<u>40,</u> 2208 (1964). ¹⁰G. K. Born, Phys. Rev. <u>169</u>, 155 (1968).

¹¹R. S. Mulliken, Phys. Rev. 136, A962 (1964).

¹²C. B. Collins and W. B. Hurt, Phys. Rev. 177,

257 (1969).

¹³The experimental conditions under which these

¹⁴M. Gryzinski, Phys. Rev. <u>115</u>, 374 (1959).

¹⁵T. Holstein, Phys. Rev. <u>72</u>, 1212 (1947).

¹⁶C. B. Collins, Phys. Rev. <u>175</u>, 160 (1968).

PHYSICAL REVIEW

VOLUME 179, NUMBER 1

5 MARCH 1969

Surface Tension at the He³-He⁴ Interface*

W. Brouwer[†]

Theoretical Physics Institute, Department of Physics, University of Alberta, Edmonton, Canada

and

R. K. Pathria

Department of Physics, Panjab University, Chandigarh, India (Received 14 October 1968)

An estimate is made of the interfacial surface tension in a phase-separated mixture of He^3-He^4 on the basis of Atkins's approach. A corresponding experimental study should elucidate the role played by the vibrational surface modes of a quantum fluid.

According to Atkins¹ and Brouwer and Pathria² the surface tension $\sigma(T)$ of liquid helium II may be written as

$$\sigma(T) = \sigma_i(T) + \sigma_v[T, \sigma_i(T)], \qquad (1)$$

where σ_i is the so-called "intrinsic" surface tension of the liquid that arises from the interatomic interactions, while σ_v is the contribution made by the quantized vibrational modes of the free surface. These vibrational modes are assumed to be similar in nature to the macroscopic capillary waves whose (frequency-dependent) phase velocity area v is given by the relationship

$$v = (2\pi\sigma/\lambda\rho)^{1/2} , \qquad (2)$$

 ρ being the mass density of the liquid and λ the wavelength of the mode in question. In the spirit of the Debye theory of solids, one chooses a cut-off frequency ν_c such that the total number of normal modes of vibration in the surface becomes equal to the number of molecules in a monomolecular layer at the surface. One thereby finds that a significant part of the total surface energy of the liquid comes from the presence of these surface modes.

At absolute zero again, the surface tension of liquid helium consists of the intrinsic surface tension $\sigma_i(0)$ and the zero-point energy of the surface modes $\sigma_{ij}(0)$. A theoretical estimate of $\sigma_i(0)$ has been made by Brouwer and Pathria² on the basis of the imperfect gas model of Gross³ and Pitaevski,⁴ where one calculates the energy associated with the nonuniformity that exists in the surface region of the liquid and identifies it with the intrinsic surface tension $\sigma_i(0)$. To obtain agreement with the experimental value⁵ of $\sigma(0)$, viz. 0.37 erg cm⁻², one concludes that $\sigma_i(0) \simeq 0.14$ erg cm⁻² and $\sigma_v(0) \simeq 0.23$ erg cm⁻². The temperature-dependent part of $\sigma(T)$, on the other hand, is reasonably well explained by the free energy contribution arising from the ripplons alone, with the result^{1,2}

$$\sigma(T) \simeq \sigma(0) - a T^{7/3}.$$

where $a = 6.5 \times 10^{-3} \text{ erg cm}^{-2} \circ \text{K}^{-7/3}$.

Subsequently, Lovejoy⁶ pointed out that the experimental results on the surface tension of liquid He³ can be understood on the basis of the existence of surface modes of vibration. It then turns out that the experimentally extrapolated value $\sigma(0)$ of the zero-point surface tension of

179

0.15 erg cm⁻² in this case, is to be regarded as made up of an intrinsic surface tension $\sigma_i(0) \simeq 0.04$ erg cm⁻² and a vibrational part $\sigma_v(0) \simeq 0.11$ erg cm⁻². It may be mentioned here that the foregoing value of $\sigma_i(0)$ can also be obtained theoretically⁷ by studying the non-uniformity existing in the surface region of liquid He³.

