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traceable largely to the terms second order in electron polarization in the denominator of Eq. (1). Although the model does not quantitatively fit the overall slope of the data in Fig. 1 with all parameters fixed, a 25% variation of the saturating ensemble width  $\Omega$  with temperature, over the entire temperature range, would be sufficient to fit the observations. More importantly, we have not taken into account possible variation of g(x)with temperature.

It should be noted that proton polarizations larger than the 67 % reported here might be possible at 0.5 K, with porphyrexide concentrations other than 0.75 %. However, this concentration was found to be approximately optimal at 1 K.

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<sup>1</sup>S. Mango, Ö. Runólfsson, and M. Borghini, to be published; M. Borghini, Bull. Am. Phys. Soc. <u>14</u>, 189 (1969).

<sup>2</sup>A butanol-water target has recently been put into operation at 1 K at Argonne National Laboratory, and similar targets are in use at CERN and at the Stanford Linear Accelerator Center.

<sup>3</sup>For a description of the <sup>3</sup>He cryostat and associated superconducting solenoid, see L. R. Windmiller and J. B. Ketterson, Rev. Sci. Instr. 39, 1672 (1968).

<sup>4</sup>The butanol was reagent grade from J. T. Baker

Company, Phillipsburg, N. J.; the porphyrexide was

obtained from K and K Laboratories, Plainview, N. Y. <sup>5</sup>Fluorinated-ethylene polymer, supplied by Cadillac Plastics and Chemical Company, Chicago, Ill.

<sup>6</sup>A polarization of 50% at 1.1 K in a 1-mg sample of ethylene glycol has been reported by H. Glättli, M. Odehnal, J. Ezratty, A. Malinovski, and A. Abragam, Phys. Letters <u>29A</u>, 250 (1969).

<sup>7</sup>From measurements on similar hydrocarbons carried out at CERN, it has been estimated that as much as 90% of the total power goes to the cavity wall. A. Masaike, private communication.

<sup>8</sup>O. S. Leifson and C. D. Jeffries, Phys. Rev. <u>122</u>, 1781 (1961).

<sup>9</sup>In the "solid effect," this behavior could be accounted for if the proton "leakage" parameter were large at 1 K and decreasing at lower temperatures. Since the (porphyrexide) electrons dominate the proton relaxation, this should not be the case.

<sup>10</sup>A. Abragam and M. Borghini, in <u>Progress in Low</u> <u>Temperature Physics</u>, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, The Netherlands, 1964), Vol. IV, Chap. 8; A. V. Kessenikk, A. A. Manenkov, and G. I. Pyatnitskii, Fiz. Tverd. Tela <u>6</u>, 827 (1964) [translation: Soviet Phys.-Solid State <u>6</u>, 641 (1964)]; M. Borghini, Phys. Letters <u>26A</u>, 242 (1968).

<sup>11</sup>C. F. Hwang and D. A. Hill, Phys. Rev. Letters <u>19</u>, 1011 (1967).

 $^{12}$ Our tests show that the maximum solubility of porphyrexide in this host is 1.5% (by weight) at room temperature. That is, the saturated concentration is about twice that used in the present experiment.

<sup>13</sup>The parameter t is proportional to microwave power. The value listed here corresponds to 20 dB on the power scale of Fig 3. The saturating ensemble was assumed to be Gaussian in shape, as in Ref. 11, with a width of  $\Omega$ .

## PERSISTENT METASTABLE STATES AND THE INHIBITED SCINTILLATION OF HE II \*

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The scintillation of liquid helium produced by  $\alpha$  particles has been measured with particular emphasis on the temperature region below 1.25°K which had not been studied previously. An interpretation of the observed inhibition of the scintillation below  $T_{\lambda}$  is proposed which attributes the effect to a reduced radiative destruction rate of metastable states in He II.

