

He-II Film Transfer Rates for Solid-Ar Beakers*

T. O. Milbrodt[†] and Gerald L. Pollack

Physics Department, Michigan State University, East Lansing, Michigan 48823

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Helium-II film transfer rates have been measured for filling and for emptying beakers of solid Ar. Liquid Ar was frozen slowly in a mold which was later removed, leaving free-standing transparent solid-Ar beakers. The beakers used were of 3-mm i.d., 5-mm o.d., and 3.5 cm in height. Experiments were carried out in the temperature range 1.45–2 K. The dependence of the transfer rate σ ($\text{cm}^3/\text{s cm}$ of circumference) on the difference between inner and outer He levels Z , height of the beaker lip above the level of the liquid source H , and temperature was studied. The transfer rates were lower than those observed with glass beakers, in agreement with theory. For beaker fillings with a level difference of 1 mm and at a temperature of 1.66 K, the measured transfer rates may be described by the relation $\sigma = 4.8 H^{-0.21} \times 10^{-5} \text{ cm}^2/\text{s}$, where H is in cm.

INTRODUCTION

The He-II film transfer effect depends on the existence of a thin He film adsorbed on cold surfaces in contact with the He liquid or vapor. This film is typically only 100–300 Å thick, but the superfluid component of He II can nevertheless flow in it quite readily. Thus if an empty beaker is partially submerged in He II, the superfluid will flow through the film, and the beaker will fill until the He levels inside and outside the beaker are equal.

Most measurements of the transfer effect have been made with beakers of glass or metal. The van der Waals attractive potential between He and glass is not very different from that between He and a metal, so that the He film thickness and the transfer rate are relatively independent of which of these materials is used for the beaker walls. Recent calculations of the van der Waals potential between He and an Ar substrate¹ have shown, however, that the He film on Ar should be about 35% thinner than it is on glass, resulting in measurably lower transfer rates. The adsorbed He film is thinner on an Ar surface because the He to Ar attractive force is weaker than the attractive force between He and glass or metal.

In this paper we present the results of measurements of He film transfer rates made using solid Ar beakers. We also describe the techniques used in making the beakers and collecting and analyzing the data, as well as the theory with which the results are compared.

APPARATUS AND PROCEDURE

A cross section of the apparatus that we used to make the argon beakers and to conduct the film transfer experiments is shown in Fig. 1. The walls of the chamber in which the experiments were carried out are made of glass, with Kovar

seals at the ends. The Kovar is soft-soldered into brass flanges. Screws hold the bottom of the chamber to the lower flange and the joint is sealed with an In O ring. The upper flange is soldered to a thin-walled stainless-steel tube, which supports the chamber. This chamber is surrounded by an annular exchange-gas region. The outer wall of this region is similarly made from two glass-to-Kovar seals, but it has demountable flanges using Pb O rings at both ends. The annular space between the chambers can be evacuated or filled with any desired pressure of exchange gas to aid in temperature control while making a beaker. A needle valve is mounted on top of the topmost flange and is connected by a small stainless-steel tube to the experimental chamber. This valve is used to admit liquid He to the chamber from the surrounding bath. The apparatus is contained in a standard glass double-Dewar system that permits evacuation of the inner Dewar and has unsilvered viewing slits.

The upper part of the mold in which the Ar beakers were formed consists of two glass tubes. The outer tube is attached at its top to a Kovar seal. The Kovar is soldered into a brass collar, which serves as a spacer, and slides in the stainless-steel tube. The inner tube forms the bore of the beaker. It is sealed at its lower end, and there is a loop of heater wire in it. It is supported in the brass collar and also, by a Teflon spacer, in the outer tube. A thin-walled stainless-steel tube is soldered into the brass collar at the top of the mold. It slides through a rubber O ring at the top of the cryostat (at room temperature), and it is used to move the upper part of the mold up and down as well as to admit Ar gas to the inside of the mold.

The base of the mold is made of Teflon. It has a conical seat with half-angle 5° into which the upper

part of the mold is pressed to form a seal. The lower, sealing edge of the mold tube must be ground flat to make the seal gas tight. The hollow spaces machined in the base fill with solid Ar . This helps to support the beaker and to hold the beaker down when the upper part of the mold is pulled away. A threaded post on the bottom of the Teflon base screws into a hole in the brass block beneath it. This holds the base in place. The base is slightly loose in the hole so that it can move a bit as the mold is fitted together. This makes it easier to obtain a good seal. The bottom of the base is coated with vacuum grease to improve thermal contact with the brass block.

