

Measurement of mass diffusion in nonsuperfluid $\text{He}^3\text{-He}^4$ solutions

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Mass diffusion in nonsuperfluid $\text{He}^3\text{-He}^4$ solutions was measured by the crystal dissolution method. The diffusion coefficient in the temperature range 0.55-1.05°K at a pressure 26 atm, and for He^3 concentrations 60-100%, lies in the range $(1.7 \text{ to } 2.5) \times 10^{-5} \text{ cm}^2/\text{sec}$, which is smaller by an approximate factor than the spin diffusion coefficient for the same solutions.

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$\text{He}^3\text{-He}^4$ solutions with uniform temperature and pressure have two diffusion coefficients. The spin diffusion coefficient characterizes the spin flux in the presence of a gradient of spin-concentration (i.e., magnetization, and is measured by the spin-echo method. The mass-diffusion coefficient connects the mass flux with the isotope-concentration gradient that produces this flux. The two coefficients are in general not equal and the question of their ratio has not been considered theoretically.

In the present study, we measured the mass diffusion coefficient in non-superfluid $\text{He}^3\text{-He}^4$ solutions with high He^3 concentration. The spin diffusion coefficient for such mixtures was measured in detail by Harrison and Hatton, as well as by Fenner and Luszczyński.^{1,2]}

The mass diffusion was measured in the instrument illustrated in Fig. 1. The experimental mixture, cooled to 0.1°K with the aid of a dissolution cryostat, was stratified in such a way that the phase boundary in the ampoule was on the "tail" of the instrument inside the nuclear magnetic resonance (NMR)

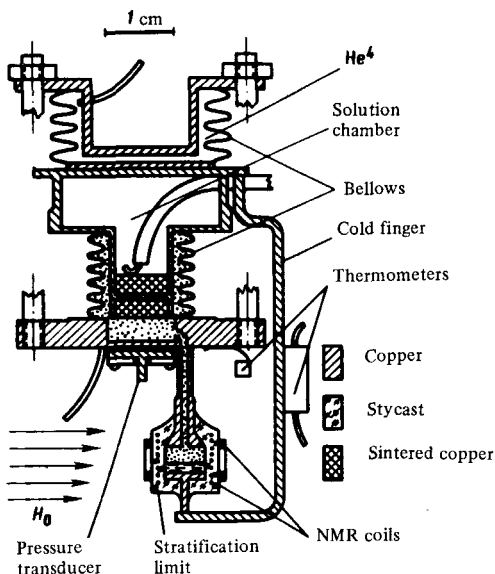


FIG. 1. Diagram of instrument.

ils. The chamber was then compressed with the aid of high-pressure He^4 and o bellows, and the lower phase was crystallized. The crystal growth was om the lower part of the ampoule, from the coldest place, and continued for hour—this resulted in a very level phase boundary. The ampoule was in a agnetic field with a gradient ($\sim 30 \text{ G/cm}$) directed perpendicular to the inter- se. The position of the interface, or more accurately the thickness of the uid layer, was determined with approximate accuracy 20μ during the passe of the NMR signal. We used continuous NMR at a frequency 2.5 MHz with system of crossed coil and with a passage time 25 sec .

At 0.1°K and 25 atm , the solid He^4 at the bottom was in equilibrium with the re liquid He^3 above. The system was then rapidly heated to a temperature at ich the He^4 concentration in the upper phase should increase, thus causing a ocess of diffusion establishment of equilibrium (the processes in the solid ase are much slower). The heating could be carried out in steps. The time thermal relaxation in the instrument was 300 sec at 0.1°K and was several es shorter at temperatures above 0.5°K , i. e., much less than the diffusion ie. The transition of the upper phase to the new concentration was accom- nished by dissolution of solid He^4 with a corresponding motion of the interface. e concentration equilibrium was established on the interface very rapidly. is follows from the fact that when convective instability is produced in the amber the interface arrives at the new equilibrium position within a time ch shorter than the diffusion time. Therefore the process can be regarded taking place at constant temperature, and the concentration of the liquid at boundary with the crystal is constant and equal to the equilibrium value. e influence of the upper part of the instrument can be neglected, since the poule is connected to it with a capillary having a diameter one-sixth as ge and ten times longer than the liquid layer in the ampoule.

