ANOMALOUS ELECTRON MOBILITY AND COMPLEX NEGATIVE ION FORMATION IN LOW-TEMPERATURE HELIUM VAPOR*

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We report in this note experimental evidence, obtained from mobility data, that slow electrons injected into helium vapor do not travel between collisions as free electrons but form rather heavy complexes with helium atoms. This conclusion is supported by the observation in the saturated vapor of a mobility four orders of magnitude lower than that expected from kinetic theory assuming free electrons. The mobility rises very rapidly as the gas pressure is lowered somewhat below the saturated vapor pressure in a manner interpretable in terms of a thermodynamic instability of the complex but not consistent with the possibility that we are observing the transport of an impurity negative ion.

Measurement of the mobility of ions[1] and electrons[2] in liquid helium has suggested interpretation in terms of a model[3] in which a charged particle surrounds itself with a massive electrostatically polarized complex of helium atoms and behaves as a particle of rather high effective mass. The stability of such complexes suggests the possibility that similar structures might also be stable in helium gas at low temperatures. We have investigated this possibility by measuring the mobility of electrons injected into helium gas at temperatures near 4°K and pressures up to the saturated vapor pressure. The experiment was performed using apparatus similar to that briefly described in connection with electron mobility measurements in liquid He II.[2] A pulsed xenon flash lamp illuminates a semitransparent cesium-antimony photocathode in a glass phototube which is immersed in liquid helium. Current pulses from the central portion of the anode are amplified and displayed on an oscilloscope. Pulse shapes observed in the gas are somewhat anomalous and are not well analyzed at present. A representative pulse is illustrated in Fig. 1(d). Figure 1 also shows for comparison theoretical pulse shapes and a pulse observed when the tube is filled with liquid helium. The time of transit has been assumed equal to the time required for the pulse to reach its peak value. Plots of reciprocal transit time so defined versus applied voltage are accurately linear for low voltages. Because of the uncertain interpretation of the pulse shape and experimental errors in the other measured quantities, the mobility data should be regarded as no more accurate than ± 25%.

Several isotherms (mobility versus pressure at constant T) are plotted in Fig. 2. The free electron low-field mobility, calculated from kinetic theory or extrapolated from room-temperature data[4] assuming $\mu \propto p^{-1}T^{1/2}$, has values of the order of 10^3 cm²/volt sec in this pressure and temperature range.

We wish to stress the following points about the data and to offer our interpretation in light of the phenomenological theory sketched below:

1. The mobility at the saturated vapor pressure is lower than the free electron value by a factor of order 10^4. We have measured the mobility of electrons injected into helium gas at 300°K and 77°K with the same apparatus and obtain values in excellent agreement with those observed by others.[4]

2. The mobility is a relatively slowly varying function of pressure (although the variation is more rapid than p^{-1}) for pressures between the saturated vapor pressure, p_s, and a critical

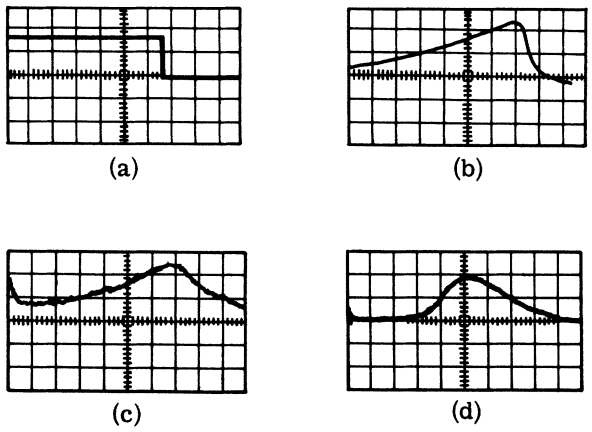

FIG. 1. Theoretical and experimental pulse shapes. (a) Theoretical for uniform electron field in drift space. (b) Theoretical for actual field (determined in electrolytic tank). (c) Experimental for electrons injected into liquid helium. (d) Experimental for electrons injected into helium gas. The theoretical pulse shapes neglect initial transient effects and space charge.

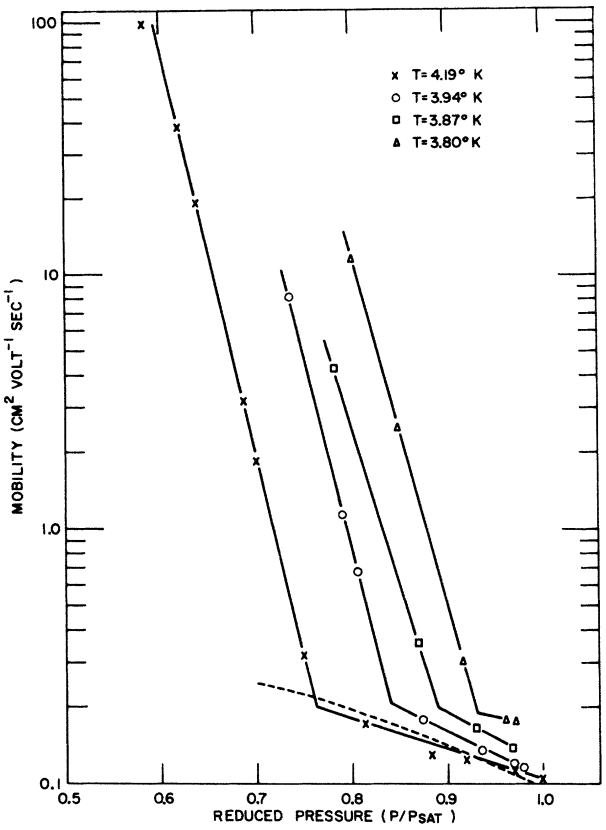

FIG. 2. Electron mobility in helium gas plotted against the ratio of the gas pressure to the saturated vapor pressure. The pressure at which the slope changed is p_c. The dashed line is calculated from the Langevin theory for $T = 4.2°$K. The solid lines have no theoretical significance.

pressure, p_c. We believe that in this pressure range a complex of definite size is stable, and that the mobility variation results principally from a decrease in the equilibrium radius of the complex as p is lowered below p_s.

