Crossing of a liquid-vapor boundary in hydrogen by negative charges

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(Submitted 27 July 1994; resubmitted 18 August 1994)
Pis'ma Zh. Eksp. Teor. Fiz. **60**, No. 6, 452–456 (25 September 1994)

The current flowing across a surface of liquid hydrogen has been measured as a function of the temperature and the polarity and magnitude of the applied voltage. Positive charges do not penetrate into the vapor from the liquid to any significant extent. Below 17 K, the temperature dependence of the current of negative charges can be described by an exponential function with an activation energy of 350 ± 70 K. The experimental results imply that two types of negative charges, differing in structure, exist in the liquid hydrogen: electron bubbles and clusters, which, like positively charged ion clusters, accumulate below the surface of the liquid.

Measurements of the mobility of charged particles (charges) which form in hydrogen subjected to ionizing radiation have shown[1] that negative charges of two types, differing in mobility, can be observed in liquid hydrogen below 20 K. In previous discussions of the properties of injected charges in liquid hydrogen, it has been assumed[2,3] 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 consisting of an H_2^+ molecular ion surrounded by a layer of solid hydrogen. However, metastable atomic H^- ions can form[4] along with electrons and H_2^+ ions in liquid hydrogen subjected to irradiation; ions of impurities dissolved in the hydrogen can also form. Which of these charges were actually observed in the experiments of Refs. 1 and 3 was not known at the outset.

Experiments on the passage of charges across a surface of liquid helium into vapor have yielded graphic confirmation of the existence of electron bubbles in helium. It has been found[5] that the current of negative charges resulting from a tunneling of bound electrons from the liquid into the vapor falls off exponentially with decreasing temperature: $J(T) \sim \exp(-G/T)$. According to the estimates of Ref. 6, the characteristic activation energy in ^{4}He is $G \approx 40$ K. Positively charged clusters do not cross the interface to any significant extent.

One might hope that a study of the crossing of a liquid-hydrogen surface by charges would also yield information on the structure of the charges moving in the interior.

The idea underlying the present experiments is quite simple. A charged particle below the surface of a liquid is acted upon by a image force which is directed into the liquid:

$$F = (e/2z)^2(\epsilon_l - \epsilon_g)/[\epsilon_l(\epsilon_l + \epsilon_g)],$$

where e is the charge of the particle, z is the distance from the surface, and $\epsilon_{l,g}$ are the dielectric constants of the liquid and the gas. The external electric field E cancels the effect of the image force and holds the charges a certain distance z_0 below the surface. This distance is determined by the relation $F(z_0)=eE$. If there is a source of charges in the liquid, a charged layer forms 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. Charges may move toward the surface as a result of thermal fluctuations. In the model of Refs. 7 and 8 it is assumed that an electron can tunnel across the interface into the vapor as a bound electron bubble approaches the surface. The current of negative charges across the surface is proportional to the concentration of electron bubbles and to the probability that an electron will cross the interface. The probability that heavy positive clusters will tunnel through the boundary is exponentially small in comparison with that for electrons, so positive charges accumulate below the surface.

EXPERIMENTAL PROCEDURE

In the experiments we studied the field dependence and the temperature dependence of the currents flowing across an interface from a liquid into a vapor. The measurements were carried in a diode partially filled with liquid. The source of charges was a radioactive plate immersed in the liquid. This plate ionized a layer of liquid about 10 μm thick around itself. The distance from the source to the collector was 5 mm. A metal guard ring 3 mm high, insulated from the source, prevented charges from escaping from the surface of the liquid to the wall of the hermetically sealed container holding the diode. The radioactive plate and the guard ring formed a cup with an inside diameter of 20 mm; hydrogen was condensed in this cup. The height of the liquid layer above the source was 0.5–3 mm. As a control measure, some measurements were repeated in a triode as in Refs. 5 and 6. The results found in the diode and triode measurements were in agreement.