The Ar gas pressure in the mold was controlled

manually by admitting or removing Ar gas through an external needle valve. A Wallace and Tiernan gauge was used to monitor the pressure. We used Matheson ultrahigh purity grade Ar gas (purity 99.999%) to make all of our beakers.

The temperature of the mold was also controlled manually, with two electric heaters, one on the brass block beneath the mold and one on the brass collar at the top of the mold. Two miniature platinum resistance thermometers were used to monitor the temperatures at these same locations.

In order to make a gas-tight seal at the bottom of the mold, the glass tube must be pressed into the Teflon seat while the apparatus is at room temperature. Upon cooling, the Teflon, which has

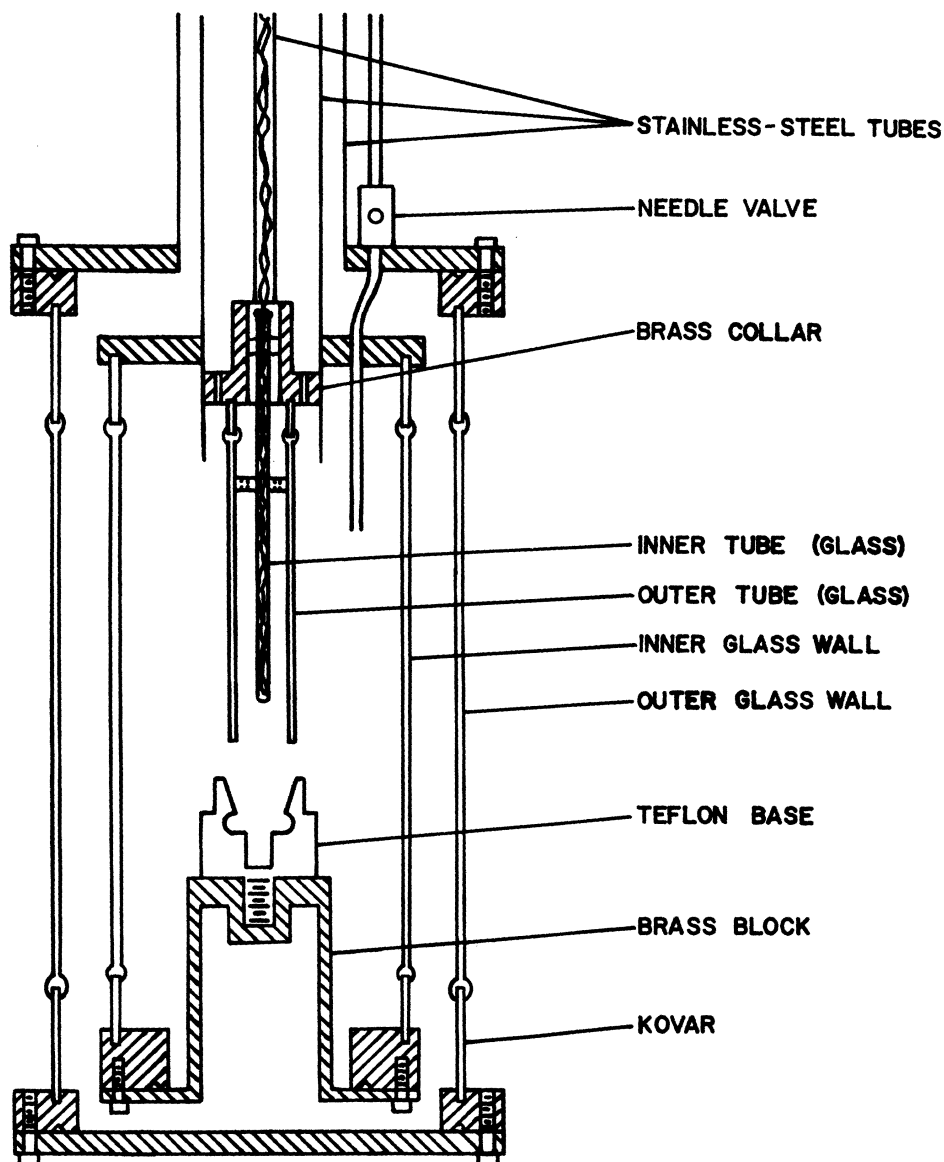


FIG. 1. A sketch of the apparatus. The upper part of the mold may be raised and lowered from outside the cryostat.

