Evidence for Roton Pair Creation in Superfluid ^{4}He†

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We have measured the drift velocity $\bar{v}$ of negative ions through superfluid ^{4}He for temperatures $T < 0.45$ K, under a pressure of 25 bar, for electric fields $20 < E < 2.6 \times 10^4$ V cm^{-1}. From the observed form of $\bar{v}(E)$, we deduce that roton emission from an ion traveling faster than the Landau critical velocity is governed by hitherto unrecognized selection rules whereby the rotons can be created only in pairs.

The discovery by Phillips and McClintock[1] that negative ions can be drawn through pressurized superfluid ^{4}He at drift velocities $\bar{v}$ considerably in excess of the Landau critical velocity v_L for roton creation,[2] without creating charged vortex rings, opened the way for a detailed investigation of supercritical energy dissipation. Allum, McClintock, and Phillips[3] showed subsequently that the velocity data were well described, for electric fields in the range $20 < E < 2000$ V cm^{-1}, by a relation of the form

$$\bar{v} = v_L + AE^{1/3}, \tag{1}$$

where A is a constant, a typical result being shown in Fig. 1: The discrepancy (2%) between the intercept and the value of v_L deduced from roton parameters is less than the possible systematic error (± 3%) in $\bar{v}$ arising from the uncertainty in our measurements of the length of the drift space.

Bowley and Sheard[4] demonstrated, on the basis of golden-rule calculations, that this result was inconsistent with energy dissipation through the expected single-roton emission process [for which $(\bar{v} - v_L) \propto E^{2/3}$ should be found], but that it was precisely the behavior to be anticipated if rotons were being emitted from the moving ion in pairs. Furthermore, they were able to show that, if the pair-creation hypothesis was correct, upward deviations from (1) should be observed in higher electric fields for which $\bar{v} > v_L + \Delta v_0$. Here, Δv_0 is the velocity decrement $(= 2\hbar k_0/m_i)$ for an ion of mass m_i when a pair of rotons each of momentum $\hbar k_0$ is emitted parallel to the electric field. Assuming $m_i = 72 m_4$ (see below), where m_4 is the ^{4}He atomic mass, progressively larger deviations from (1) would therefore be anticipated if $\bar{v}$ was increased above 55 m sec^{-1}. The purpose of this Letter is to report the result of experiments designed to test Bowley and Sheard's prediction and thus the veracity of their two-roton-emission hypothesis.

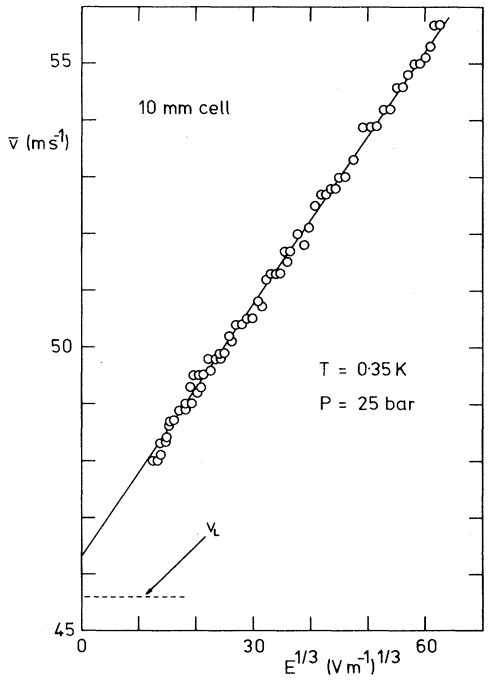

FIG. 1. The relationship between the ionic drift velocity $\bar{v}$ and the electric field E is well described by (1) when $\bar{v} < 55$ m sec^{-1}. The dashed line indicates the value of the Landau critical velocity v_L deduced from roton parameters.

