

$b_2 \sim b_3 \sim \omega_1/\gamma$, $b_1 \sim b_4 \sim 10^{-2}\omega_1/\gamma$, and $H_0 < b_1$.

To analyze the behavior of the frequencies at H a it is necessary to use the appropriate phase diagram [4].

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STABILIZATION OF NITROGEN ATOMS IN SUPERFLUID HELIUM

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It is shown that the stationary concentration of nitrogen atoms that are stabilized in superfluid helium exceeds 1.6%. The peculiarities of the recombination radiation have made it possible to estimate the recombination activation energy at 0.1 kcal/mole and to advance the hypothesis that solid nitrogen granules experience thermal explosion on going through the λ point.

The purpose of the present paper was to ascertain whether the recombination of atoms is an activated process (i.e., whether its realization calls for a supply of heat from the outside) and consequently determine whether it is possible, at least in principle, to stabilize large concentrations of atoms when the temperature approaches absolute zero. In the past, many experiments aimed at freezing atoms from a discharge on a substrate at 4.2°K (see the reviews [1, 2]) have led to the conclusion that the concentrations of the stabilized atoms do not exceed several tenths of one per cent. Similar results were obtained in experiments on radiative accumulation of atoms in solid molecular substances [1 - 3]. The developed theories [4 - 8] connected the existence of a limiting concentration atoms with instability to thermal explosion (the recombination process was assumed to be non-activated, and the diffusion of the atoms in the matrix was assumed to be activated). Analysis has shown, however, that the experiments described in the literature were performed at temperatures on the order of several times 10°K: at the instant of deposition, the temperature of the substrate increased to about 20°K [2]; an even higher temperature was possessed by the "hot points" in the case of radiative accumulation [1 - 3]. A significant lowering of the temperature in the region of intense sticking together of the atoms can lead to an appreciable increase of their stationary concentrations, both as a result of stabilization of the atoms in shallower traps of the matrix, and as a result of the appearance of a hypothetical activation energy of the recombination process.

Within the known procedures, it is impossible in principle to lower the temperature appreciably in the region of intense sticking. We have therefore decided to introduce the atoms in the form of a beam into the interior of cooled liquid helium, through its surface. Such a method would make it possible immediately (i) to lower the observation temperature to 1.2 - 1.5 K, (ii) thermalize the atoms before they arrive into the region of intense sticking, (iii) ensure the possibility of working with larger fluxes (10^{20} particles per second and more), owing to the effective heat removal in the superfluid helium.

To realize this method it was absolutely essential to produce a directional beam that reached directly the surface of the liquid; otherwise the atoms would be carried away by the intense rising flow of evaporating helium. Quite unexpectedly, the outflow of the warm (100 - 200°K) gas from a small opening into cold (1.5 - 3.0°K) helium gas under the influence of small pressure gradients ($\Delta P \approx P_{He}$) leads to the formation of a strongly pronounced beam. The

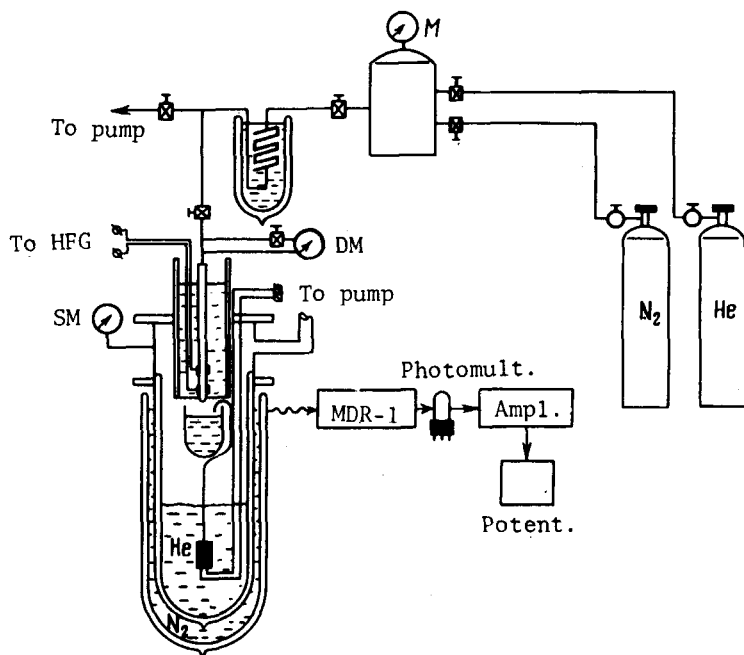


Fig. 1. Diagram of setup: M - manometer, DM - differential manometer, SM - standard manometer - vacuum meter, HFG - high-frequency generator.

reason is that the cold helium, which is denser by several orders of magnitude, "squeezes" the jet of warm gas as a result of heat transfer, as is the case in the flame of a gas burner. (The possibility of introducing directed gas beams into liquid helium is in our opinion also of independent interest.

The experimental setup is illustrated in Fig. 1. The experiments were performed mainly with nitrogen atoms, owing to the convenience with which recombination can be revealed with the aid of chemiluminescence in the visible region. The atoms were obtained by dissociating N_2 molecules in an electrodeless HF discharge, the discharge tube being cooled with liquid nitrogen. For a more complete dissociation of the N_2 molecules, the mixtures used in the HF discharge ($f = 40$ MHz, $P = 80$ W) were strongly diluted with helium, $[N_2]:[He] = 1:20$ to $1:1000$. The total gas flow ranged from 10^{18} to 10^{20} particles/sec (the outlet aperture of the discharge tube was ~ 0.75 mm). The distance between the source and the helium surface in the upper vessel was varied in the range from 2 to 5 cm.

