

<sup>8</sup>M. S. Feld, thesis, Massachusetts Institute of Technology, 1967 (unpublished); M. S. Feld and A. Javan, "Laser-Induced Line-Narrowing Effects in Coupled Doppler-Broadened Transitions," to be published.

<sup>9</sup>There are several relevant misprints in Eq. (33): In line (33e),  $E_1 E_2$  should read  $E_1 E_2^2$ ; line (33d) should read  $+(|\mu_{12}\mu_{31}|^2/\gamma_1)(N_3-N_1)^{1/2}E_1 E_2^2[\gamma-i(\omega_B-\nu_B)]^{-1}$ .

<sup>10</sup>H. R. Schlossberg and A. Javan, Phys. Rev. Letters **17**, 1242 (1966); G. W. Flynn, M. S. Feld, and B. J. Feldman, Bull. Am. Phys. Soc. **12**, 669 (1967).

<sup>11</sup>For explanation of (i), see Ref. 7 and M. S. Feld, B. J. Feldman, and A. Javan, Bull. Am. Phys. Soc. **12**, 669 (1967), and "Frequency Shifts of the Fine Structure Oscillations of the 8446-Å Atomic Oxygen Laser," to be published; (ii) is merely due to the presence of the weak fine-structure component with gain, which overlaps the high-frequency wing of the 2-0 transition ( $^3P_2-^3S_1$ ). The 1-0 transition ( $^3P_1-^3S_1$ ), however, is completely symmetrical and free of overlap.

<sup>12</sup>For oxygen linewidths, see W. L. Wiese, M. W. Smith, and B. M. Glennon, Atomic Transition Probabilities, U. S. National Bureau of Standards National Standard Reference Data Series-4 (U. S. Government Printing Office, Washington, D. C., 1966), Vol. 1.

<sup>13</sup>W. R. Bennett, Jr., W. L. Faust, R. A. McFarlane, and C. K. N. Patel, Phys. Rev. Letters **8**, 470 (1962).

<sup>14</sup>A study of the intensity of the 1-0 laser oscillations as a function of cavity length would be of interest.

<sup>15</sup>In this case the time-dependent wave function  $\Psi$  is

obtained from a three-level Schrödinger equation to which radiative decay terms have been added. For

$$\Psi = \sum_{j=0}^4 c_j \exp(-iW_j t) u_j,$$

with  $u_j$  the eigenfunction of level  $j$  of energy  $\hbar W_j$ , the coupled equations are  $\dot{c}_i + \sum_j (a_{ij} - \frac{1}{2}\gamma_j \delta_{ij}) c_j$ , in which  $a_{ij} = -[\mu_{ij} E(t)/\hbar] \exp[i(W_i - W_j)t]$ , and  $E(t)$  is the sum of the two traveling-wave fields as seen in the atoms' rest frame.

<sup>16</sup>As an example, for an atom in level 0 at initial time  $t_0$ ,  $|c_j(t=t_0, t_0)| = \delta_{j0}$  and the 0-1 transition rate at a later time  $t$  is  $\gamma_1 |c_1(t, t_0)|^2$  (see preceding footnote). Thus, the total stimulated power emitted by background atoms in level 0 is  $\hbar \Omega_1' n_0 \gamma_0 \gamma_1 \int_{-\infty}^t |c_1(t, t_0)|^2 dt_0$ .

<sup>17</sup>In extending Eq. (2) to the spontaneous-emission case, the population of level 1,  $n_1$ , should be set equal to 0 and the energy density of the weak probe field,  $(E_1^0)^2/8\pi$ , should be replaced by  $(\hbar \Omega_1'^3/8\pi^3 c^3) d\Omega_1' dS$ , where frequency interval  $d\Omega_1' \ll \gamma$  and  $dS$  is a small solid angle in the forward direction (+z axis);  $E_2$ , the laser field, remains in its classical monochromatic form. Similar remarks apply to Eq. (3); note, in particular, that  $G(\Omega_1)$  becomes the usual Doppler-broadened spectrum of the power emitted spontaneously into  $d\Omega_1 dS$  with given polarization.

<sup>18</sup>See Feld, Feldman, and Javan, Ref. 11.

## EVIDENCE FOR A NEW KIND OF ENERGETIC NEUTRAL EXCITATION IN SUPERFLUID HELIUM\*

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We present evidence showing that neutral excitations which are not photons are produced in superfluid liquid helium in the presence of a  $\text{Po}^{210}$  alpha-particle source. At low temperatures these excitations travel through the liquid with negligible scattering. They have sufficient energy to generate positive ions and electrons at the surface of the liquid or at a suitable metal plate immersed in the liquid.