a larger coefficient of thermal expansion, contracts tightly around the glass mold tube. The apparatus is cooled with liquid nitrogen and the heaters adjusted so that the temperature of the mold is brought to about 0.5 K above the Ar triple-point temperature of 83.8 K. Argon gas is then admitted to the mold until a sufficient amount of liquid Ar has condensed in it. A small reduction in the electric current in the lower heater causes the temperature of the bottom of the mold to slowly fall to slightly below the triple-point temperature, and the Ar then begins to freeze from the bottom. The chamber around the mold is kept evacuated to about 10^{-6} Torr while the Ar is freezing so that the heat of fusion is carried away through the solid Ar and the mold. This keeps the freezing rate slow and even. After a few mm of solid have formed, a further reduction of the temperature of the bottom of the mold is begun until, after several hours, it has fallen to 78 K. It takes about 12 h for all the liquid Ar to freeze under these conditions.

After the last of the Ar has frozen, the temperature of the top of the beaker begins to fall below the triple-point temperature and solid Ar begins

to condense on top of the beaker from the Ar gas remaining in the mold. In order to prevent this, and the associated unevenness of the beaker top, Ar must be slowly pumped from the mold as the sublimation pressure of Ar corresponding to the temperature of the top of the beaker falls from the triple-point value of 517 to 220 Torr at 78 K. A small amount of He exchange gas is added to the chamber to help bring the solid Ar to a uniform temperature.

When the temperature gradients in the solid have been eliminated, we resume slowly pumping Ar gas from the mold. The Ar pressure in the mold is reduced to about 0.5 Torr over a period of about an hour. The resulting sublimation of some of the solid Ar cools the remaining solid and frees the Ar beaker from the mold. A little current in the heater in the glass tube in the middle of the beaker ensures that some solid Ar sublimates from it. When the beaker is loose, the upper part of the mold is pulled up so that the beaker is left standing free on the Teflon base of the mold. Liquid He is then transferred into the cryostat, cooling the beaker to 4.2 K.

Our beakers are typically 3.5 cm in height, with 0.55-cm o.d., 0.30-cm i.d., and 2.5-cm bore length. The Ar is transparent, but usually has a few visible defects. Quite often one of these defects will penetrate the wall of a beaker, allowing it to leak He I and rendering it useless for transfer experiments. Figure 2 is a photograph of one of our beakers. We have stored beakers for over a week at 4.2 K. It is necessary to protect the beaker from contaminating vapors with a liquid-nitrogen-cooled cold trap at all times.

A beaker-filling run is initiated by admitting He from the bath through the needle valve into the chamber until the liquid level in the chamber is about 1 cm higher than it is inside the beaker. A beaker-emptying run is initiated by using the stream from a He II fountain in the chamber to fill the beaker. We observe the liquid level in the beaker with a Wild cathetometer and record the level height every 30 s. After a series of beaker fillings and emptyings, the liquid He is pumped from the chamber with a mechanical vacuum pump. This pumping is done through a series of two liquid-nitrogen-cooled cold traps.

Figure 3 is a plot of the liquid level inside the beaker as a function of time for three filling runs. The ordinate Z is the difference between the inner and outer levels in cm. Since the volume of the chamber is much greater than that of the beaker, the outer level may be taken to remain constant during a run. The transfer rate σ ($\text{cm}^3/\text{s cm}$ of circumference) is proportional to the slope of the $Z(t)$ curve.