EXPERIMENTAL RESULTS

As in the case of helium, the current of positive charges across a liquid-hydrogen surface in a diode is negligible (less than 1×10^{-15} A) at applied electric fields up to 1000 V/cm. The magnitude of the negative-charge current depends on the temperature, on the voltage on the source, and also on the voltage on the guard ring.

Figure 1 shows the current of negative charges versus the voltage on the guard ring, 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 different values of the applied voltage U. As the potential of the guard ring approaches that of the source (the collector is essentially grounded), the current is observed to drop sharply to zero. In a diode filled completely with liquid (curve 3 in Fig. 1; measurement temperature of 16.5 K), the current goes through a maximum as a function of the voltage on the guard ring and then falls off slowly at $V>U$. This result can be attributed to a partial cancellation of the vertical component of the applied electric field in the liquid.

The sharp decrease in the current across the surface in the case of the partially filled diode (curves 1 and 2) is apparently due to a cancellation of the applied field by negative charges whose structure differs from that of electron bubbles: clusters which accumulate under the surface and which cannot penetrate into the gas phase. Two facts point in this direction. First, the surface is curved in the absence of a current, and when an alternating

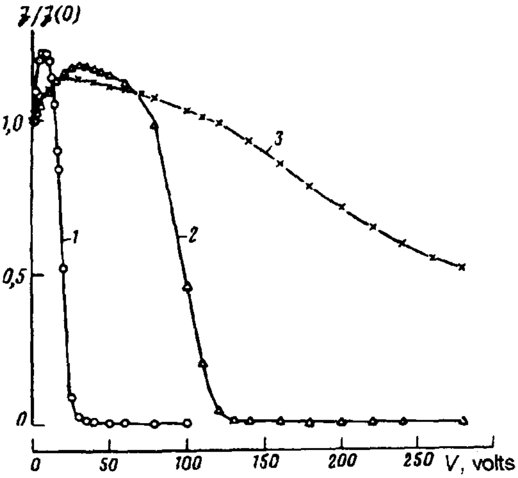

FIG. 1. Current of negative charges in the diode versus the voltage on the guard ring, V [actually shown here is the ratio $J(V)/J(0)$]. 1, 2—Current crossing the surface for source–collector voltages $U=20$ and 100 V, respectively, at $T=16$ K; 3—for a diode completely filled with liquid, at a voltage $U=100$ V and a temperature $T=16.5$ K.

voltage is applied to the guard ring we can observe standing waves at the surface of the liquid (a curvature of the surface and the onset of standing waves were also observed in the experiments with positive charges). Second, when a step voltage $V<U$ is applied to the guard ring, the relaxation time of the negative current is less than 10 s, while at $V>U$ the current crossing the surface and flowing to the collector reaches a new steady-state value over a time on the order of 10^3 s. In other words, over this time, charge accumulates under the surface and cancels the applied electric field.

Curve 1 in Fig. 2 shows the temperature dependence of the current of negative charges, $J(T)$, for the case of a potential difference $U=10$ V between the source and the collector, with a voltage $V=5$ V on the guard ring (this is the steady-state current). Corresponding curves of the temperature dependence were observed at $U>V$ over the voltage range $U=1$ –30 V. Curve 2 was measured as the temperature of the liquid was

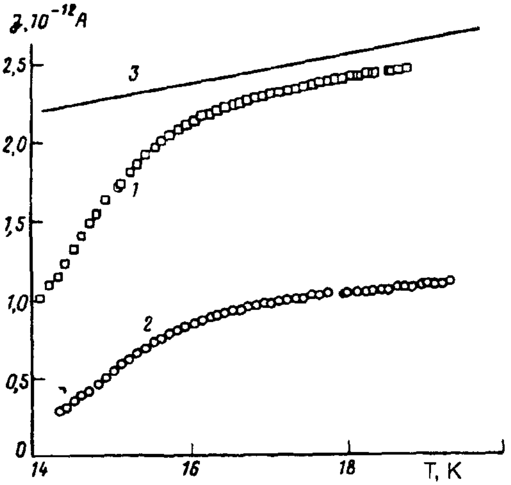

FIG. 2. Temperature dependence of the current of negative charges in the diode. 1, 2—Current crossing the surface at source–collector voltages $U=10$ and 3 V, respectively, at a guard-ring voltage $V=5$ V; 3—for a diode completely filled with liquid at $U=10$ and $V=5$ V.

