Scintillation of Liquid Helium under Pressure. II. $2 > T > 0.3\,^{\circ}\mathrm{K}$*

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Measurements of the α-particle-produced scintillation intensity of liquid He^4 under pressure have been performed at temperatures ranging down to 0.3 °K. The data show a strongly temperature-dependent component of the pulse intensity which drops to zero for $T \lesssim 0.75$ °K. The principal pressure dependence of the intensity is in the component remaining below 0.75 °K. A physical interpretation of the results is presented which invokes metastable systems and the superfluidity of He II to explain the temperature dependence, and ionic recombination to explain the pressure dependence.

I. INTRODUCTION

Measurements of the scintillation intensity produced by α particles in pressurized liquid helium are reported in the preceding paper by Manning, Agee, Vincent, and Hereford[1] (MAVH, hereafter) for temperatures between 1.3 and 4.2 °K. As described in the introductory paragraphs of MAVH, [1] recent investigations, both in this laboratory[2] and in others, [3-5] have demonstrated that metastable atoms and molecules play an important role in the luminescence mechanism in He II.

We report here measurements of the scintillation intensity of pressurized liquid He^4 for temperatures ranging down to 0.3 °K. Furthermore, we show that at least a partial understanding of the observed phenomena can be achieved by considering the radiative destruction rate of the metastables produced near the α-particle's path.

II. EXPERIMENTAL ARRANGEMENT

The details of the pressurized scintillation chamber and the He^3 cooling system are shown in Fig. 1. The continuously operated He^3 refrigerator was capable of cooling the He^4 scintillation sample to 0.3 °K. The extreme uv radiation produced in the liquid helium by Po^{210} α particles was wavelength shifted by a coating of POPOP on the walls of the chamber and the radiation detected by a photomultiplier situated as shown. Scintillation current pulses (collection time ≈ 1.25 μsec) were amplified and analyzed as described by MAVH. [1] Again the position of the α peak (in channels) in the pulse-height spectrum was a measure of the scintillation pulse intensity (photons per α).

The temperature of the scintillation chamber was monitored continuously by standard resistance-thermometry techniques. A Speer carbon resistor, which had been ground down and varnished into a copper sleeve attached to the He^3 pot with Wood's metal, was calibrated against the vapor pressure of the He^3. The resistance value was measured using an ac bridge. Constant temperature was maintained by monitoring the null signal of the bridge on a lock-in amplifier and varying the pumping speed of the He^3 refrigerator with a parallel combination of valves to maintain a null at a given resistance reading. It was estimated that the temperature was held constant to within 3×10^{-3} °K.

He^4 was admitted to the scintillation chamber through a charcoal cold trap maintained at liquid nitrogen temperature in order to reduce impurities. By means of a throttling and bleeding system, the pressure in the chamber was maintained at a constant value to within approximately 0.5% above atmospheric pressure and to within 1 mm Hg below.

III. RESULTS AND DISCUSSION

In the initial series of observations the pulse intensity was measured for varying pressure with the temperature held fixed. Typical data are shown in Figs. 2 and 3. The data exhibit the same type of pressure dependence as that reported by MAVH. [1]

In a second series of experiments the pressure was held fixed, and the temperature was varied between 0.3 °K and approximately 2 °K. These results are shown in Figs. 4 and 5 along with certain empirically normalized data at higher temperatures taken from MAVH. [1] As the temperature is lowered, the pulse intensity begins to drop and levels off at a value approximately 15% lower at a temperature of about 0.75 °K.

The first striking feature of the data is the fact that, at temperatures below 0.75 °K, where the temperature dependent component of the pulse intensity has been eliminated, the characteristic pressure dependence (for fixed T) is still apparent. Hence, it is reasonable to assume that the pressure dependence is due principally to an intensity component which does not depend on temperature, viz., the component remaining in the pulse at the lowest temperatures.