The main purpose of the present note is to suggest that on the basis of the foregoing results, one can make a theoretical estimate of the He³- He⁴ interfacial tension at T = 0°K and thereby provide an experimental test for the validity of the considerations on which Eq. (1) is based.

For this purpose we note that, according to the "classical" theory of surface tension, in which the zero-point energy of the surface modes of vibration plays no part, the interfacial tension $\sigma_{34}(0)$ may be obtained by applying Antonov's rule,⁸ whence it follows that

$$\sigma_{34}(0) \simeq \sigma_4(0) - \sigma_3(0), \tag{4}$$

 $\sigma_4(0)$ being the equilibrium surface tension between liquid He⁴ and its vapor⁹ and $\sigma_3(0)$ the equilibrium surface tension between liquid He³ and its vapor. Correcting $\sigma_4(0)$ for the fact that, on phase-separation at 0°K, the He⁴ part contains a 6% admixture of He³ in it, ¹⁰ we obtain for the zero-point interfacial tension

$$\sigma_{34}(0) \simeq 0.36 - 0.15 \simeq 0.20 \text{ erg cm}^{-2}$$
. (5)

On the other hand, if we believe the philosophy behind Eq. (1) to be correct, then the interfacial tension $\sigma_{34}(0)$ must as well be regarded as made up of two contributions, i.e.,

$$\sigma_{34}(0) = \sigma_{34i}(0) + \sigma_{34v}(0) , \qquad (6)$$

Where, as usual, σ_{34i} is the intrinsic interfacial tension while σ_{34i} is the contribution made by the capillary waves at the interface. Clearly enough, it is only the intrinsic interfacial tension σ_{34i} which can be obtained by applying the Antonov rule; and taking into account the 6% concentration of He³ in He⁴ at T = 0°K, we obtain

$$\sigma_{34i}(0) \simeq \sigma_{4i}(0) - \sigma_{3i}(0)$$

 $\simeq 0.094 \text{ erg cm}^{-2}.$ (7)

The contribution σ_{34v} of the capillary waves can also be readily calculated. The phase velocity ot these interfacial waves is given by¹¹

$$v = [2\pi\sigma/\lambda(\rho_1 + \rho_2)]^{1/2} , \qquad (8)$$

where ρ_1 and ρ_2 are the respective densities of the liquids concerned. The cutoff frequency ν_C of these modes can be determined by requiring that their total number per unit area of the interface be equal to the difference between the number of molecules per unit area in a monomolecular layer of the high-density phase on the one hand and the low-density phase on the other. It turns out that $\nu_C \simeq 4.3 \times 10^{10} \text{ sec}^{-1}$ which may be compared with the values $9.5 \times 10^{10} \text{ sec}^{-1}$ and $1.5 \times 10^{11} \text{ sec}^{-1}$ for the pure cases of liquid He³ and liquid He⁴, respectively. Carrying out the relevant calculation for the zero-point energy of the interfacial waves, in the same way as was done in Refs. 1 and 2, we obtain

$$\sigma_{34v}^{(0)} \simeq 0.007 \ \mathrm{erg} \, \mathrm{cm}^{-2},$$
 (9)

which is extremely small in comparison with the intrinsic surface tension. ⁷ Consequently, the theoretical estimate for the He^3-He^4 interfacial tension would be

$$\sigma_{34}(0) \simeq 0.10 \text{ erg cm}^{-2},$$
 (10)

which is about one-half of the "classical" estimate. An extrapolation of $\sigma_{34}(T)$ determinations should, therefore, be able to decide the issue between the two estimates and provide a practical justification for expressing the surface tension of a quantum liquid in the form of Eq. (1).

In conclusion, it appears worthwhile to point out that evidence has already been obtained for the existence of gapless surface excitations in liquid He^4 , in that the specific heat measurements of Brewer *et al.*¹² are found to be "consistent with the presence of excitations associated with the free liquid surface ...".

ACKNOWLEDGMENT

We are grateful to Dr. J. P. Franck for comments on our earlier work,² that led to the present investigation.

*Supported in part by the National Research Council of Canada.

[†]Present address: Institute for Theoretical Physics,

University of Amsterdam, Amsterdam, The Netherlands. ¹K. R. Atkins, Can. J. Phys. <u>31</u>, 1165 (1953).

