August 1972

Conf-720833--8

Dielectric Constant and Viscosity of Pressurized
 $\text{He}^3\text{-He}^4$ Solutions Near the Tricritical Point*

C. M. Lai

Brookhaven National Laboratory, Upton, New York

and

State University of New York, Stony Brook

and

T. A. Kitchens

Brookhaven National Laboratory, Upton, New York

Measurements of the dielectric constant ϵ and the product of the normal density ρ_n and the viscosity η have been made on $\text{He}^3\text{-He}^4$ solutions at pressures up to the solidus. In this experiment both $\rho_n \eta$, determined from the damping on a vibrating wire, and ϵ , determined by the frequency shift of a tunnel diode oscillator, can be measured simultaneously in both the He^3 -rich and the He^4 -rich phases below the separation. Data taken for 63.6% to 75.1% He^3 for temperatures between 0.45 and 1.2°K are presented. Viscosity and $\rho_n \eta$ results near the tricritical point, and the resulting phase diagram for elevated pressures obtained from the measured dielectric constant are given.

* Work supported by the U. S. Atomic Energy Commissior.

—NOTICE—

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

JLH

As shown by Graf, Lee, and Reppy⁽¹⁾ the second-order superfluid transition in $\text{He}^3\text{-He}^4$ liquid solution appears to end in the first-order phase separation. In fact, they found the superfluid transition temperature to be a linearly decreasing function of the He^3 concentration and the phase boundaries for both the superfluid He^4 rich and the normal He^3 rich solutions to also be linear. This behaviour indicates that these rather different types of ordering are related, indeed strongly related, at the point of intersection. Griffiths has investigated some of the thermodynamic features of this rather unusual situation and has dubbed this point the tricritical point². Here we report an experiment to investigate the region about this point in which a static property, the dielectric constant, and a dynamic property, the effective viscosity, can be simultaneously measured for $\text{He}^3\text{-He}^4$ liquid solutions under pressure. These measurements will be reduced to determine the general features of the phase diagram and to determine the variation of the viscosity as the transition is approached.

The experimental apparatus is capable of simultaneous measurement of both the dielectric constant and the effective viscosity of both the He^3 and the He^4 rich solutions below the phase separation. The technique and the apparatus has been described previously⁽³⁾ and will be only briefly reviewed here. The sample cell was an integral part of a vapour-pressure-regulated He^3 refrigerator. The cell has symmetrical upper and lower sections each containing a carbon resistance thermometer, a vibrating bent-wire viscometer equivalent to that used by Black, Hall and Thompson⁽⁴⁾, and a low volume capacitor in the extreme upper (lower) position. These capacitors, as well as a third capacitor located in the center of the cell, where there was a large excess volume, were in

the tank circuits of three independent oscillators driven by tunnel diodes located at 4.2°K. These oscillators were stable to one part in 10^7 which is equivalent to less than .05% change in He^3 concentration in these liquid solutions. The temperature of the cell was determined by a calibrated germanium thermometer and could be regulated by a controller using one of carbon resistance thermometers and driving a heater attached to the He^3 refrigerator body.

The 80 micron diameter tungsten wire viscometers, which constituted arms of AC bridges, were in a uniform six kOe magnetic field produced with a persistent superconducting solenoid in the main He^4 bath of the cryostat. The wires were driven to a maximum amplitude of about four microns by current oscillators synchronized with the natural frequency of the wires. The decay or increase in the amplitude of oscillation of each wire is proportional to the error signal of the bridge and, after suitable amplification, was observed on an oscilloscope.

The samples were pressurized by a cryogenic toepler technique; the sample gas was condensed in a container of minimum volume filled with Zeolite cooled to 4°K; the Zeolite container was then warmed to room temperature, thereby pressurizing the sample cell. The He^3 concentration of the gas recovered after the completion of the measurements was determined. Corrections due to the small volumes at high temperatures in the sample system were much less than the uncertainties in the

isotopic gas analyses, which were typically 1%. Comparison of the measured phase separation temperature with that expected from Graf, Lee, and Reppy⁽¹⁾ for saturated-vapor-pressure (svp) were also within 1%.

Analysis of Dielectric Constant Data

From an analysis of the dielectric constant measured in each of the three capacitors it is not only possible to determine the temperature of phase separation for the given He^3 concentration of the sample but also to determine the complete phase diagram for lower temperatures. We have assumed the molar volume of the solution V_s may be expressed by $V_s = xV_3 + (1-x)V_4 - x(1-x)\Delta$ (1) where V_3 and V_4 are the molar volumes of pure He_3 and He_4 , respectively, and are known functions of pressure and temperature⁵. The form of the last term is consistent with measurements at svp by Kerr⁶; from these measurements we know $\Delta = \Delta(p \rightarrow 0, T)$ for T above 1.2°K . It is positive yet decreasing with decreasing temperature. At higher pressures Δ can be determined from the data of Watson, Reppy, and Richardson⁷ for $T \rightarrow 0$. Here Δ is found to be small and positive at low pressures and turning slightly negative at high pressures. We have roughly accounted for Δ in the following analysis with resultant phase separation curve shifts of less than 0.5%.