Furthermore, the other component in the pulse,

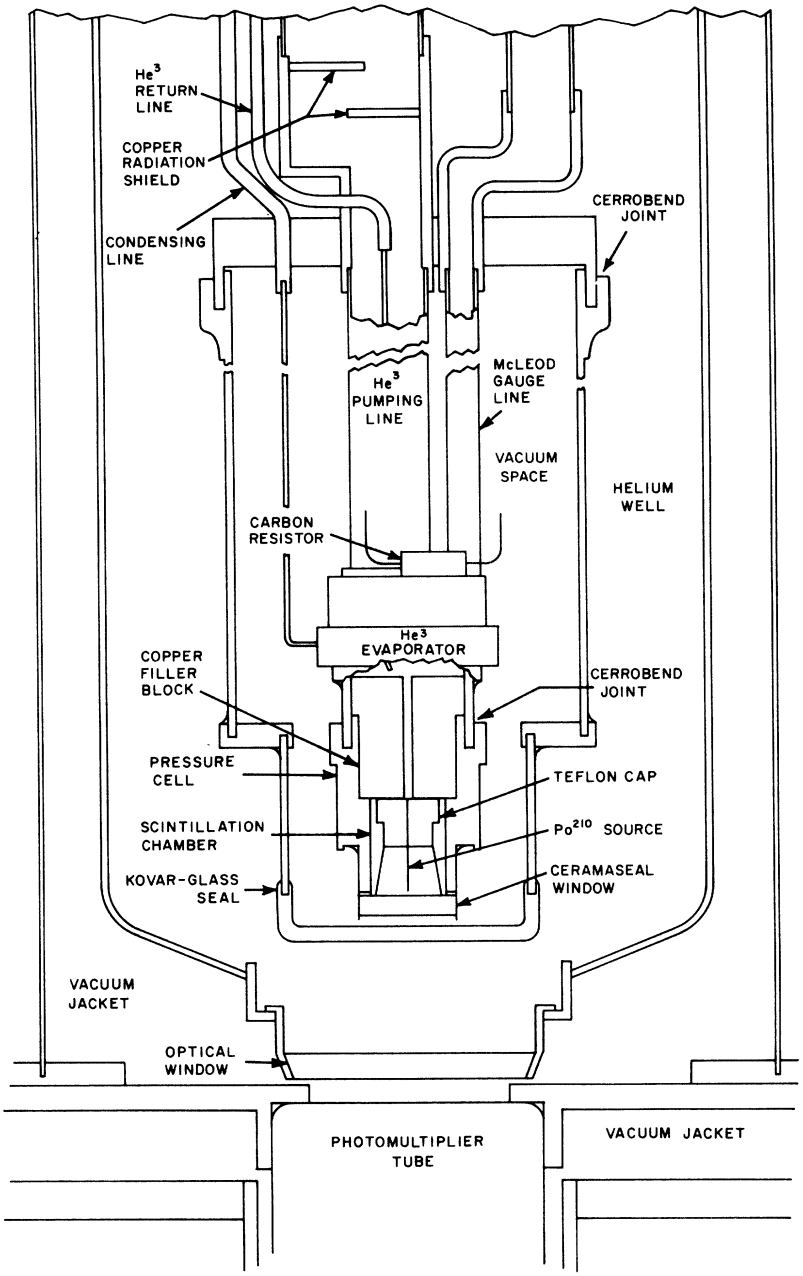

FIG. 1. Details of the experimental apparatus.

which appears as T is raised to T_λ, is seen to be only weakly dependent on the pressure. Hence, we assume that the total intensity $I(T, P)$ may be written as the sum of two components, $I_1(P)$ expressing the main pressure dependence, and $I_2(T, P)$ expressing the temperature dependence (with a weak pressure dependence):

$$I(T, P) = I_1(P) + I_2(T, P) . \quad (1)$$

We consider the I_2 component first and note that previously[2] we have shown that its temperature dependence at the vapor pressure can be fitted by assuming that it derives from some form of metastable excitons with a radiative destruction frequency proportional to ρ_n/ρ. This assumption leads to[2]

$$I_2(T, P) = I_0\{1 - e^{-KR_n}\} , \quad (2)$$

where $R_n(T, P) = \rho n(T, P)/\rho(T, P)$ and K is a constant.

In the present case both T and P are varied. The total density as a function of T and P is known, and values of $\rho_n(T, P)$ can be calculated from the Landau model, where (see, for example, Springett[6] or Mills[7])