²W. Brouwer and R. K. Pathria, Phys. Rev. <u>163</u>, 200 (1967).

³E. P. Gross, Nuovo Cimento <u>20</u>, 454 (1961).

⁴L. P. Pitaevski, Zh. Eksperim. i Teor. Fiz. <u>40</u>, 646 (1961) [English transl.: Soviet Phys. - JETP 13, 451

(1961)].

- ⁵K. R. Atkins and Y. Narahara, Can. J. Phys. <u>33</u>, 49 (1955).
 - ⁶D. R. Lovejoy, Can. J. Phys. <u>33</u>, 49 (1955).
 - ⁷W. Brouwer and R. K. Pathria, unpublished.
 - ⁸G. N. Antonov, J. Chim. Phys. 5, 372 (1907).
 - ⁹R. Defay, I. Prigogene, A. Bellemans, D. H. Everett,

¹⁰J. C. Wheatley, Am. J. Phys. <u>36</u>, 181 (1968).

¹¹L. M. Milne-Thomson, <u>Theoretical Hydrodynamics</u>

(Macmillan and Co., London, 1960), p. 409. ¹²D. F. Brewer, A. J. Symonds, and A. L. Thomson, Phys. Rev. Letters 15, 182 (1965).

PHYSICAL REVIEW

VOLUME 179, NUMBER 1

5 MARCH 1969

Properties of an Afterglow Helium Plasma*

J. W. Poukey, J. B. Gerardo, and M. A. Gusinow Sandia Laboratories, Albuquerque, New Mexico (Received 1 November 1968)

The role played by individual atomic reactions and transport processes in the decay of a helium afterglow plasma in a long cylindrical vessel is studied by solving numerically a complete set of three-temperature transport equations. Comparison with the measured decay of a laboratory afterglow shows the equations to be realistic. Reactions which are found to be particularly important in determining the decay include electron-ion recombination, metastable formation, metastable-electron and metastable-metastable collisions, resonant charge exchange, and electron-ion collisions. The room-temperatures wall bounding the plasma causes spatial gradients in the densities and temperatures to evolve, resulting in appreciable thermal conduction and particle convection. The relative importance of the various reactions and processes is presented quantitatively. Conclusions are reached which apply at least semiquantitatively to any afterglow helium plasma with densities, temperatures, and geometry comparable to those studied here.

I. INTRODUCTION

The properties of a partially ionized, decaying helium plasma in an infinite cylindrical tube were studied by numerical solution of a set of transport equations. The role played in the decay by individual atomic reactions, transport processes and spatial nonuniformities of constituent parameters was investigated. Gray and Kerr¹ previously studied the spatial and temporal behavior of the electron density in a bounded plasma, but did not allow for spatial nonuniformities other than the free electron and ion densities. Considered here is the more general case in which all plasma quantities are permitted to evolve nonuniformly, and various atomic reactions and transport processes are included explicitly. The relative importance of the role played by each in governing the decay of electron density and temperature was quantitatively studied. The reactions and processes which were given major attention include: electron-ion recombination, metastable-electron and metastable-metastable collisions, thermal

conduction and metastable formation.

The study was carried out for a partially ionized helium plasma with an electron temperature of a few thousand degrees, a background gas pressure of a few Torr, a degree of ionization of < 2% and contained in a long cylindrical tube with a roomtemperature wall. The set of initial conditions was chosen to correspond to those of an extensively investigated laboratory plasma.² This permitted comparison of the theoretical plasma model with the laboratory plasma. This served to illustrate that the model is realistic. The results presented here strictly apply only to the particular plasma which served as the basis for the theoretical model. However, conclusions are drawn from this study which are applicable in part to many laboratory afterglow helium plasmas.

Convective particle flow is commonly estimated in studies of the type carried out here by inclusion of diffusion coefficients and particle gradients. This is a good approximation only if the flux is that of a minority constituent in a spatially uniform background; a condition which is not typical-

Surface Tension and Adsorption (Longmans, Green and Co. Ltd., London, 1966), p. 294.