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SELF-DIFFUSION COEFFICIENT AND NUCLEAR SUSCEPTIBILITY OF LIQUID He³†

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The self-diffusion of the magnetization has been measured at pressures ranging from 5 to 9 cm-Hg in pure liquid He³ (He⁴ impurity less than 2:10⁵) and in liquid He³ containing 1.2% He⁴ from 0.15°K to 0.032°K.¹ The nuclear susceptibility relative to its value at the He⁴ bath temperature has been measured at similar pressures in pure He³ from 1.3°K to 0.031°K, and in He³ containing 1.2% He⁴ at 1.3°K and at a few low temperatures, the lowest being 0.032°K. No evidence has been found for a transition of the liquid into some sort of cooperative state.²

The diffusion coefficient, D , was measured using a spin echo method at a frequency of 85 kc/sec. A 90°-180°-180° pulse sequence was used with a fixed time delay between the 90° pulse and the first 180° pulse and a variable time, τ , between the two echoes. The resulting echoes were displayed on an oscilloscope and photographed. The ratio, R , of the amplitudes of the two echoes is $R = \exp(-\tau/T_2 - \gamma^2 G^2 D \tau^3 / 12)$, where G is the magnetic field gradient, $\gamma = 2.038 \times 10^4$ (gauss-sec)⁻¹, and T_2 is the transverse relaxation time.³ The diffusion coefficient was calculated neglecting the effect of T_2 . It was found that T_2 was greater than 0.74 sec at $T = 0.034$ °K. This magnitude for T_2 would lead to a +2.2% error in D at the lowest temperatures. The cell containing the He³ was a cylinder 0.5 cm i.d. \times 0.8 cm long. The copper cooling wires were embedded in the cylindrical surface and did not penetrate into the space occupied by He³; hence, it is believed that the measured D is characteristic of the bulk liquid. Measurements of D were made as a function of G and with G alternately

parallel and antiparallel to the steady field. Convection effects were assumed to be absent. The apparatus outside the cryostat and the method used to obtain the echoes were essentially the same as described by Hart and Wheatley.⁴

The relative nuclear susceptibility, χ_γ , was measured using the apparatus described above but with no applied gradient and with one 180° pulse applied just a few milliseconds after the 90° pulse. The amplitude of the echo was taken to be proportional to the susceptibility provided the magnetizing time was long enough to allow the thermal equilibrium magnetization to develop. All echo amplitudes were divided by those measured at the He⁴ bath temperature at the end of a day's measurements to obtain a quantity proportional to χ_γ . As the temperature decreased, the amplitude became dependent on the time after the 90° pulse. The temperature-dependent gradient responsible for this defect in the experiment probably came from a weak magnetism of the "Epibond 104"⁵ used to construct the He³ cell. The time dependence was extrapolated out assuming that it arose from diffusion. However, the resulting values of χ_γ , which have been increased by the extrapolation from the measured amplitudes by about 4% at 0.03°K, may still be as much as 7% too small at 0.03°K since it is not clear empirically how the extrapolation should be made. The correction was less than 1% for $T > 0.065$ °K.

Temperatures were measured using a slurry of cerium magnesium nitrate and Dow-Corning 200 Fluid of 50 cs viscosity encased in a shell of Epibond 104. At the conclusion of the meas-

urements of D and χ_T it was shown that Epibond 104 was weakly magnetic. Since this material had been used rather extensively in construction, it was necessary to make very careful magnetic temperature calibrations of all parts of the apparatus. In the 1-4°K region, 27% of the Curie constant of the thermometer came from Epibond 104. A special experiment was made to determine the T - T^* curve for Epibond 104 using cerium magnesium nitrate as thermometer.⁶ The presence of the Epibond constitutes a serious defect in the precision of the experiment; however, the measured temperatures below 0.15°K are less susceptible to error than those at higher temperatures where a thermal shield containing Epibond varied in temperature in a relatively unknown way.