The apparatus and experimental techniques were substantially as described previously,[1,5] except that the length d of the space in which the ionic time of flight was to be measured was made considerably shorter in order to raise the maximum electric field attainable using the same circuitry and power supplies. In addition to our original 10-mm cell, we have employed two new cells with drift lengths nominally of 2.5 and 1 mm,[6] enabling us to measure $\bar{v}$ in electric fields of up to 26 kV cm^{-1}. Because d could be measured only to an accuracy of a few percent, a scaling procedure has been applied to data obtained from the two shorter cells: For the 2.5- and 1-mm cells, d has accordingly been adjusted by +4% and +5%, respectively, from the directly measured values (which were themselves uncertain to about ±8% and ±15%) in order to obtain agreement with the (more accurate) 10-mm data in the region of overlap below 3 kV cm^{-1}.

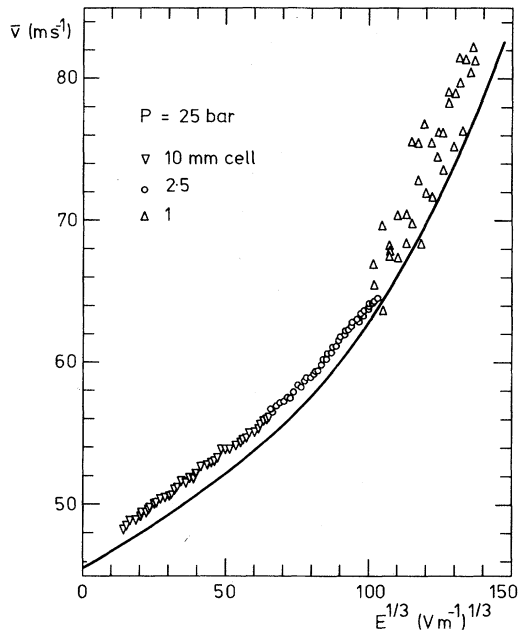

FIG. 2. Experimental results from three different cells (points) compared with the two-roton emission theory (full curve). In the interests of clarity many data points, including all those in the regions of overlap, have had to be omitted.

All the velocity measurements were made at $P = 25$ bar, $T < 0.45$ K, for electric fields such that $\bar{v}$ was independent of T. Results are shown in Fig. 2. Measurements made using the 1-mm cell show a large scatter, partly because of the relatively long rise time (3 μsec) of the measuring circuit, partly because of its modified design,[6] and partly because the signals became progressively smaller as the electric field was raised beyond a few kV cm^{-1}. The latter effect, which appeared also to depend in a complicated manner on T and P, is believed to be a consequence of vortex-ring creation, and will be discussed in more detail elsewhere. It is immediately evident from Fig. 2 that in high electric fields there are significant deviations from (1), that these are in the expected direction, and, furthermore, that they start approximately at the predicted value of $\bar{v}$.

To compare the data quantitatively with the two-roton-emission theory we start from the golden-rule equation for the emission rate,

$$R(v) = \frac{2\pi}{\hbar} \sum_{\vec{k}} \sum_{\vec{q}} \frac{|V_{\vec{k},\vec{q}}|^2}{\Omega^2} \delta\left(\epsilon_{\vec{k}} + \epsilon_{\vec{q}} - \hbar(\vec{k}+\vec{q})\cdot\vec{v} + \frac{\hbar^2(\vec{k}+\vec{q})^2}{2m_i}\right), \tag{2}$$

where a pair of rotons of momenta $\hbar\vec{k}$, $\hbar\vec{q}$, and energies $\epsilon_{\vec{k}}$, $\epsilon_{\vec{q}}$ is emitted by an ion traveling initially at an instantaneous velocity v; $V_{\vec{k},\vec{q}}$ is the unknown matrix element; and the Landau dynamics are, of course, implicit in the δ function which ensures conservation of energy. Assuming that, for the region of interest in $\vec{k}$, $\vec{q}$, and $\vec{v}$, the matrix element is constant ($= V_{k_0,k_0}$), and ignoring certain negligible higher order terms, (2) yields

$$R(v) = \frac{k_0^4 |V_{k_0,k_0}|^2 m_r (v-v')^2}{[2\pi^2\hbar^3(v-\hbar k_0/m_i)^2]}, \tag{3}$$

