liquid (curve *3* in Fig. 2; measurement temperature 16.5 K) the current goes through a maximum as a function of the voltage on the guard ring V and then falls off slowly at $V > U$. This result

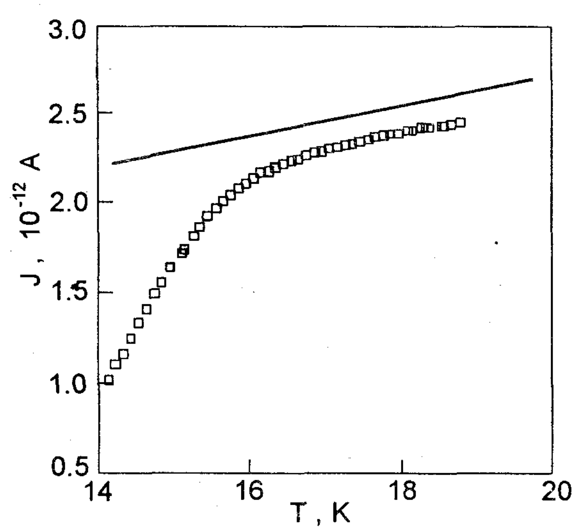

Fig. 3. Temperature dependence of the current of negative charges in the diode: curve *1* is the current through the surface at source-collector voltage $U = 10$ V; curve *2* is for a diode completely filled with the liquid at $U = 10$ V and the guard ring voltage of 5 V.

can be attributed to a partial cancellation of the vertical component of the applied electric field in the liquid.

In the case of the partially filled diode (curves *1* and *2*) the sharp decrease in the current across the surface is apparently due to a cancellation of the holding field by the negative charges whose structure differs from that of electron bubbles, i.e., by clusters that accumulate under the surface and cannot penetrate into the gas phase.

Curve *1* in Fig. 3 is the temperature dependence of the current of negative charges, $J(T)$, through the surface for $U = 10$ V and $V = 5$ V. Curve *2* describes the temperature dependence of the collector current $J_f(T)$ when the diode is completely filled with liquid, for the same voltages as for curve *1*. By comparing curves *1* and *2* we can conclude that at temperatures below 17 K the magnitude and the temperature dependence of the current $J(T)$ in partially filled diode are determined by the conditions for the charges to cross the surface. Above 17 K, the surface has a negligible effect: $J(T)$ is close to $J_f(T)$.

The charged layer in our cell is formed below the surface as a result of the external field. The sign of the charges in this layer is determined by the polarity of the applied voltage. The charges can approach the surface due to thermal fluctuations. It is natural to suggest that, as in helium [6,7], an electron can tunnel through the interface over distances from the surface on the order of the bubble radius. The current of negative charges across the surface is proportional to the concentration of electron bubbles and to the probability for an electron to cross the interface. The probability for heavy positive or negative clusters to tunnel through the boundary is exponentially small as compared to that for free electrons: hence, positive or negative charged clusters accumulate below the surface.

So, observations of the crossing of the liquid-vapor interface by negative charges give evidence that electron bubbles exist in liquid hydrogen. A bubble is prevented from approaching the surface by electrical image forces. In order to determine the effective height of the barrier G that hinders the crossing of negative charges from the liquid into the vapor, we can represent the experimental $J^{-1}(T)$ dependence as a sum $J^{-1}(T) = J_1^{-1}(T) + J_2^{-1}(T)$, where $J_1(T)$ and $J_2(T)$ correspond to the high- and low- temperature asymptotic behavior of the current (respectively, curves *2* and *1* in Fig. 3). We assume that $J_1(T) = a J_f(T)$ and $J_2(T) = b \exp(-G/T)$, where a and b are numerical parameters. By fitting the analytic curve to the experimental data, we find a barrier height of $G = 350 \pm 70$ K.