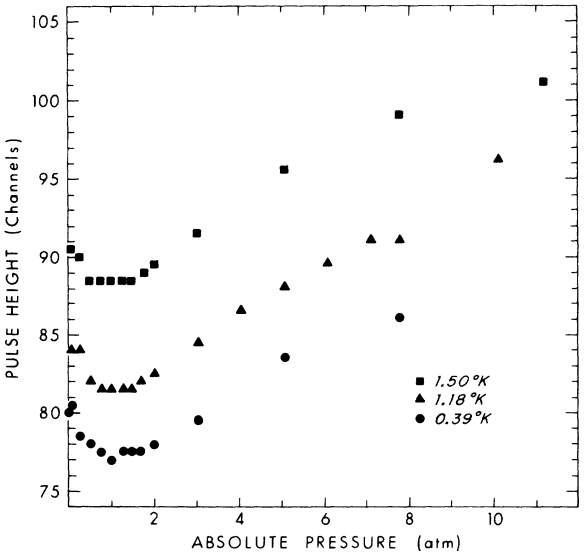

FIG. 2. Variation of scintillation intensity with pressure, with the temperature fixed at the indicated values.

$$\rho_n = \rho_{ph} + \rho_r ,$$

$$\rho_{ph} = \frac{\pi^2 (kT)^4}{45 \hbar^3 c^5} ,$$

$$\rho_r = \frac{2\mu^{1/2}\hbar}{3(2\pi)^{3/2}(kT)^{1/2}} \left(\frac{P_0}{\hbar}\right) \left[1 + \frac{6\mu kT}{\hbar^2} \left(\frac{P_0}{\hbar}\right)^{-2}\right] \times e^{-\Delta/kT} .$$

In these expressions, Δ and P_0 are the energy and momentum coordinates of the minimum in the Landau dispersion curve; μ is a constant related to the curvature of the minimum; and c is the velocity of first sound. All four of these quantities vary with temperature and/or pressure. Neutron scattering data yield the following expressions[6]:

$$\mu = 0.16\ (1 - 0.0217P) m_{He} ,$$

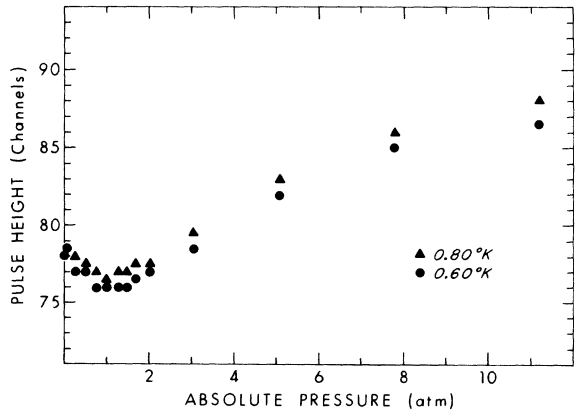

FIG. 3. Variation of scintillation intensity with pressure, with the temperature fixed at the indicated values.

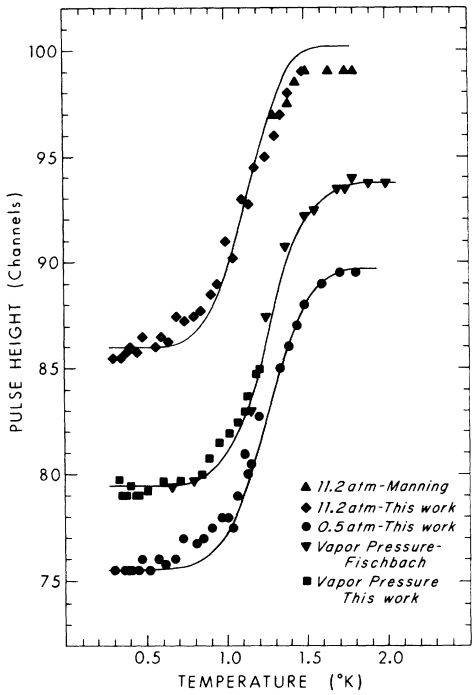

FIG. 4. Variation of scintillation intensity with temperature, with the pressure fixed at the indicated values. Data of Manning *et al.* (Ref. 1) and Fischbach *et al.* (Ref. 2), normalized to the data reported in this paper, are shown as indicated. The solid curves are calculated as described in the text.

$$P_0/\hbar = 1.91\ (1 - 0.0029P)\ \text{Å}^{-1} ,$$

$$\Delta/k = (8.68 - 0.0084T^7)(1 - 0.0075P)\ ^\circ\text{K} .$$

If the values of $R_n(T, P)$ are calculated from the above equations, then using Eq. (2), the data can be fitted quite well by the expression

$$I(T, P) = I_1(P) + 14.25\ (1 - e^{-KR_n(T,P)}) . \qquad (3)$$

The solid curves in Figs. 4 and 5 were calculated from Eq. (3), for $K = 20$, using for $I_1(P)$ the average of the measured intensities at 0.39 and 0.60 °K shown in Figs. 2 and 3. The experimental data in Figs. 4 and 5 suggest that the intensity is the same at these two lower temperatures, despite the difference of approximately 1% in the results shown in Figs. 2 and 3. This latter difference is within the run-to-run repeatability of the measurements.