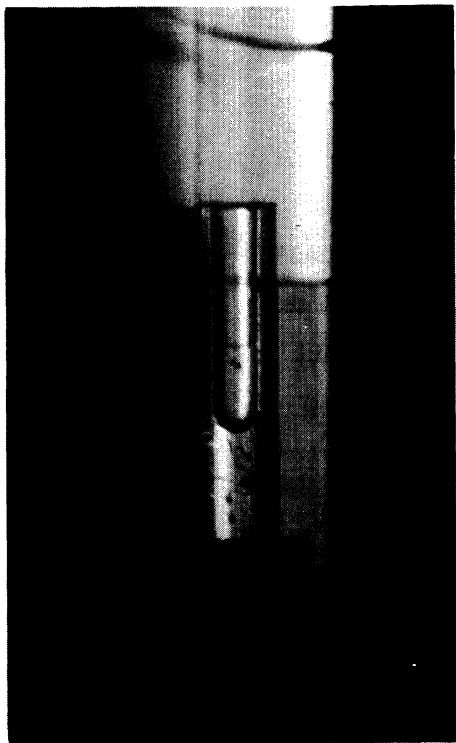


FIG. 2. Photograph of one of the Ar beakers. The beaker is filling, and the liquid He levels inside and outside the beaker can be seen. Some defects are visible in the solid Ar, especially below the end of the bore.

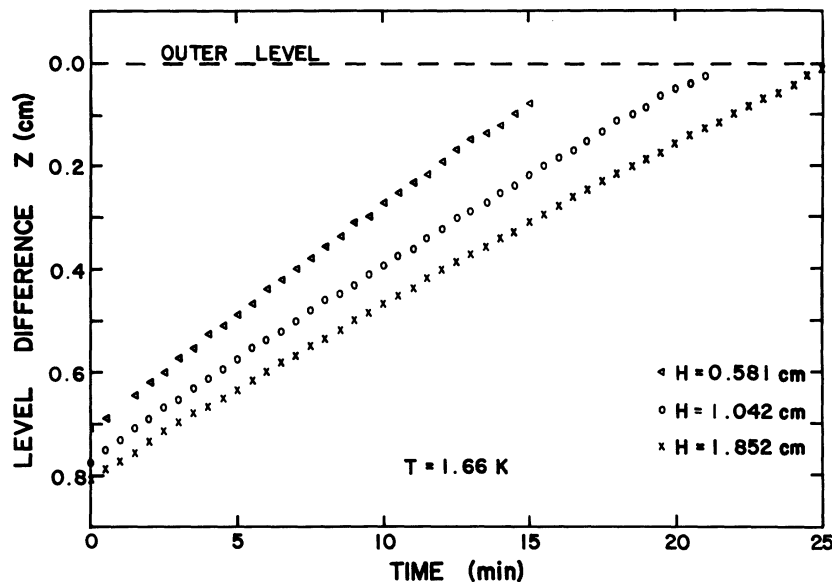


FIG. 3. He level difference as a function of time for three beaker fillings at three different heights H from the outer He level to the beaker rim.

Differentiating the $Z(t)$ data directly results in a great deal of scatter because of random errors in the height measurements. The scatter is reduced by using the following smoothing procedure: A line $Z(t) = at + b$ is fit to each set of eight adjacent data points. The slope of the line is used to calculate the transfer rate associated with the center of the smoothing interval. The geometric relation between the transfer rate and Z is just

$$\sigma = \frac{\gamma}{2} \frac{dZ}{dt}, \quad (1)$$

where γ is the inner radius of the beaker. There is a slight flare in the i.d. near the lip, so that

the i.d. at the lip is usually about 7% greater than γ . The flare is never more than 2 mm long and should have little effect when the liquid He levels are not very near the lip. The radius γ was measured to within 3% with a horizontal cathetometer. Corrections were made for the index of refraction of solid Ar and for the independently measured optical effects of the glass Dewar system.

The transfer rates corresponding to the data shown in Fig. 3 are plotted in Fig. 4.

THEORY

We relate the transfer rate to the experimental parameters in the usual way with the equation²