rapidly raised, under conditions such that the blocking surface layer had not yet had time to form. For this curve we have $U=3$ V, $V=5$ V, a heating time of 6 min, and a current relaxation time of more than 1 h. Curve 3 shows the temperature dependence of the collector current in a diode completely filled with liquid, for the same voltages as for curve 1.

In the diode completely filled with liquid, the current in the liquid, J_f, depends only weakly on the temperature over the temperature interval 14–20 K (Fig. 2). In a diode partially filled with liquid, the current across the surface, J, increases rapidly as the temperature is raised from 14 to 17 K. Above 17 K, the temperature dependence $J(T)$ approaches the $J_f(T)$ curve. It can be concluded from a comparison of curves 1, 2, and 3 that at temperatures below 17 K the magnitude and temperature dependence of $J(T)$ are determined by the conditions for the crossing of the surface by the charges. Above 17 K, the surface has a negligible effect.

DISCUSSION

Observations of the crossing of a liquid–vapor interface by negative charges thus confirm that electron bubbles exist in liquid hydrogen. It is natural to suggest that, as in helium,[7,8] an electron can tunnel through the interface over distances from the surface on the order of the bubble radius. A bubble is prevented from approaching the surface by electrical image forces. In order to work from the $J(T)$ curves to determine the effective height of the barrier opposing the crossing of negative charges from the liquid into the vapor, we can describe the experimental $J^{-1}(T)$ dependence by the sum $J^{-1}=J_1^{-1}(T)+J_2^{-1}(T)$, where $J_1(T)$ and $J_2(T)$ correspond to the high- and low-temperature asymptotic behavior of the current on curves 1 and 2 in Fig. 2. It is being assumed here that the temperature dependence $J_1(T)$ is governed by the bulk component, i.e., that we have $J_1=aJ_f(T)$, where a is a numerical parameter. It is also assumed that an electron bubble is prevented from approaching the surface by the energy barrier set up by electrical image forces. In other words, we are assuming $J_2=b\exp(-G/T)$, where b is a numerical parameter, and G is the barrier height. Fitting analytic curves to the experimental data, we find a height $G=350\pm70$ K. The error in the determination of G is determined by the width of the temperature interval.

We can work from the value of G to 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

$$kG=[e^2(\epsilon_l-\epsilon_g)]/[4R^*\epsilon_l(\epsilon_l+\epsilon_g)],$$

where k is the Boltzmann constant. Inserting the value $G=350\pm70$ K, we find $R^*=10.6\pm2$ Å . It follows from mobility measurements[1] that the radius of a bubble is ~9 Å (the values of r given in the paper should be multiplied by a factor of 1.25 to reflect the effect of space charge on the velocity at which the front of charged particles moves in the diode). Theoretical work in Ref. 2 yields approximately the same value, $r\approx10$ Å ; i.e., bound electrons tunnel out of the liquid into the vapor if the effective distance to the surface, R^*, is comparable to the radius of the bubble. This conclusion looks completely plausible.

In summary, it can be concluded from measurements of the passage of a current across a liquid–vapor interface that irradiation gives rise to both electron bubbles and negatively and positively charged ion clusters in liquid hydrogen. The probability for the clusters to cross the interface is small in comparison with that for electrons localized in the liquid.

We wish to thank V. N. Khlopinskiĭ for assistance in the preparation for the experiments and in the experiments themselves.

This study had partial support from the Soros International Science Foundation.

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Translated by D. Parsons