Charged particles introduced into superfluid liquid helium form ion complexes which have been used to study scattering effects due to rotons, phonons, and  $\text{He}^3$  atoms.<sup>1,2</sup> Such charged particles have also been used to produce quantized vortex rings<sup>3</sup> and to investigate the properties of such rings as well as of vortex lines.<sup>4</sup> In this Letter we report evidence for the existence of energetic neutral excitations in He II and a study of the emergence of charged particles through the surface of the liquid at temperatures below 1°K.

Our experimental arrangement, illustrated in Fig. 1, is similar to that of Rayfield and Reif.<sup>3</sup> The ion source  $S$ , plated with  $\text{Po}^{210}$ , is immersed

in liquid helium; it emits alpha particles which generate ions within a 0.2-mm thick layer of liquid adjacent to  $S$ . Electric potentials in the apparatus are adjusted by means of metal grids. The current arriving at the collector  $C$  is measured with a vibrating-reed electrometer. All the electrodes are gold plated to avoid formation of insulating oxide layers. At the temperatures below 0.6°K used in most of our experiments, the helium vapor pressure is so low that atomic mean free paths in the vapor above the surface of the liquid are long compared to the dimensions of the apparatus; hence the vapor region is effectively a vacuum.

Consider the experimental situation of Fig.

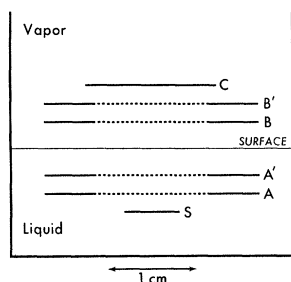


FIG. 1. Typical experimental arrangement showing the ion source  $S$ , the collector  $C$ , and several grids  $A$ ,  $A'$ ,  $B$ , and  $B'$ . The surface of the liquid lies between grids  $A'$  and  $B$ .

1 with an electric field applied normal to the surface of the liquid in the region between grids  $A'$  and  $B$ . It has previously been found that at temperatures above  $1^\circ\text{K}$ , charges can be extracted from the liquid into the vapor above its surface.<sup>5</sup> The resulting current arriving at the collector  $C$  is strongly temperature dependent and becomes vanishingly small as the temperature is decreased toward  $1^\circ\text{K}$ . We find below  $0.7^\circ\text{K}$ , however, a distinctly new temperature regime where current again appears in the vapor above the liquid and increases with decreasing temperature. This current can be shown to consist of two contributions of comparable magnitude. One of these, which we shall call the anomalous current, is completely independent of any electric fields existing within the liquid; it is the only current observed if these fields are such as to stop the passage of charged particles from the source  $S$  to the liquid surface. The remaining part of the current, operationally defined as the total current minus the anomalous current, will be called the normal current since it does respond to electric fields within the liquid.

The normal current consists of negative charges which emerge from the liquid surface with negligible energy (less than  $0.2\text{ eV}$  as measured by retarding potentials in the vapor). The charge-to-mass ratio of these charges, measured by a magnetic deflection experiment in the vapor, showed them to be electrons. The temperature dependence and magnitude of the normal current are similar to those of the negative vortex-ring current arriving at a collector  $C$  immersed in the liquid. Previous work<sup>3</sup> has shown that at temperatures below  $0.7^\circ\text{K}$ , charged vortex rings are formed in the liquid and travel appreciable distances without much energy loss. Hence we are led to the following conclusion:

In the temperature region below  $0.7^\circ\text{K}$ , charged vortex rings arrive at the surface of the liquid; in the case of negatively charged rings, the electrons carried by these rings are able to escape through the liquid surface and thus give rise to the observed normal current.

The nature of the anomalous current was the subject of primary interest in our investigation. This anomalous current, emerging from the surface of the liquid, can be either positive or negative, depending on the direction and magnitude of the electric field applied across the liquid surface. The fact that the anomalous current is not influenced by electric fields within the liquid suggests that this current is due to some kind of neutral excitations which travel through the liquid and then are converted to charged particles when they arrive at its surface. In order to substantiate this inference, the following experiments were performed to rule out any spurious effects. The anomalous current was observed to be an increasing function of the source strength and to disappear when the source is absent; thus the anomalous current clearly depends on the presence of the source  $S$ . The anomalous current was shown to persist even when the source  $S$  was completely enclosed in a can of metal solid except for one wall in the form of a grid; thus the anomalous current cannot be stray current originating at the source and traveling along unsuspected directions. By time-of-flight and magnetic-deflection experiments, the ions and electrons which constitute the anomalous current were shown to travel in the vapor from the surface to the collector; hence the anomalous current is not due to charges arriving at the collector after traveling in the helium film covering the container walls. Finally, the anomalous current can be detected not only by the collector above the liquid, but also (as will be described later) by a suitable metal plate immersed in the liquid. All these tests confirm the conclusion that the anomalous current is indeed due to some kind of neutral excitations which are created in the region near the source and travel through the liquid.