The results of the diffusion measurements are shown in Fig. 1 and those for the relative susceptibility measurements are shown in Fig. 2. These results may be summarized as follows: (i) Adding 1.2% He⁴ to pure He³ does not change either D or χ_T within our experimental error. (ii) Between 0.1°K and 0.03°K, D obeys a power law $(1/T)^n$, where $n = 1.55 \pm 0.05$ for pure He³ and $n = 1.56 \pm 0.05$ for He³ with 1.2% He⁴. In reference

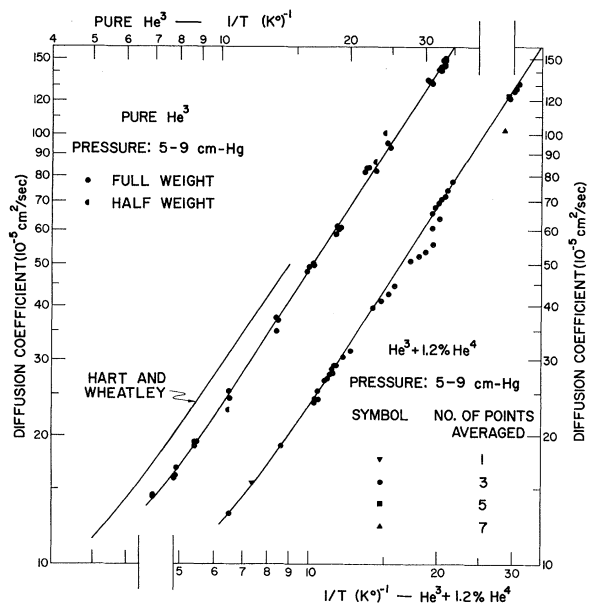


FIG. 1. Plot of the logarithm of the diffusion coefficient against the logarithm of the reciprocal of the temperature for He³ containing less than 2 parts in 10⁵ of He⁴ and for He³ containing 1.2% He⁴.¹ The curve labeled "Hart and Wheatley" represents smoothed data from reference 4. The curve drawn through the data for He³ with 1.2% He⁴ is the one having maximum slope.

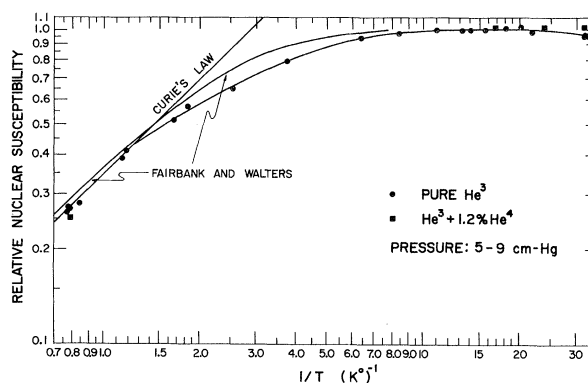


FIG. 2. Plot of the logarithm of the relative nuclear susceptibility against the logarithm of the reciprocal of the temperature for He³ containing less than 2 parts in 10⁵ of He⁴ and for He³ containing 1.2% He⁴. The relative nuclear susceptibility is the ratio of the susceptibility at temperature T to that below 0.1°K. The curve labeled "Fairbank and Walters" represents smoothed data from reference 10. The straight line labeled "Curie's Law" is the curve the relative susceptibility would follow if He³ obeyed Curie's law. The values of relative susceptibility have been corrected for changes in the density of He³ with temperature.

4, it was found that $n = 1.46 \pm 0.1$ in a more restricted temperature range with a 1.1% He⁴ impurity and with a He³ cell in which diffusion could have been limited somewhat by the boundaries. (iii) Between 0.1°K and 0.04°K, χ_T is constant within an experimental error of about 3%. At 0.03°K, χ_T appears to be a few percent lower, but this effect is thought to be spurious. (iv) Deviations at higher temperatures of χ_T from constancy occur in the same temperature range as deviations of D from a strict power-law dependence on temperature. (v) The ratio of the susceptibility at 1.293°K to that below 0.1°K is 0.268 ± 0.008 . If He³ obeyed Curie's law at all temperatures, the susceptibility below 0.1°K would correspond to a temperature of (0.347 ± 0.010) °K. (vi) No evidence has been found down to temperatures of 0.03°K either in D or in χ_T for a transition to some sort of cooperative state.²