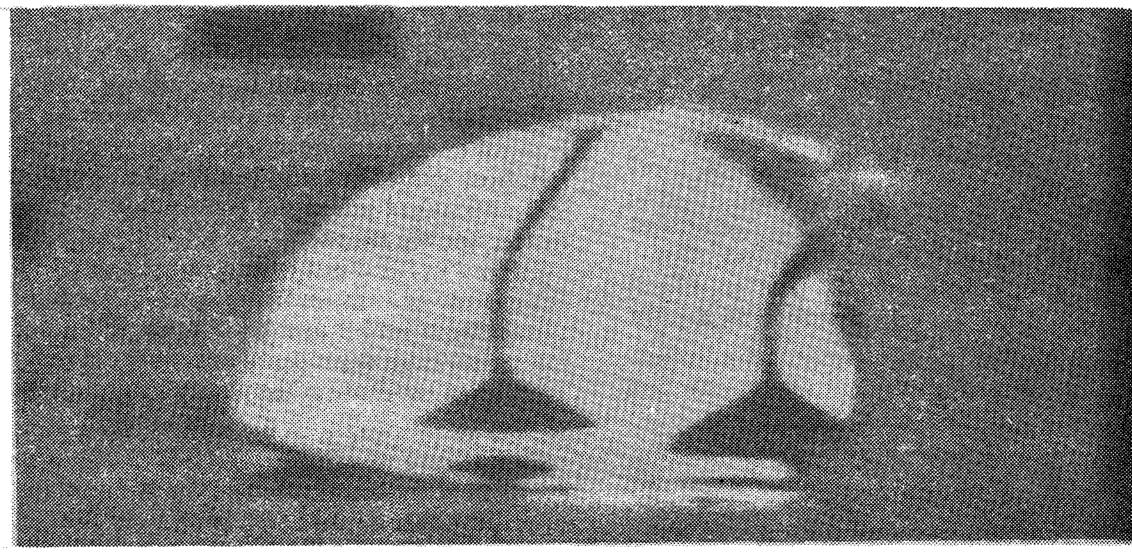

Fig. 4. Charged surface of liquid hydrogen in a double diode cell. The lifted charged surface is seen under the left diode containing the source only.

From the value of G we can estimate the effective distance R^* from the surface over which an electron tunnels from the liquid into the vapor. If we ignore the external electric field, we can write

$$k_B G = [e^2(\varepsilon_l - \varepsilon_g)]/[4R^*\varepsilon_l(\varepsilon_l - \varepsilon_g)] ,$$

where k_B is the Boltzmann constant; e is the electron charge; $\varepsilon_{l\,(g)}$ are the dielectric constants of the liquid (gas). Putting $G = 350 \pm 70$ K, we find $R^* = 10.6 \pm 2$ Å. From mobility measurements [1] the bubble radius is estimated to be ≈ 9 Å (the values of R, given in Ref. 1, should be multiplied by a factor of 1.25 to account for the effect of space charge on the velocity of the leading edge of charged particles). Theory [2] yields approximately the same value, $R \approx 10$ Å, i.e. bound electrons tunnel from the liquid into the vapor if the effective distance to the surface, R^*, is comparable with the radius of the bubble. This conclusion looks completely plausible.

Strong electric fields

As in the case of low applied voltages, the voltage dependence of the current of negative charges, $J(U, V)$, through the interface exhibits a drastic change at the value $V \approx U$. Note that when a voltage $V > U$ is step-wise applied to the guard ring, the current decreases not instantaneously but over quite a long period of time. This duration exceeds substantially the time needed for the charges to fly the distance between the source and the surface. This is because negative charged clusters, which are kept from running to the walls of the cell when driven by the voltage V applied to the guard ring, gradually accumulate under the liquid surface. The increasing (in time) curvature of the charged surface is also evidence that the density of accumulating charges increases.

In experiments with positive charges the current across the surface was below 10^{-15} A. The change in the surface curvature with applied voltage was, however, indicative of accumulation of positive charges under the surface of the liquid.

Visual inspection shows that as the voltage at the guard ring and at the source are increased (provided that the current across the interface remains negligible, $J < 10^{-15}$ A) the surface of the liquid in the diode (or triode) gets strongly curved: a hump forms in the center of the diode. In higher fields, nearly periodic oscillations of the liquid surface can be observed in the diode at 14.6 K at a constant voltage above a certain critical one, namely ≈ 500 V for positive charges and ≈ 800 V for negative charges in the diode with a guard ring diameter of 7.5 mm. The height of the hump grows in time, reaches a maximum and then the surface instantaneously relaxes to the initial planar state. Then the process is repeated.