The neutral excitations responsible for the anomalous current can be generated by the  $\text{Po}^{210}$  source which injects into the liquid alpha particles with energies of several MeV. On the other hand, a tunnel cathode, immersed in the liquid<sup>6</sup> and injecting into it electrons of about  $1\text{-eV}$  energy, produces normal current but no

measurable anomalous current.

Detection of the neutral excitation was achieved, as already mentioned, by its conversion into positively and negatively charged particles at the surface of the liquid helium. The energy distribution of charged particles emerging from the liquid was ascertained by retarding-potential measurements in this region. (The kinetic energy of the positive particles was found to be negligible.) Determination of the charge-to-mass ratio of these charged particles in the vapor region was achieved by a time-of-flight experiment for the positively charged particle and by a magnetic-deflection experiment for the negatively charged particle. The masses of these particles, assuming them to be singly charged, identify them as  $\text{He}_2^+$  ions and electrons, respectively. The fact that the neutral excitation can produce charged particles at the liquid surface indicates that the neutral must be endowed with appreciable energy, comparable with that required to ionize a He atom in vacuum (24.6 eV).

An alternative method for detecting the neutrals was obtained by allowing them to impinge upon, and thus eject electrons or ions from, a metal plate immersed in the liquid. To function as a detector, this plate was subjected to a large electric field by means of a potential applied to a closely spaced grid.<sup>7</sup> Even at the highest electric fields used, the efficiency of the metal plate as a detector of the neutral excitations was only about 1% that of the free liquid surface. Hence we used the liquid surface as a detector in most of our experiments. It is worth noting that essentially the same temperature dependence of the anomalous current, shown in Fig. 2, is obtained by measurements using either detection method.

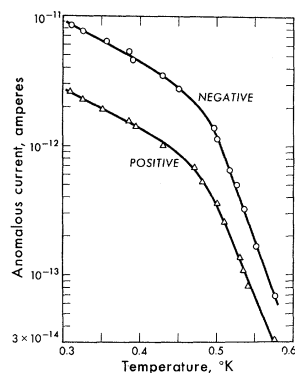


FIG. 2. Temperature dependence of the anomalous current emerging from the liquid surface.

At low temperatures ( $\leq 0.45^\circ\text{K}$ ), the neutral excitations travel in liquid helium along straight lines with negligible scattering. This conclusion was derived from an experiment in which "venetian-blind" baffles were placed between the source  $S$  and the liquid surface as shown in Fig. 3. Two collectors  $C_1$  and  $C_2$  were placed in the vapor above the liquid surface. The anomalous current collected by  $C_2$  was found to be much greater than that collected by  $C_1$ . This asymmetry caused by the directionality of the baffles indicates that, at these low temperatures, the neutrals travel in straight lines from the source  $S$  to the liquid surface about 1 cm away.<sup>8</sup>

The flux of neutral excitations is not attenuated after traversing a liquid thickness of the order of 1 cm. This is shown by noting that the anomalous current arriving at the collector in the vapor is unchanged if the level of the liquid above the source is increased while the solid angle subtended by the liquid surface at the source is kept constant. Thus, unless the neutral has a very large velocity in the liquid, its lifetime must be relatively long. In addition, the absence of attenuation of the neutrals by the liquid, combined with the temperature-insensitivity of the detection mechanisms, indicates that the temperature dependence shown in Fig. 2 is characteristic of the number of neutral excitations generated in the liquid near the source.

Finally, we have evidence that the neutral excitation is not a photon. If it were, it should have an energy of the order of 25 eV (the ionization potential of a He atom) in order to account for the production of ions and electrons at the liquid-helium surface. But a 900-Å thick, self-supporting aluminum film, interposed be-

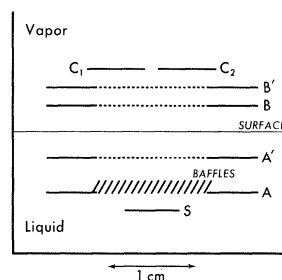


FIG. 3. Experimental arrangement designed to demonstrate straight-line propagation of the neutral excitations in the liquid. In the presence of the interposed baffles, the anomalous current collected by  $C_2$  is much greater than that collected by  $C_1$ .

tween the source  $S$  and the liquid surface, stops the anomalous current collected by  $C$  although it transmits all photons with energy between 16 and 55 eV.<sup>9</sup> (A crystal of LiF, interposed between the source and the liquid surface, also suppresses the anomalous current although it transmits all photons with energy between 0.1 and 12 eV.<sup>10</sup>)