In comparing the calculated curves of Figs. 4 and 5 with the experimental points, it should be noted that the pressure dependence is more subtle than simply the addition of a *single* calculated curve for $I_2(T)$ to the averaged experimental data for $I_1(P)$ at 0.39 and 0.60 °K. The curves accurately reflect the fact that, as T is lowered, the decrease in $I_2(T, P)$ begins at decreasingly smaller values of

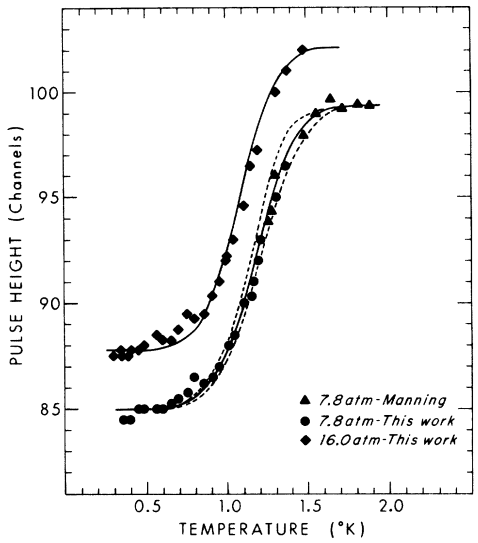

FIG. 5. Same as Fig. 4. The dotted curves show the effect of a ± 25% variation of the constant K in Eq. (3) of the text.

T for progressively higher values of P (1.4 °K for 16 atm, as compared to 1.75 °K at the vapor pressure). The origin of this lies simply in the decrease of T_λ with increasing pressure and the corresponding increase of R_n (for a given temperature below T_λ). The fact that this pressure dependence, as well as the temperature dependence, of $I_2(T, P)$ is provided by Eq. (3) is additional evidence of its validity.

IV. PHYSICAL MODEL FOR $I_2(T,P)$

A mechanism must sought for component I_2 which provides a destruction probability for metastables proportional to the density of the normal fluid. This requirement, however, is not too precise. From Fig. 5 it can be seen that with K changed by ± 25%, Eq. (3) still fits the data quite well. In other words, the strong temperature dependence of R_n is the predominant factor, and the exponent in Eq. (3) could contain a multiplicative factor which could be quite temperature dependent. For example, $\sqrt{T}$ (or its reciprocal), which changes by about 30% between 1.5 and 0.75 °K, could be inserted into the exponent with an appropriate adjustment of the constant K, and Eq. (3) would still fit the data reasonably well.

With this in mind, one can examine various metastable destruction mechanisms to determine whether they could provide a physical basis for Eqs. (2) and (3). The first obvious possibility is collisions with phonons and rotons, the densities of which are proportional to ρ_n (apart from a factor T). However, it seems most unlikely that interaction with such excitations of the fluid could be as effective as interaction with the large density of ions and other metastable systems in the vicinity of an α particle's path. This conjecture is supported strongly by the fact that there is no inhibition in He II of scintillation pulses produced by β particles, [8] for which ion and metastable densities near the particle track are orders of magnitude less than for α's. Presumably the larger densities near an α track yield radiation from certain metastables within the pulse, this radiation being delayed beyond it in the case of β's (which are known to produce metastables[3–5]). If phonon or roton interactions destroyed the metastables, it would be difficult to understand why there is no reduction in the pulse intensity below T_λ with β's.

The possible influence of impurities should be mentioned. We do not believe that they play a significant role in the observations reported here. Although the He^4 was admitted to the scintillation chamber through a liquid-nitrogen-cooled charcoal trap, POPOP contamination from the walls and other impurities could be present after liquefaction. It is difficult to see why POPOP suspended in the bath would function differently as a wave shifter than that on the walls, unless it settled out below T_λ. In this case again the inhibition effect should be present for β's as well as α's, which is not the case. Furthermore, unpublished work in this laboratory has shown that significant heat currents produce no change in scintillation intensity. If impurities were operative in the scintillation mechanism, a heat current could reasonably by expected to have an effect.