Figure 4 shows the instant when the charged surface of the liquid in the left diode has reached a maximum value of about 0.5 mm. The temperature of observation is about 15 K. These are no charges below the surface inside the right diode. The linear dimension of the hump is approximately half the diameter of the collector (about 3 mm). In the triode, as the guard ring diameter was increased to 20 mm and the collector diameter to 19.5 mm, the critical voltage above

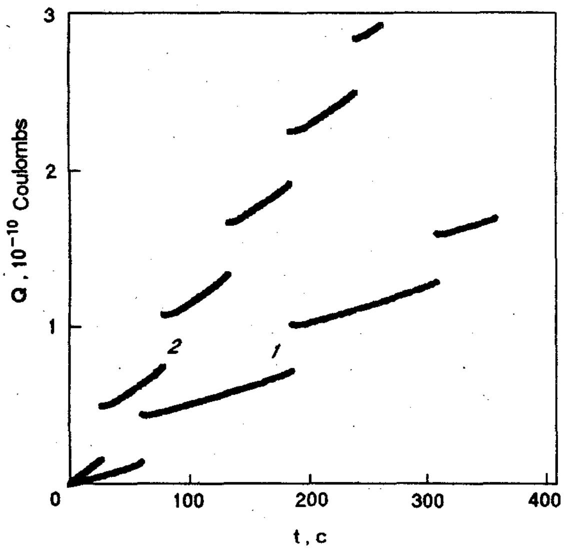

Fig. 5. Time dependence of the charge accumulated at the collector at high voltages. Positive charges are extracted from the surface at two different holding voltages. Curve *1* is for $U = 550$ V; curve *2* is for $U = 700$ V. Guard ring voltage 550 V.

which such oscillations were observed was ≈ 1 000 V for charges of either sign. It is noteworthy that the visible dimensions of the disturbed surface area did not change whatever in this case. The critical voltage decreased as the temperature of the liquid was increased.

Arrival of the charges at the collector connected with the electrometer with open input (charge accumulation regime) is recorded at the moment of surface relaxation, which means that the surface has discharged. We did not observe any break-away of individual droplets from the surface of the liquid. The surface relaxed in a jump-like manner to the initial state and the process was repeated. Figure 5 shows the time dependence of the total amount of positive charges, $Q(t)$, accumulated at the collector at two voltages of $U = 550$ V (curve *1*) and $U = 700$ V (curve *2*). The voltage $V = 550$ V on the guard ring is the same.

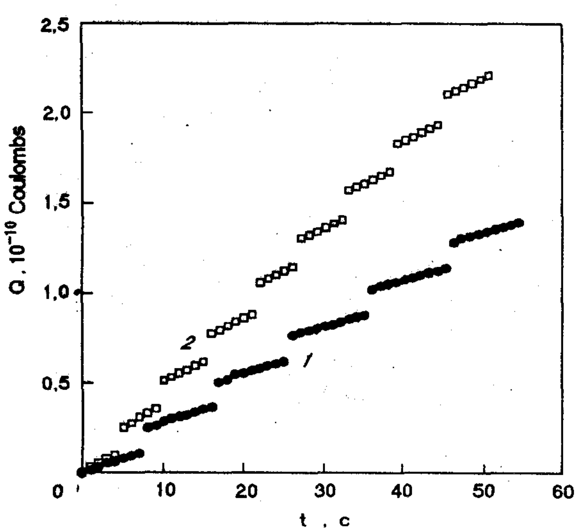

Fig. 6. Time dependence $Q(t)$ for the negative charges extracted through the surface at high voltages. Curve *1* was taken at holding voltage $U = 1\,050$ V and guard ring voltage $V = 1\,200$ V and temperature $T = 16$ K. Curve *2* : $U = 900$ V; $V = 950$ V; and $T = 18.2$ K

Figure 6 shows similar curves $Q(t)$ for negative charges: curve *1* was taken at $U = 1\,050$ V and $V = 1\,200$ V, the temperature was 18 K; curve *2* was taken at $U = 900$ V, $V = 950$ V, and $T = 16$ K. One can see that the period of oscillations of the charged surface decreases as either the applied voltage U, or the temperature of the liquid increases.