We measured the displacement of the phase boundary as a function of the

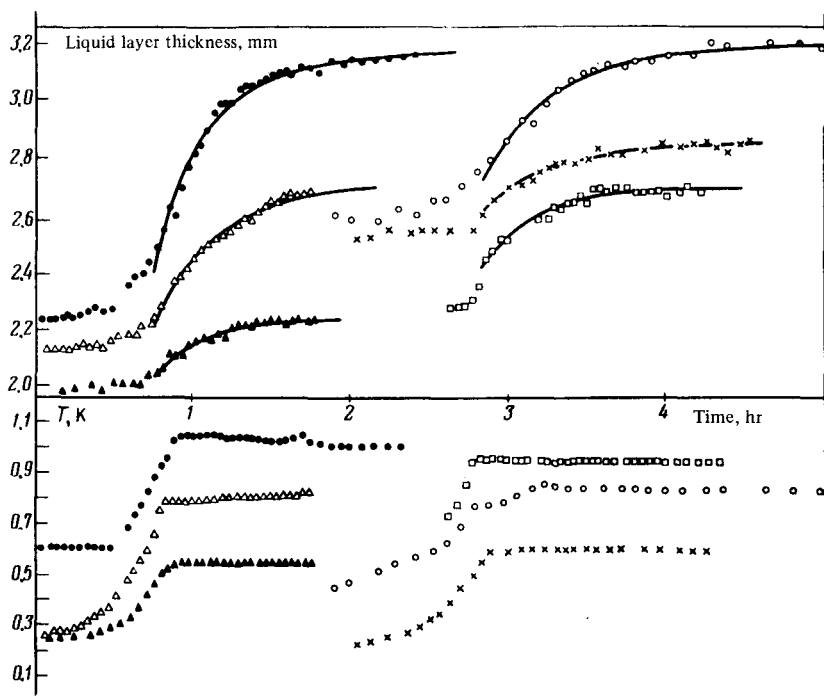


FIG. 2. Measurement of the thickness of the liquid layer and of the temperature in the course of six experiments. Curves—solutions of the corresponding diffusion problems.

time. The results of six experiments are shown in Fig. 2, and the time reference for each experiment is chosen arbitrarily. The lower part of the figure shows the variation of the temperature of the chamber in each experiment. The temperature was measured with a carbon thermometer mounted on the chamber wall and calibrated against the He^4 and He^3 vapor pressures and the He^3 melting curve.

The motion of the boundary is described by the solution of the Stefan problem for a homogeneous diffusion equation

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} \quad (1)$$

with mobile boundary, and with boundary conditions

$$\frac{\partial c}{\partial x} = 0 \quad \text{at} \quad x = 0; \quad \frac{\partial c}{\partial x} = \frac{\Delta c}{D} \dot{y}, \quad c = c_p \quad \text{at} \quad x = y \quad (2)$$

and initial condition

$$c = 0 \quad \text{for} \quad 0 \leq x < y. \quad (3)$$

Here $c = c(x, t)$ is the concentration of the He^4 in the liquid, D is the diffusion coefficient, c_p and Δc are the equilibrium concentration on the boundary and

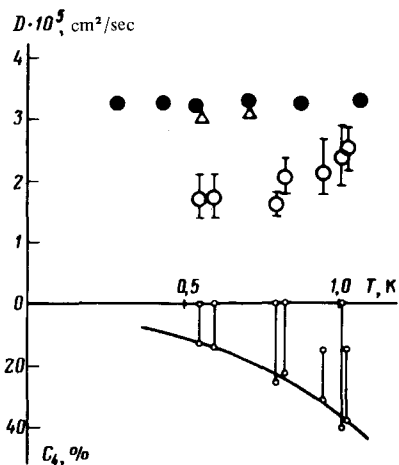


FIG. 3. Dependence of the diffusion coefficient on the temperature, and also the initial and final concentrations of the liquid in each experiment. Mass diffusion: circles with error bars—present work at 26 atm; spin diffusion: circles—85% He^3 , 20 atm,^[1] triangles—100% He^3 , 26 atm.^[4] He^4 concentration in liquid: small circles—present work at 26 atm, solid curve—data on crystallization of solutions at 26 atm.^[5]

e difference between the volume concentrations of the liquid and the crystal, and y is the position of the boundary.

In view of the special boundary conditions (2), this problem is nonlinear and can be reduced to an infinite system of ordinary differential equations.^[3] However, by excluding the initial section (20% of the displacement), the motion of the boundary can be described by the first approximation with 2% accuracy. This was verified by numerically solving the system of equations up to fifth order. The corresponding curves are shown in Fig. 2 for each experiment. The least squares method was used to determine the mass diffusion coefficient D , which is a parameter of the curve. The results are shown in Fig. 3. The segments mark the region of D for which the rms error increases by not more than $\sqrt{2}$ times. The same figure shows the results of the measurement of the spin diffusion for an He^3 concentration 85% and a pressure 20 atm,^[1] as well as the data of Garwin and Reich^[4] for pure He^3 at 26 atm. Figure 3 shows the initial and final concentrations C_4 of the liquid in each experiment. The initial concentration of the liquid was determined from the displacement of the boundary to a new equilibrium position, by the formula

$$C_4 = \frac{\Delta l / V_4}{\frac{\Delta l}{V_4} + \frac{l}{V_3}} = \frac{1}{1 + \frac{l}{\Delta l} \frac{V_4}{V_3}}. \quad (4)$$

where l is the initial thickness of the liquid layer, Δl is the displacement of the boundary, and V_4 and V_3 are the molar volumes of the solid He^4 and the liquid He^3 . The calculated concentrations agree well with the data of Zinov'eva^[5] on the crystallization of solutions.

The obtained values of the coefficient of mass diffusion turned out to be less than for the spin diffusion by approximately 1.5 times. For superfluid mixtures, it follows from the available data by Ptukha^[6] on the mass diffusion and

the data^[4,7] on the spin diffusion that the mass diffusion coefficient is larger than or equal to the spin diffusion coefficient. In addition to the opposite dependence of the spin diffusion on the He^3 concentration in superfluid and nonsuperfluid mixtures, which was indicated in^[1], this shows that the diffusion process is essentially different in normal and superfluid He^3 — He^4 mixtures.

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