The question then remains as to whether destruction in ion-metastable or metastable-metastable collisions can provide a physical model for Eq. (2). Since most ions should recombine quickly following emission of an α, one might expect metastable-metastable collisions to be the more important process. The results of Hill *et al.*, [9] in fact, indicate that in the case of $a^3\Sigma_u$ metastables produced by an electron beam, their "lifetime" is about 0.7 msec, and their deexcitation is dominated by $a^3\Sigma_u - a^3\Sigma_u$ encounters, yielding a nonexponential decay. On the other hand, they find an approximately exponential decay in the case of the 2^3S_1 atomic state.

If we assume that the metastable species responsible for the I_2 component are destroyed through collisions with some "other" species of longer lifetime, a physical basis for Eqs. (1) and (2) can be established which is at least qualitatively satisfactory. The problem can be approached using the approximate treatment employed by Jaffé[10] for the columnar recombination of ions near an α track.

Assume that a metastable species of density $n(r, t)$ is responsible for I_2 and undergoes radiative destruction through collisions with another more

stable species of density $N(r, t)$. The variable r is the radial distance from the α track, and t is measured from the time of alpha emission. Assume further that the more stable systems simply diffuse outward at a rate determined by a diffusion constant D, such that

$$\frac{\partial N}{\partial t} = D\nabla^2 N .$$

For the case of an initial Gaussian distribution of mean square radial width b^2, the cylindrically symmetric solution of this equation is

$$N(r, t) = \frac{N_0}{\pi(4Dt + b^2)} \exp\left(-\frac{r^2}{4Dt + b^2}\right) . \quad (4)$$

In the case of the metastables of density $n(r, t)$, they undergo destruction as well as diffusion, and we assume $n(r, t)$ to be determined by

$$\frac{\partial n}{\partial t} = -\nu N n + D\nabla^2 n , \quad (5)$$

where ν is a destruction frequency, assumed to be a constant. Following the method of Jaffé, [10] we solve this equation approximately by neglecting the diffusion term in Eq. (5), and by assuming that $n(r, t)$, although decreasing as a result of destruction, diffuses outward with a Gaussian form

$$n(r, t) = f(t) \frac{n_0}{\pi(4Dt + b^2)} \exp\left(-\frac{r^2}{4Dt + b^2}\right) , \quad (6)$$

where n_0 is the initial number of metastables per unit track length, and $f(t)$ represents their decrease with time due to destruction (assumed to be the same at all values of r).

Substituting expressions (4) and (6) into Eq. (5), omitting the last term, multiplying by $2\pi r dr$, and integrating over r from 0 to ∞, we obtain the following differential equation for $n_0 f(t)$, the total number of metastables surviving destruction:

$$\frac{d(n_0 f)}{dt} = -\frac{\nu N_0}{2\pi(4Dt + b^2)} (n_0 f) .$$

Solving for $n_0 f(t)$, we find that

$$n_0 f(t) = n_0 \exp\left[-\frac{\nu N_0}{8\pi D} \ln\left(\frac{4Dt + b^2}{b^2}\right)\right] .$$

The pulse intensity component I_2 will then be simply the difference between the initial number of metastables n_0 and the number surviving at t' (≈ 1.25 μsec) or

$$I_2(T, P) = n_0 \left\{1 - \exp\left[-\frac{\nu N_0}{8\pi D} \ln\left(\frac{4Dt' + b^2}{b^2}\right)\right]\right\} . \quad (7)$$

The temperature and pressure dependences are contained in the diffusion coefficient D which rises exponentially as T drops below T_λ and is also pressure dependent.

Consideration of the magnitudes of the quantities in the logarithmic term of the exponent shows that this term depends only weakly on D. Tenner[11] has estimated the radius of the electron cloud along a particle's ionization column to be about 3×10^{-7} cm (the positive-ion cloud is even smaller). We shall assume the initial radius (after thermalization) of the metastable distribution (which derives partially from ionic recombination) to be at the most 3×10^{-6} cm; hence, b^2 is of the order 10^{-11} cm^2.

The value of D should be at least about 10^{-4} cm^2 sec^{-1}, which is the diffusion coefficient of He^3 atoms in very dilute liquid He^3-He^4 solutions at 2 °K. Hence, for $t' = 1.25$ μsec

$$4Dt'/b^2 \gtrsim 50 .$$

As the temperature drops and D increases, this ratio will grow even larger. Consequently, the logarithmic term will depend very weakly on D, and the $1/D$ factor will dominate the temperature and pressure dependence of the exponent in Eq. (7).