In Figs. 5 and 6 the height of the steps is approximately $3.4 \cdot 10^{-11}$ Coulomb for positive charges and $1.5 \cdot 10^{-11}$ Coulomb for negative ones at 16 K. The density of charges under the disturbed surface, as estimated from the height of the step on $Q(t)$, is approximately 10^8 cm^{-2}. This value is close to the estimated density of charges, n_s, in the layer that completely cancels the effect of the external field (found from the relation $E = 2\pi e n_s$, where E is the intensity of the critical electric field).

The critical electric field required for the charges to escape can be estimated [8] to be

$$E_c = 1.27 \cdot 10^4 \, (\Delta \rho \, \sigma)^{1/4} \text{ V/cm} ;$$

and the critical radius of the convex surface site is

$$r_c = (3\sigma / \Delta\rho g)^{1/2} \text{ cm} ,$$

where σ is the surface tension; $\Delta\rho$ is the density difference between liquid and vapor; g is the gravitational potential. The calculated value of E_c in hydrogen is 8 500 V/cm at 15 K. This value exceeds the experimental value of 5 000 V/cm for positive and negative charges. At the same time the calculated critical radius of the convex surface site r_c amounts to 3.5 mm, a value twice as large as that observed in experiment (close to the capillary length).

The surface in our cell oscillates as a whole because, given the cell dimensions, only one disturbed site can be formed in electric fields insignificantly exceeding the critical value (several disturbed sites can be formed if only the distance between them is much longer than r_c). Once the charges have escaped from the liquid-vapor interface, the time required for a new portion of charges to accumulate (critical for the given temperature), i.e. the oscillation period, is determined by the source capacity and the velocity of charges in the liquid. Therefore, the period of oscillations decreases as the value of the applied field is increased or the temperature is raised.

The triode geometry enables estimation of the residence time of charges below the surface of the liquid. As seen from Fig. 4, the accumulation of charges under the surface at constant U and V gives rise to a convex surface site. The curvature radius of this site depends on the magnitude of the holding field. If the source is grounded and the voltage between the grid and the collector is kept constant, then the rate of relaxation of the curved surface can yield the velocity at which the charges escape. In our experiments the change in the surface curvature was registered from broadening of a laser beam reflected from the surface of the liquid. It turned out that over one hour the surface curvature and, hence, the density of both positive and negative charges are constant within 2%. The current flowing across the surface of the liquid to the collector was less than 10^{-15} A with the source grounded. So, one may conclude that the life time of positive and negative charged clusters which accumulate under the surface at $T = 14-20$ K exceeds 1 hour. On the other hand, we noted above that the electrons localized in bubbles are capable of crossing the liquid-vapor boundary. The life time of a bubble under the surface, as estimated from a change in the current across the liquid surface in the triode at 14 K and low voltages $U < V$, was less than 10 s. One therefore may conclude that negative charges of two types (bubbles and clusters) exist in the liquid. Bubbles, containing an electron inside, govern the magnitude of the current flowing across the liquid-vapor interface. Clusters consist of a negative charged H^- ion [4] or an impurity ion. These clusters are surrounded by a layer of solidified molecular hydrogen. Such clusters accumulate below the surface like positive charges do.

Note that in Ref. 1 concerned with measurements of the mobility of charges in liquid hydrogen we suggested that there should be negative charges of two types. From mobility measurements it follows that at low temperatures the radius of negative charges is 4 Å. This value is close to the estimated radius [4] of a negative charged cluster containing an atomic ion H^-.

Conclusion

It has been shown that both negative charged electron bubbles and ion clusters which, like positive charges, are capable of accumulating below the surface can form in liquid hydrogen under the action of β-radiation. The electron bubble radius is about 10 Å and that of a negative charged cluster is about 4 Å. It has been demonstrated that a layer of positive or negative charges can be accumulated below the liquid hydrogen surface and kept there for a sufficiently long time. The density of charges under the surface reaches 10^8 cm^{-2}. Quasi-periodic oscillations of the charged surface have been observed in fields in excess of certain critical values.

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