The data were analyzed assuming that the tank circuit was composed of a capacitor, C_0 , filled with the sample solution and a parallel capacitor C_s due to stray capacitance in the circuit, which is not filled with sample. The dielectric constant of the solution is related to the number densities, N_3 and N_4 , of He^3 and He^4 , respective-

ly through the Clausius-Mossotti Equation

$$(\epsilon-1)/(\epsilon+2) = (4\pi/3)(\alpha_3 N_3 + \alpha_4 N_4)$$

where α_M is the atomic polarization for the isotope with atomic mass M. Our analysis follows that of previous work ^{7,5} except we have accounted for the fact that α_3 and α_4 are not equal and depend on the density as recently shown by Kerr and Sherman⁸. Here we have used $\alpha_3(\text{mole/cm}^3) = 0.12341 - 0.0068/V_s$ and α_4 0.03% higher.

The analysis⁷ contains two parameters f_0 and $\gamma = C_0/(C_0 + C_s)$ for each of the oscillator circuits. f_0 is the frequency when the sample capacitor is void of sample and γ can be determined from the frequency measured above 1.2°K and the known ³He concentration of the sample. The results of such an analysis on a run at svp is shown in Fig. 1 and the agreement with Graf, Lee and Reppy⁽¹⁾ is excellent. A compilation of results at other pressures agrees with those obtained by Johnson and Graf.⁽⁹⁾

Analysis of Viscosity Data

From an analysis of the decay in the amplitude of vibration of the bent-wire viscometers, the product of the normal density and the viscosity $\rho_n \eta$ can be obtained near the top and bottom of the cell provided (1) the vibrational amplitude is a small fraction of the wire diameter a , (2) the viscous penetration depth $\lambda \ll a$, and (3) the nonlinear terms in the fluid velocity are negligible in the Navier-Stokes Equation.^(10,11) Under these assumptions, well satisfied in this investigation,

$$\rho_n \eta = \frac{1}{4\pi f} \left(\frac{\rho_w \cdot a}{\tau} \right)^2$$

within .5% where τ is the decay time constant, ρ_w is the wire density, and f is the frequency of the oscillation.

In addition to a measure of $\rho_n \eta$ as the phase separation is approached and for both the He^3 -rich and the He^4 -rich phases below the phase separation, it is possible to determine the superfluid transition $T_\lambda = T_\lambda(P, x)$ and $\rho_n \eta$ near T_λ for an initial He^3 concentration x less than the tricritical value x_{tc} . Such data are illustrated in Fig. 2.

Finally, the viscosity in the normal fluid has been calculated from these data near the tricritical point. The data illustrated qualitatively in Fig. 3 are not precise enough to obtain accurately the exponent derived from the scaling hypothesis but it appears that it is rather more sensitive to concentration than to pressure. However, the curves corresponding to 67% He^3 solution at svp and 65% He^3 solution at 22.9 atmospheres terminate very near the tricritical point yet have quite different slopes.

We wish to acknowledge many helpful conversations with M. Vanneste, D. F. Brewer, and P. P. Craig, and the technical support rendered by T. Oversluizen.

References

1. E. H. Graf, D. M. Lee, and J. D. Reppy, Phys. Rev. Letters 19, 417 (1967).
2. R. B. Griffiths, Phys. Rev. Letters 24, 715 (1970).
3. C. M. Lai, P. P. Craig, T. A. Kitchens and D. F. Brewer, Proceed. 12th Internat. Conf. Low Temp. Physics, Kyoto, Japan, Sept. 1970, Ed. E. Kanda, (Academic Press, 1971) p. 167.
4. M. A. Black, H. E. Hall, and K. Thompson, J. Phys. C: Solid State Physics 4, 129 (1971).
5. C. Boghosian, H. Meyer, and J. Rives, Phys. Rev. 146, 110 (1966); C. Boghosian and H. Meyer (private communication).
6. E. C. Kerr, Low Temperature Physics and Chemistry, editor Dillinger (Univ. of Wisconsin Press, 1958), p 158.
7. G. E. Watson, J. D. Reppy and R. C. Richardson, Phys. Rev. 188 384 (1969).
8. E. C. Kerr and R. H. Sherman, Journal of Low Temp. Physics 3, 451 (1970).
9. R. Johnson and E. Graf, (private communication.)
10. J. T. Tough, W. D. McCormick, and J. G. Dash, Rev. Sci. Inst. 35, 1345 (1964); Phys. Rev. 132, 2373 (1963).
11. R. G. Hussey, B. J. Good, and J. M. Reynolds, Phys. Fluids 10, 89 (1967); R. G. Hussey and P. Vujacic, Phys. Fluids 10, 96 (1967).

Figure Captions

- Fig. 1. The phase diagram determined from dielectric constant data from the three tunnel diode oscillators in the experimental cell. The initial solution was 67% He^3 in He^4 at saturated vapor pressure.
- Fig. 2. The product $\rho_n \eta$ for the two bent-wire viscometers for an initial solution of 67% He^3 at saturated vapor pressure.
- Fig. 3. The qualitative features of the viscosity in the neighborhood of the tricritical point. The initial concentration of He^3 and the pressure in atmospheres label each curve. The terminal points indicate phase separation and the intermediate point in the $x < x_{tc}$ curves designate the superfluid transition.

FIGURE 1

FIGURE 2

FIGURE 3