Among the unusual properties of superfluid helium, which are not yet understood, is the inhibition of its scintillation<sup>1,2</sup> (produced by  $\alpha$  particles) compared with that of normal-liquid helium. Experimental studies, all at temperatures above 1.25°K, of the effect on the scintillation intensity of an electric field,<sup>3</sup> a heat flux,<sup>4</sup> and rotation of the fluid<sup>5</sup> have failed to provide sufficient information to establish a mechanism for the scintillation process and the inhibition effect. It has been possible to conclude only that the radiation derives in roughly equal degree from (1) the deexcitation of some sort of atomic system or exciton and (2) processes in which ion recombination plays a contributing role.

We have now extended the measurement of the

scintillation intensity down to approximately  $0.3^{\circ}$ K and have been able to correlate the data with the density of the normal fluid below  $T_{\lambda}$  by means of a rather simple model.

A He<sup>3</sup> refrigerator was employed to cool a liquid-He<sup>4</sup> scintillation chamber which contained a Po<sup>210</sup> source of 5.28-MeV  $\alpha$  particles, and which was coated on its inner surfaces with a wavelength shifter.<sup>4,5</sup> Analysis of the pulses from a photomultiplier which viewed the chamber yielded the scintillation intensity (photons per  $\alpha$  particle) in arbitrary units. The results are shown in Fig. 1 and indicate that as the temperature is reduced below  $T_{\lambda}$ , the intensity drops by about 15%, leveling off at 0.9°K, and remaining roughly constant at lower temperatures.

A subsidiary experiment which was performed is pertinent to interpretation of the results. In the scintillation measurements, photons were detected only during a 1.25- $\mu$ sec period following emission of an  $\alpha$  particle. Using a currentto-frequency conversion technique, we have made an effort to detect <u>all</u> photons, regardless of emission time. The results indicate that the total photon emission per alpha is not inhibited below  $T_{\lambda}$  and remains constant to within approx-



FIG. 1. The scintillation intensity versus temperature. The solid curves below  $T_{\lambda}$  are calculated as described in the text, and the dotted curve above  $T_{\lambda}$  is drawn to fit the experimental points.

imately 3%. This additional result suggests that the inhibition of scintillation is caused by <u>delayed</u> photon emission from some type of atomic system, and one thinks immediately of metastable states of helium atoms or molecules.

Previous investigators<sup>6,7</sup> have proposed collision-induced decay of metastable atoms as a source of scintillation of helium gas. Furthermore, the decay of metastables has been observed in the afterglow of helium discharges.<sup>8</sup> In this latter work, both the 2<sup>1</sup>S and 2<sup>3</sup>S states were studied, the first undergoing radiative destruction much faster than the second, which (after conversion to  $2^{3}\Sigma$  molecules) can persist for up to 0.05 sec. In the case of liquid helium, metastable states have been proposed by Surko and Reif<sup>9</sup> to explain neutral excitations observed at temperatures below about 0.7°K which propagate through He II after being produced by  $\alpha$  particles.

Without specifying a particular metastable state, one can infer from Phelps' work<sup>8</sup> that at high gas pressures the metastable density *n* will be determined by a destruction frequency  $\nu$ , which depends on the atom density *N*:

$$\partial n/\partial t = -\nu(N)n$$
.

The data below  $T_{\lambda}$  in Fig. 1 can be accounted for by the simple assumption that the destruction frequency is proportional to the fractional density of the normal fluid  $\rho_n(T)/\rho(T)$ , that is,  $\nu$ =  $[\rho_n(T)/\rho(T)]\nu_0(N)$ . One then obtains for the density of radiative destruction events occurring within time t' after emission of an  $\alpha$  particle

$$n(0)-n(t')=n(0)\left\{1-\exp\left[\frac{\rho_n(T)}{\rho(T)}\nu_0t'\right]\right\}$$

With the assumption that metastables account for approximately 15% of the intensity immediately below  $T_{\lambda}$ , and using the known temperature dependence of  $\rho_n/\rho$ ,<sup>10</sup> the data in the He II region can be fitted remarkably well by the expression

$$I(T) = 82.5 + 15 \left\{ 1 - \exp\left[ -\frac{\rho_n(T)}{\rho(T)} \nu_0 t' \right] \right\}$$

The two solid curves in Fig. 1 represent Eq. (1) for  $\nu_0 t'$  values of 11.6 and 14.5 and show that a change in this coefficient of 25% does not appreciably alter the temperature dependence, demonstrating the dominant role of the normal-fluid density.