Using He^3 diffusion data and the known relation between temperature and the normal-fluid density,[12] we can express $1/D$ (employing a constant C) as

$$\frac{1}{D} \sim \exp\left[-\frac{\Delta}{kT}\right] = C\sqrt{T}\, R_n(T, P) .$$

Equation (7) may now be written as

$$I_2(T, P) = n_0 (1 - e^{-KR_n(T,P)}) ,$$

where

$$K = \frac{C\nu N_0\sqrt{T}}{8\pi} \ln\left(\frac{4Dt' + b^2}{b^2}\right) .$$

Thus, within the flexibility stated at the beginning of this section, we obtain an approximate expression for $I_2(T, P)$ of the form of Eq. (2).

Whether the $2\,^3S_1$ states observed by Hill *et al.*[9] are responsible for the I_2 component in the pulse measurements reported here cannot be stated definitely, but these states seem to be likely candidates. Although Hill *et al.* indicate $2\,^3S_1$ lifetimes of the order of 0.5 msec when produced by an electron beam, the lifetime could be significantly less in the much larger excitation density near an α track. On the other hand, singlet metastable states could be the species involved.

Although we believe metastable–"other-metastable" collisions to be the dominant destruction mechanism, we do not think that destruction in collisions with ions can be ruled out. We intend to investigate the effect of an electric field in order to clarify the mechanism further. Also, it should be pointed out that the proposed mechanism does not preclude other processes which could be operative at times considerably later than the scintillation pulse.

V. $I_1(P)$ COMPONENT

The question remaining at this point is the physical origin of $I_1(P)$. This component presumably derives from recombination and from the fast deexcitation of excited atoms and molecules. The pressure dependence shown in Figs. 2 and 3 is present for both He I and He II, and we have observed it recently in liquid He^3 (unpublished results). It is not peculiar to He II.

An obvious origin of the effect is the compression of the vapor bubbles which are known to form around both negative ions and metastable systems.[9] Since the application of several atmospheres of pressure increases $I_1(P)$, *but does not restore the intensity lost from* $I_2(P)$ *at low temperatures*, we assume that the pressure effect involves negative ions rather than metastable states. Previous observations[13] have demonstrated that ionic recombination plays a contributing role in the scintillation process in liquid helium.

In considering the role of recombination, one should recognize first the fact that a certain fraction of the ions produced will escape prompt recombination. Onsager[14] has shown that in high pressure gases the probability of an electron escaping recombination with its parent is given by $e^{-r_c/r}$, where $r_c = e^2/kT$, the critical distance at which the electrical potential energy is equal to kT. In gases in which electron attachment occurs, the electron is presumed to escape unless it is captured to form a negative ion within r_c. A similar situation should occur in liquid helium in which the electron forms a cavity of radius about 16 Å with an effective mass of approximately 100 m_{He}.

We suggest that the probability of an electron escaping prompt recombination, and thereby reducing the scintillation pulse intensity in liquid helium is pressure dependent. The effect could be quite complex, involving the rate of bubble formation as the electron drifts outward, the mobility of the bubble structure once formed, and the ultimate "tunneling out" of the electron to recombine with a positive ion.

It is tempting to attribute the effect to mobility alone, since the mobility of negative ions under pressure first rises to a maximum at several atmospheres (near the minimum in Figs. 2 and 3) and then decreases along with the positive-ion mobility.[15] However, the $I_1(P)$ component fails to respond to the substantial increase in mobility which occurs as T is lowered below T_λ. Hence, we conclude that the bubble-formation process or the "tunneling" of the electron to recombine must be dominant here.

A pressure dependence of the escape probability of electrons from the ion column would be consistent with the results of Onn and Silver.[16] They observe a pressure dependence of the injection into liquid He^4 of hot electrons from cold-cathode emitters. They are able to account for their results by introducing a "thermalization range" which decreases with increasing pressure causing more electrons to be repelled by an injection barrier and return to the cathode.

In the recombination case, such a decrease in the range within which thermalization and/or "bubble" formation occurs would increase the recombination radiation, as observed for $P \gtrsim 1$ atm (Figs. 2 and 3). However, we are unable to account for the small decrease between 0 and 1 atm.

We wish to express our indebtedness to Dr. Frank E. Moss for many helpful discussions regarding interpretation of these experiments.

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