Smoothed data from reference 4 are shown on Fig. 1. At low temperatures the curves are nearly parallel but shifted by about 14% from one another. This shift probably arises from a systematic error in calibration of the thermometer in the earlier experiment due to neglect of the weakly magnetic properties of some of the materials used in construction. The measure-

ment of the temperature dependence in the present experiment is marred by the necessity of accounting for the deviations of Epibond 104 from Curie's law. In the present experiment the ferric alum refrigerator, the thermometer, and the He³ cell were completely different from those in reference 4. Since the same temperature dependence of D was observed in both experiments, within experimental error, it seems unlikely that serious systematic errors in the temperature dependence are present.

The straight line drawn through the experimental values of D for the He³ containing 1.2% He⁴ has the maximum reasonable slope. Experimental points to the right of this line were obtained on days when the warmup rate was much larger than normal, with the exception of an unexplainable point at $1/T = 28.7^\circ\text{K}^{-1}$, $D = 102 \times 10^{-5}$ cm²/sec. Hence the effect of the heat leak is to decrease the value of n in the law $D \propto (1/T)^n$. Similar effects were not observed in the measurements with pure He³. Approximately three hours after demagnetization D reached a constant value characteristic of 0.03°K .

The ratio of the susceptibility at the He⁴ bath temperature (corrected for variations in this temperature and for density⁷) to that below 0.1°K had a maximum spread of 3% for three measuring days and was independent of the calibration of the magnetic thermometer. Combining the measured value of the specific heat⁸ with the above ratio, one finds that the ratio $\chi T/C_v$, where C_v is the specific heat and χ is the susceptibility below 0.1°K , is 4.8 ± 0.4 times larger than it would be in the absence of spin-dependent interactions.⁹

Values of χ_T for pressures near the saturated vapor pressure have been calculated from the graphs given by Fairbank and Walters¹⁰ and plotted on Fig. 2, χ_T arbitrarily being set equal to 1.0 at their lowest temperature, 0.13°K . If their curve is normalized to agree with ours at the bath temperature, it is found that their extrapolated χ_T agrees with the value measured in this experiment to within 2%. This agreement is within the experimental error of both experiments, which differ from one another quite considerably in details. Between $(1/T) = 2$ (K^o)⁻¹ and $(1/T) = 6$ (K^o)⁻¹, again normalizing Fairbank and Walters' curve to agree with ours at the bath temperature, the present values of χ_T are as much as 8% lower than those of Fairbank and Walters. However, in this temperature range the errors in thermometry due to the presence of Epibond 104 in a thermal shield are difficult

to assess and may be as large as 5 to 10%. Hence the deviations from the data of Fairbank and Walters may not be significant.

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¹The 1.2% He⁴ impurity is in the gas. It is possible that in the temperature range of our experiments there is a phase separation which changes the concentration of He⁴ in the He³-rich phase. K. N. Zinov'eva and V. P. Peshkov, J. Exptl. Theoret. Phys. (U.S.S.R.) 37, 33 (1959) [translation: Soviet Phys.-JETP 10, 22 (1960)].

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⁵Furane Plastics, Inc., 4516 Brazil Street, Los Angeles 39, California.

⁶"Epibond 104" has a susceptibility per gram in the region from 1°K to 2°K which is 31% of that for cerium magnesium nitrate. In the region of 0.04°K , $T^* \sim 2T$. The magnetism seems to come from the Bentonite filling material and hence probably arises from some impurity. Complete details of the T - T^* experiment will be supplied on request.

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¹⁰W. M. Fairbank and G. K. Walters, Proceedings of the Symposium on Solid and Liquid He³ (Ohio State University, Columbus, 1957), p. 211.