where $v' = v_L + \Delta v_0/2$ is the minimum value of v for which roton emission is possible, and k_0 and m_r are roton parameters.[7] We have found $\bar{v}$ from[8]

$$\bar{v} = \frac{\int f(v) v\, dv}{\int f(v)\, dv}, \tag{4}$$

having first obtained the distribution function $f(v)$ of ionic velocities by solving the appropriate Boltzmann equation which, for equilibrium, is

$$\frac{df(v)}{dt} = \frac{\partial f(v)}{\partial v}\frac{dv}{dt} - R(v)f(v) + R(u)f(u)\frac{du}{dv} = 0. \tag{5}$$

Here, the first term (with $dv/dt = eE/m_i$) gives the rate of change of $f(v)$ caused by the electric

field, the middle term gives the rate of change of $f(v)$ caused by roton emission by ions traveling initially at velocity v, and the last term refers to ions traveling initially at velocity u but which, after roton emission, travel at velocity v. The average difference between u and v, for the large velocities we are considering, is not well approximated by Δv_0, as was assumed previously,[4,5] because rotons can be emitted at relatively large angles to the electric field: We find that the average velocity decrement parallel to the field, for an ion traveling with initial velocity u, is to a good approximation

$$u - v = \Delta v_0 [1 - 2(u - v')/3u]. \quad (6)$$

With the single exception of the matrix element V_{k_0,k_0}, the values of all the constants appearing in the above equations are known from independent measurements. We have therefore been able without ambiguity to solve (3)–(6) numerically for $\bar{v}(E)$, treating V_{k_0,k_0} as an adjustable parameter in fitting the theory to the experimental data. Choosing the value $V_{k_0,k_0} = 3.7 \times 10^{-52}$ J m^3, which gives the correct slope for small E, we obtain the full curve of Fig. 2. In carrying out this procedure we have assumed an ionic effective mass which is entirely hydrodynamic ($= \frac{2}{3}\pi\rho r_i^3$) in nature; an ionic radius of $r_i = 1.1$ nm, based on Ostermeier's analysis[9] of earlier experimental data; and a liquid density at 25 bar of 0.172 g cm^{-3}. That the experimental points lie systematically about 2% above the theoretical curve gives no cause for concern, being merely an indication of the small discrepancy (see above), within experimental error, between the value of v_L which may be deduced from our data and the accepted value derived from roton parameters. It must strongly be emphasized that the remarkably good agreement between theory and experiment shown in Fig. 2, extending over more than three orders of magnitude in E, has been obtained *with the use of only one adjustable parameter.*

A number of observations should be made, however, about the range of applicability of our model. (a) We assumed, in the absence of other information, that the matrix element in (2) is constant: This is probably a reasonable approximation for $E < 10$ kV cm^{-1}, where the total range in velocity is less than a third of its absolute value. (b) We have ignored departures from parabolicity of $\epsilon(k)$, which can be shown to become important when $E > 10$ kV cm^{-1}, but which will have an insignificant effect at lower fields. (c) For our highest electric fields, the spatial separation of successively emitted rotons could be as small as 2 Å (if they were all created at the same position in relation to the ion) in which case they might perhaps interfere with each other. (d) We have ignored the possibility of multiroton (and other types of multiple-excitation) emission processes. For our highest electric fields, the simultaneous emission of up to sixteen rotons is energetically allowed, and would have the effect of increasing the drag on the ion. The fact that the high-field data do *not* deviate to the right of the theoretical curve of Fig. 2 may thus be taken as an indication that such processes do not occur to any significant extent. (e) In sufficiently high electric fields, the energy of the ion will become poorly defined owing to its short lifetime between emission events. However, by replacing the delta function in (2) with a Lorentzian, itself containing $R(v)$, and solving self-consistently, we have shown that, although this phenomenon also would tend to increase the drag on the ion, it can safely be ignored for $E < 50$ kV cm^{-1}.