The principal results of the experiments are the following:

1) It was possible to deliver a well-shaped beam¹⁾ up to a pressure of 20 Torr directly to the surface of HeII. The beam can be clearly seen because of the bright recombination glow. At a distance 3 cm from the source to the liquid, a crater up to 10 mm deep could be seen on the liquid surface (Fig. 2). The color of the flare varied over its length from orange-yellow at the source (the principal lines in the visible region at 585.4, 580.4, and 575.5 nm, corresponding to the $B^3\Pi \rightarrow A^1\Sigma_u^+$ transition of the N_2 molecule) to green (523, 540, and 560 nm). When the liquid helium evaporated from the vessel and the temperature rose, the flare became spread out, and its color became the same, orange-yellow, over its entire length.

2) Inside the liquid, and particularly pronouncedly in experiments with 1:1000 dilution,

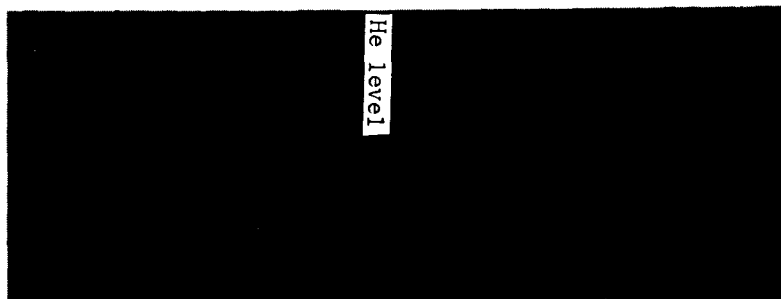


Fig. 2. Photograph of beam and of crater on the surface of liquid helium.

a dark-green ($\lambda \approx 523$ nm) glow was observed, propagating towards the bottom of the vessel at a constant speed. The glow intensity inside the liquid helium (probably due to the presence of finely dispersed particles containing N atoms) decreased with a time constant $\tau = 10 - 20$ sec. After turning off the discharge and after the almost complete disappearance of the glow, when the helium temperature increased slowly in the region $2.2 - 2.3^\circ\text{K}$ (i.e., near the λ point), sharp flashes of yellow glow were observed near the bottom of the vessel. Repeated subsequent passage through this temperature up and down, we observed each time (with decreasing intensity) the appearance of this glow, but only during the heating part of the cycle. Heating above 2.7°K again caused the appearance of the afterglow, which became gradually stronger. After evaporation of the helium from the vessel, the glow disappeared, and flashes with $\lambda \approx 523$ nm appeared again only at approximately 10°K , just as in [9, 10].

3) In the experiments with the 1:20 dilution, prolonged irradiation produced on the helium surface near the vessel walls a glowing edge ($\lambda \approx 523$ nm), which sometimes dropped to the bottom, still continuing to glow. Its afterglow time was also $10 - 20$ sec.

The intense recombination of the nitrogen atoms observed in the present study at 1.5°K (afterglow), and 2.2°K (flash near the λ point, and in the $2.7 - 4.2^\circ\text{K}$ range indicates that the concentration of the stabilized atoms increases noticeably with decreasing temperature. We can obtain a lower bound for this concentration from the following considerations: When the level of the HeII drops in the vessels, the vessel walls remain to be covered with a uniform dull deposit; immediately after the evacuation is stopped, however, this deposit is completely sublimated in explosive fashion at the expense of the recombination heat of the atoms present in the solid deposit. The sublimation heat of molecular nitrogen, Q_c , is ≈ 1.8 kcal/mole at $T = 20^\circ\text{K}$. The N_2 dissociation energy (E_D) per atom is 112.5 kcal/mole.^c The relative concentration of the atoms is therefore $n_N/n_{\text{N}_2} > Q_c/E_D = 1.6\%$.

The activation energy of the gross-process of recombination of the excess part of the atoms can be estimated from the universally used formula for the characteristic recombination time, $\tau = \tau_0 \exp(E_A/RT)$ [4, 5, 8], where $\tau_0 = 10^{-12} - 10^{-14}$ sec. For $\tau = 10 - 20$ sec at 1.5°K we have $E_A \approx 0.1$ kcal/mole, which is lower by one order of magnitude than the diffusion barrier assumed in [4 - 8]. To ascertain whether this activation energy is due to the lower-temperature diffusion or to the recombination act proper it is necessary to perform additional experiments.

The sharp recombination-glow flashes at $2.2 - 2.3^\circ\text{K}$ (i.e., near the λ point) are apparently connected with thermal explosion of granules containing nitrogen atoms. This explosion may be due to the recombination heat and the sharp decrease of the thermal conductivity of the helium on going through the λ point. Indeed, the relative increase of the temperature inside the granules on going through the λ point is quite appreciable and amounts to $\Delta T/T \approx 2(\kappa_{\text{N}_2}/\kappa_{\text{He}}) \geq 1$, where κ_{N_2} and κ_{He} are the thermal conductivities of the solid nitrogen and HeI, respectively².

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1) It contains apparently, besides the N atoms and N_2 molecules, also the clusters N_3 , N_5 , etc.

2) Estimates show that in general it is necessary to take also the Kapitza temperature jump into account.

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