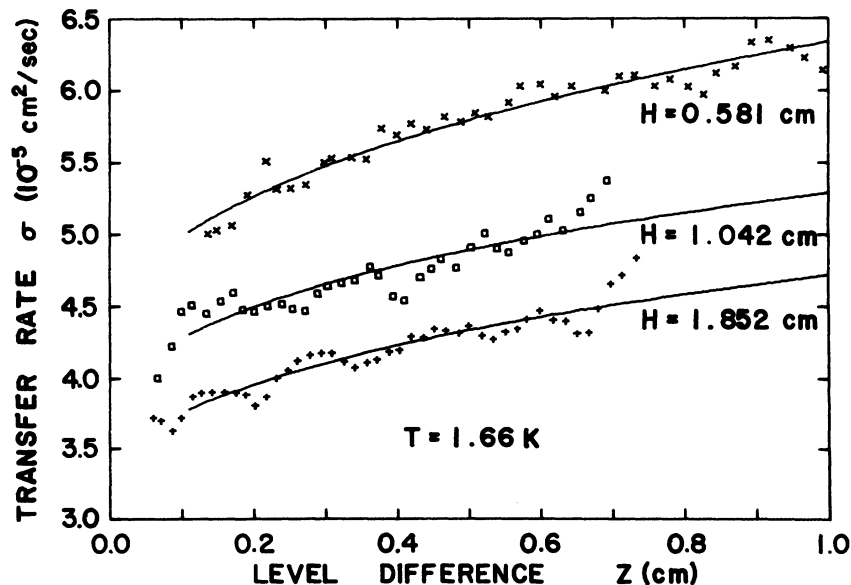


FIG. 4. Transfer rate as a function of level difference for the three beaker fillings of Fig. 3. The solid curves are best fits of the form $\sigma = 1/(A - B \ln Z)$ to the data.

$$\sigma = (\rho_s/\rho)v_s d. \quad (2)$$

In Eq. (2), d is the thickness of the adsorbed He film at the lip of the beaker, v_s is the maximum superfluid velocity in the film, which occurs near the lip, and ρ_s/ρ is the superfluid fraction.

For the He flow in channels of width less than 10^{-3} cm, many experiments have given results that are represented well by the expression³

$$v_{s,c} \approx d^{-1/4} \quad (\text{cgs units}), \quad (3)$$

where $v_{s,c}$ is the critical superfluid velocity, the superfluid velocity under a vanishingly small driving pressure. In particular, when coupled with independent measurements of the saturated He film thickness on glass⁴ and Eq. (2), this relation has successfully described He film transfer rates observed in a number of careful experiments with glass beakers.^{5,6}

As can be seen from Fig. 4, our data show a weak dependence of σ on the level difference Z . This is a special case of a general dependence of v_s on driving chemical-potential difference that has been observed and studied in film flow⁶⁻⁸ as well as other cases of He II superfluid flow.^{9,10} It has also been treated theoretically.^{11,12}

The steady-state relation for superfluid flow driven by a chemical-potential gradient $\nabla\mu$ is of the form

$$-\nabla\mu = C e^{b(T)[1-v_0(T)/v_s]}. \quad (4)$$

The parameters C , $b(T)$, and $v_0(T)$ are constants with a weak temperature dependence. Since $\nabla\mu \approx \rho g Z/L$, where L is some length along the direction of flow in the film we have $\nabla\mu \propto Z$, approximately, if the length L does not change. In agreement with this we make a two-parameter fit with the data from each filling run to

$$\sigma = 1/(A - B \ln Z). \quad (5)$$

The solid curves in Fig. 4 are fitted curves of this form.

The He film thickness on the beaker wall at a height h above the bulk liquid may be calculated by applying the condition that the potential energy of a He atom on the surface of the film must equal the potential energy of an atom on the surface of the bulk liquid.^{13,14} In order to calculate the He film thickness, therefore, one needs to know the attractive van der Waals potential $V(d)$ between a He atom and an Ar substrate a distance d away.

The London theory¹⁵ has usually been used to determine the form of $V(d)$. It is based on the instantaneous dipole moment that a neutral atom or molecule has and which enables it to interact with neighboring atoms to produce an attractive potential. This potential varies inversely as the

sixth power of the distance r' between the centers of the atoms or molecules. The dependence on r' yields $V(d) \propto d^{-3}$, and if one equates the gravitational and van der Waals potentials one obtains

$$d = kh^{-1/3}, \quad (6)$$

where k is a constant. We have used published potential parameters for He² and Ar¹⁶ together with a combining rule¹⁷ consistent with the London theory, from which we calculate that for He on an Ar substrate $k = 2.92 \times 10^{-6}$ cm^{4/3}. This is about the same magnitude for k as has been measured for He on glass. However, the London theory described above is incomplete and must be supplemented as indicated below.