The data which we have presented are not sufficient to lead to an unambiguous model for the neutral excitation observed in our experiments. We wish, however, to make the following speculative remarks. The fact that the neutral can eject ions and electrons when arriving at the liquid surface indicates that it must have an energy of the order of 25 eV; some kind of "exciton" or excited helium atom ( $\text{He}^*$ ) would have this property. This exciton might correspond to one of the long-lived metastable states of the isolated He atom (e.g., to the  $2^3S_1$  or  $2^1S_0$  states having energies about 20 eV above the ground state), and thus would tend to have a relatively long lifetime in the liquid. The mean free path of such an exciton, if it were sufficiently localized, should be similar to that of a  $\text{He}^3$  atom in liquid helium and should thus be quite long at low temperatures; in this case, the straight-line propagation of the neutral excitation would not be surprising. Alpha particles emitted by the source would indeed be expected to produce excited states of He, both by direct excitation of ground-state He atoms and by recombination of positive helium ions and electrons; the temperature dependence of the production of the observed neutrals might then be due to the strongly temperature-dependent diffusion of ions and neutrals near the source. The emergence of  $\text{He}_2^+$  ions from the surface of the liquid is also not too surprising since  $\text{He}_2^+$  molecular ions are commonly encountered when ions are studied in sufficiently dense helium gas.<sup>11</sup> The predominant creation mechanism of  $\text{He}_2^+$  in the gas phase is the reaction  $\text{He}^* + \text{He} \rightarrow \text{He}_2^+ + e^-$ , and a similar reaction might account for the conversion of the neutral ( $\text{He}^*$ ) at the liquid surface.<sup>11</sup> The precise energy relationships in such a reaction are, however, difficult to assess quantitatively since the excitation and ionization energies differ by unknown amounts in the condensed phase compared with those in vacuum.<sup>12</sup>

Further experiments are continuing with the aim of determining in greater detail the nature

of the neutral excitation discussed in the preceding paragraphs. We are also interested in studying the mechanism of electron escape from vortex rings at the liquid surface.

We gratefully acknowledge discussions with Dr. M. Silver concerning the functioning of tunnel-cathode electron sources in liquid helium, and discussions with Dr. Om P. Rustgi concerning the fabrication of thin aluminum films for use as far-ultraviolet filters. We also wish to thank Professor Marvin L. Cohen, Mr. T. I. Kamins, and Mr. G. J. Dick for helpful conversations.

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<sup>1</sup>G. Careri, in Progress in Low Temperature Physics, edited by C. J. Gorter (Interscience Publishers, Inc., New York, 1961), Vol. 3, pp. 58-79.

<sup>2</sup>F. Reif and L. Meyer, *Phys. Rev.* **119**, 1164 (1960), and *Phys. Rev. Letters* **5**, 1 (1960).

<sup>3</sup>G. W. Rayfield and F. Reif, *Phys. Rev.* **136**, A1194 (1964).

<sup>4</sup>R. J. Donnelly, Experimental Superfluidity (University of Chicago Press, Chicago, Ill., 1967), Chap. 6.

<sup>5</sup>G. Careri, U. Fasoli, and F. S. Gaeta, *Nuovo Cimento* **15**, 774 (1960); L. Bruschi, B. Maraviglia, and F. E. Moss, *Phys. Rev. Letters* **17**, 682 (1966).

<sup>6</sup>M. Silver, D. G. Onn, P. Smejtek, and K. Masuda, *Phys. Rev. Letters* **19**, 626 (1967).

<sup>7</sup>This detector is similar to a surface ionization detector conventionally used for detecting metastable atoms in vacuum. The large electric field (up to  $2 \times 10^5$  V/cm in our experiments) is essential when the detector is immersed in the liquid, presumably to overcome the large image force tending to keep ejected electrons near the metal plate. (Similar effects are encountered with tunnel-cathode electron sources operated in liquid helium; see Ref. 6.)

<sup>8</sup>No significant data could be obtained in this experiment at temperatures higher than 0.45°K since the magnitude of the anomalous current then becomes too small in the presence of the baffles.

<sup>9</sup>A discussion of such aluminum-film far-ultraviolet filters can be found in Om P. Rustgi, *J. Opt. Soc. Am.* **55**, 630 (1965).

<sup>10</sup>The LiF crystal was obtained from the Harshaw Chemical Company, Cleveland, Ohio. The transmission characteristics may be found in D. F. Heath and P. A. Sacher, *Appl. Opt.* **5**, 937 (1966), and H. W. Hohls, *Ann. Physik* **29**, 433 (1937).

<sup>11</sup>J. A. Hornbeck and J. P. Molnar, *Phys. Rev.* **84**, 621 (1951); see also F. L. Arnot and M. D. M'Ewen, *Proc. Roy. Soc. (London), Ser. A* **171**, 106 (1939).

<sup>12</sup>G. Baldini, *Phys. Rev.* **128**, 1562 (1962).