A value of 13 for  $\nu_0 t'$  (with  $t' = 1.25 \ \mu sec$ ) yields an average cross section for the destruction of the metastables of  $5 \times 10^{-20}$  cm<sup>2</sup> in reasonable agreement with Phelps' value of  $3 \times 10^{-20}$  cm<sup>2</sup> for the destruction of the  $2^{1}S$  state. However, the validity of a comparison of data for liquid helium with that for helium gas at 300°K is doubtful.

The observations reported here were carried out for temperatures below  $T_{\lambda}$ . For the sake of completeness, previous data<sup>5</sup> (normalized at  $2^{\circ}$ K) are shown in Fig. 1 for temperatures above  $T_{\lambda}$ . The dotted line above  $T_{\lambda}$  is simply a smooth curve drawn through the experimental points with a sharp drop in intensity at  $T_{\lambda}$ .<sup>5</sup> Since this drop has been observed to disappear when a small overpressure is maintained, it may be related to the cessation of internal boiling at  $T_{\lambda}$ . The rise in intensity above  $T_{\lambda}$  is probably related to the 15% decrease in fluid density between  $T_{\lambda}$ and  $4.2^{\circ}$ K, and we hope that a refinement of the simple model proposed may account for it. The structure exhibited near 0.5°K is barely outside of the reproducibility of the data and may be instrumental in nature. In any case, we are unable to offer an interpretation.

As for the inhibition of scintillation below  $T_{\lambda}$ , the role of metastable atoms was suggested by Moss and one of us (F.L.H.)<sup>1</sup> in the first report of the anomalous scintillation behavior of HeII. We believe that the data below  $T_{\lambda}$  show that the simple correlation between the destruction frequency and  $\rho_n$  has meaning and represents a reduced destruction rate of metastable atoms in

He II. Superfluid liquid helium may be a uniquely suitable medium for the production and retention of high densities of metastable helium atoms and molecules.

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HEAT CAPACITY AT CONSTANT PRESSURE NEAR THE SUPERFLUID TRANSITION IN He<sup>4</sup>

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We present high-precision data for  $C_p$  near  $T_{\lambda}$  and give three interpretations in terms of scaling predictions. We find no interpretation fully in agreement with the predicted symmetry of the transition and a divergent  $C_p$ .

Several measurements near the superfluid transition temperature  $T_{\lambda}$  of<sup>1-5</sup> He<sup>4</sup> have yielded quantitative confirmation for this critical point of theoretical predictions based upon "scaling."6-10 It is the purpose of this communication to present measurements of the heat capacity at saturated vapor pressure  $C_s$  near  $T_{\lambda}$ . These results agree in detail with theoretical predictions<sup>6-8</sup> only if the heat capacity at constant pressure  $C_p$  is finite at  $T_{\lambda}$ . In order to confirm these observations measurements also were made of the heat capacity at constant volume  $C_{\nu}$  and of  $(\partial P/\partial T)_{\nu}$ (P is the pressure) at a molar volume V=26.81cm<sup>3</sup>. From these quantities  $C_{\rho}$  can be calculated reliably. These results, although less precise, are consistent with the conclusions based on  $C_s$ .

The measurements were made in the apparatus used for studies of the thermal conductivity of HeI near  $T_{\lambda}$ .<sup>3</sup> The temperature resolution was  $10^{-7}$  °K which limited the precision of  $C_s$  for |t|