We conclude that the excellent agreement between theory and experiment for $E < 10$ kV cm^{-1} constitutes an impressive vindication of the two-roton-emission hypothesis, but that the agreement at the highest fields may, in view of (a)–(d), perhaps be fortuitous. No feature of our present, admittedly rather vague, physical picture of the roton has enabled us, even in retrospect, to account for the existence of selection rules which apparently favor roton creation in pairs but forbid their emission singly or in other combinations. This intriguing and quite unexpected result is worthy of further study.

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M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975), Vol. 1, p. 165.

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[6]The 1-mm cell incorporated a 2-mm-long medium-field (1 kV cm^{-1}) space on each side of the 1-mm high-field region. This arrangement facilitated temporal separation of the wanted signal from the gate-switching transients, and prevented high-field "leakage" into the gate or collector spaces.

[7]R. J. Donnelly, Phys. Lett. 39A, 221 (1972).

[8]The high-field forms of $\bar{v}(E)$ developed in Refs. 4 and 5 were based on a number of approximations which become poor for the higher values of E used in the present experiments. In particular, because of the different shape of $f(v)$ at very large E, compared to that in small E, we have found it essential to compute $\bar{v}$ by solving a Boltzmann equation, rather than by using the simpler approach via the concept of a relaxation time.

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Surface Structure and Bonding of Acetylene to the Platinum (111) Surface

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Analysis of low-energy-electron-diffraction beam intensities for a 2×2 chemisorbed layer of acetylene on the Pt(111) surface shows that the molecules are adsorbed in either a threefold position or a twofold bridge position at a distance of 1.95±0.10 Å above the topmost plane of platinum atoms.

In recent years the technique of low-energy-electron-diffraction (LEED) beam-intensity–voltage (I-V) analysis (surface crystallography) has been used primarily to determine bonding geometries for (unreconstructed) clean metal surfaces and simple overlayer systems of chemisorbed atoms. Here we wish to report the first such investigation of an ordered overlayer of molecules on a surface, the hydrocarbon acetylene (C_2H_2) associatively chemisorbed on the (111) surface of platinum.[1] We find that the method can distinguish between competing model geometries of the metal-hydrocarbon system and that the optimum bonding arrangement gives calculated I-V profiles which are in consistently good agreement with experiment for several incident beam angles and a large number of fractional and integral-order beams.

The exposure of a clean Pt(111) surface to C_2H_2 gas at 300 K under ultrahigh vacuum conditions followed by gentle heating to 400 K for 1 h gives rise to a 2×2 LEED diffraction pattern with sharp fractional-order spots indicative of long-range order in the overlayer.[2] The LEED beam intensities from the C_2H_2 overlayer were measured photographically at several incident beam angles for energies 10–200 eV. The experimental apparatus, single-crystal platinum sample, cleaning procedure, and the photographic technique have been previouly described.[3] The gas was introduced into the vacuum chamber via a stainless-steel needle directed at the crystal surface. Although gas exposures have not been accurately determined, it was found that at 300 K a well-ordered C_2H_2 overlayer was obtained only at low exposures (~1 L = 10^{-6} Torr sec), in agreement with earlier work.[4] The adsorption and ordering characteristics of C_2H_2 were independent of the electron beam. However, the fractional-order beams lost intensity under electron-beam exposure at a rate approximately proportional to the incident beam current. The electron-beam exposure was therefore limited so that the maximum loss in fractional-order beam intensity was about 5% during the period of photographic data collection (2 min).

The surface structure analysis was carried out by comparing calculated I-V profiles from models of the Pt-C_2H_2 geometry to the experimental results. The model geometries considered were the likely ones in which the C-C axis of the molecule is parallel to the surface plane with the molecule oriented in various symmetric ways with respect to the substrate atoms (Fig. 1). The observed three-fold symmetry of the diffraction pattern could arise from three 120°-rotated domains of either 2×2 or 2×1 packing of C_2H_2 on the substrate, and both types of translational symmetry were considered in the anaysis. The calculations employed a multiple-scattering theory using a beam representation and the layer-doubling method.[5] The Pt geometry and scatter-