3. For pressures below p_c the mobility rises very rapidly as the pressure is lowered, presumably tending toward the free electron value. For reasons of resolving time and signal-to-noise ratio we have not yet obtained data at pressures below those shown. We believe that for $p < p_c$ the probability that an electron be found attached to a complex decreases rapidly, but continuously, from a value near unity to a value near zero leading to the observed dependence of μ upon p. The mobilities observed at the lowest pressures are too high to be explained by formation of negative impurity ions, although some impurities might be expected to be evolved from the photocathode during the light flash.

In the high-pressure range ($p > p_c$) a good account of the data can be given by assuming that a small droplet of liquid helium forms on each electron. As is familiar from the theory of the cloud chamber,[5] small charged droplets can be stable when the gas pressure p (at large distances from the droplet) is below the saturated vapor pressure p_s. In the present case the relationship between these pressures and the radius R of the droplet can be written as

$$\ln(p/p_s) = (nkT)^{-1}(2\gamma/R + \partial\gamma/\partial R) - \alpha e^2/2R^4kT, \quad (1)$$

where n and γ are the atomic density and surface tension of the drop and α is the polarizability of a helium atom. An approximate value for $R(p, T)$ can be derived if we insert for γ the surface tension observed in the bulk liquid and neglect $\partial\gamma/\partial R$. Equation (1) then predicts an equilibrium radius decreasing from 12.6 A at 4.2°K and $p/p_s = 1$ to 9.6 A at $p/p_s = 0.75$ where the apparent onset of instability occurs. R also decreases along the vapor pressure curve as T is decreased and should become large as the critical temperature is approached. The mobility of such a complex can now be calculated from the Langevin theory[6] assuming hard-sphere (isotropic) collisions at radius R plus electrostatic (polarization) interaction at larger distances. The mobility, calculated by this method, agrees well with the experimental data in the range $p > p_c$. A calculated curve for $T = 4.2°$K is plotted in Fig. 2. The agreement between theory and experiment is probably better than could be expected considering the crudeness of the surface tension ansatz,[7] and the fact that we have considered the nearly saturated vapor as an ideal gas. (The correct density was used in the Langevin theory, however.) Nonetheless, the agreement with the data is impressive in view of the fact that the theory contains no adjustable parameters.

We can interpret the rapidly varying portion of the data for $p < p_c$ in terms of the following kinetic argument. The observed transit time and mobility are determined by the time-average drift velocity of an electron. In steady state there will be a finite probability, P_e, that an electron be free, and a probability, $P_d = 1 - P_e$, that it be attached to a droplet. The measured mobility, μ, will then be connected with the free electron mobility, μ_e, and the droplet mobility, μ_d, by

the relation

$$\mu = \mu_e P_e + \mu_d P_d. \quad (2)$$

In the high-pressure range $P_d \cong 1$, but as the pressure is decreased P_e increases. In fact in steady state we would have $P_d/P_e = w\tau$, where w is the probability per unit time per free electron for droplet formation and τ the mean lifetime of a droplet. This interpretation then requires that as p (and R) decrease, $w\tau$ must decrease rapidly.

The droplet interpretation of the data discussed above implies the existence of a rather stable state in which an electron is bound to a complex of helium atoms approximately 10 A in radius. It is in fact difficult to see how this is possible. The high zero-point energy of a confined electron, with its associated pressure, has suggested to a number of workers that the electron would tend to expel helium atoms from the region where the electron wave function is greatest. In the present case the "droplet" would then become a "bubble" of inner radius b and outer radius R. The value of the inner radius b is determined by a pressure balance equation and can be estimated either by assuming a surface tension at $r = b$ equal to the bulk value or by the method of Kuper.[8] Both methods predict b values larger than 10 A. Thus we are not at present able to present a consistent model for the wave function of an electron bound to such a complex of helium atoms. We are hopeful that additional study will shed light on the structure of the negative ions observed in both gaseous and liquid helium.

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DETECTION OF THIRD SOUND IN LIQUID HELIUM FILMS

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The present note describes experiments leading to the detection of "third sound" in thin films of liquid helium II. As is well known, Tisza[1] and Landau[2] predicted that for bulk liquid helium II, besides ordinary (or first) sound, in which the normal and superfluid components oscillate in phase, giving a pressure wave, there should also exist another type of wave motion, second sound, in which the normal and superfluid components oscillate out of phase, giving a temperature wave. More recently one of us[3] has suggested that, in the liquid helium film, there might be yet another type of wave motion, termed third sound, in which the superfluid component oscillates but the normal component remains locked to the wall. Detailed analysis suggested that third sound should appear as an oscillation in the thickness of the film. It is therefore somewhat similar to classical waves on shallow water, but the superfluidity of liquid helium is essential to its existence since such a wave would be rapidly attenuated in a thin film of an ordinary viscous liquid. Apart from being an oscillation in thickness rather than pressure or temperature, the most striking characteristic of third sound is that the predicted velocity is much lower than