It has been shown¹⁸ that for large separations between atoms, retardation causes the attractive force to be weaker than the London theory predicts and the potential to be proportional to r'^{-7} rather than r'^{-6} . Sabisky and Anderson¹ have recently made machine calculations of $V(d)$ using the macroscopic theory constructed by Lifshitz^{19,20} that is very general and takes into account many-body forces and retardation. Their results show that $V(d)$ is no longer proportional to d^{-3} for $d > 10$ Å, and also that the magnitude of the potential is only about $\frac{2}{3}$ of that determined above, where retardation was neglected. Since the relationship between d and h is no longer simple in this more complete and correct theory, we will continue to use Eq. (6) in the following development, but with k replaced by $k(d)$, a slowly varying function of d .

Substituting Eqs. (3) and (6) into Eq. (2), and setting h equal to H , one obtains

$$\sigma = (\rho_s/\rho)k^{3/4}H^{-1/4}. \quad (7)$$

We now use for k values of $k(d)$ from the curve for a solid Ar substrate in Fig. 8 of the paper of Sabisky and Anderson.¹ For the range of d relevant to our experiment, 140–250 Å, we find that to a very high degree of accuracy (better than 0.2%) we can represent the predicted transfer rates by the equation

$$\sigma = 5.05(\rho_s/\rho)H^{-0.21} \times 10^{-5} \text{ cm}^2/\text{s}. \quad (8)$$

The dependence of $k(d)$ on d is absorbed very well by changing the exponent of H from -0.25 to -0.21 . We continue to use the form $\sigma = \sigma_0 H^{-n}$ because it is convenient and correctly describes the theoretical result in the range of interest.

RESULTS

We have made five Ar beakers that did not leak liquid He I at an observable rate, and which were kept free of surface contamination. Of the transfer rates observed with these five beakers, the trans-

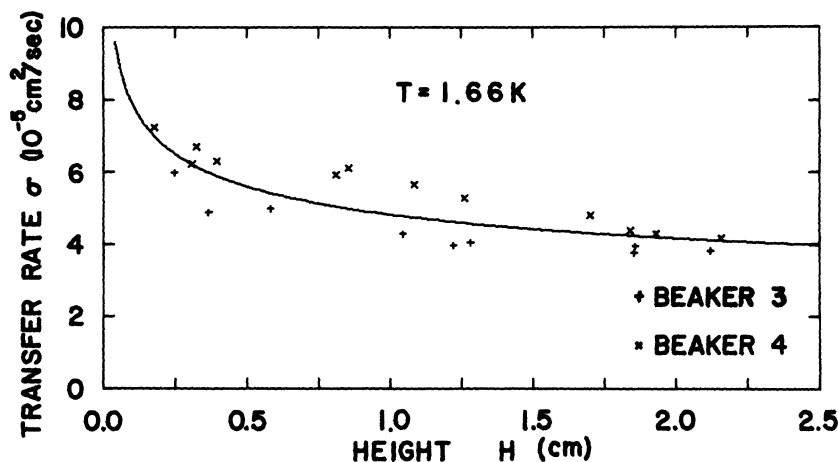


FIG. 5. Transfer rate at 1-mm level difference as a function of the height H from the outer He level to the top of the beaker for beaker fillings at a temperature of 1.66 K. The solid curve is a fit to the data of the form $\sigma = \alpha_0 H^{-n}$. For this curve the parameter values are $\alpha_0 = 4.8 \times 10^{-5}$ cm²/s and $n = 0.21$.

fer rates with two of the beakers are the lowest and are in reasonably good agreement with each other. It is the results of our experiments with these two beakers that we report here. The other three beakers showed transfer rates from 15% to 50% higher than the ones reported here. These higher rates may have been due to an increased microscopic surface area of the beaker walls or quite possibly due to tiny leaks in the walls, much too small to be seen with He I. For a given beaker, in general, transfer rates were reproducible to better than 10% and obeyed the expected flow rate dependences on the height to the beaker lip and the level difference.

For beaker fillings, best fits to Eq. (5) result in a value for B of $(3.2 \pm 1.7) \times 10^3$ s/cm², with Z in cm. Scatter in B masks any dependence of it on temperature. This value of B yields a value of

$b(T)$ in Eq. (4) of the order of 15. This agrees in order of magnitude with the results of Cannon, Chester, and Jones,⁸ who found for $b(T)$ temperature-dependent values ranging from 30 to 70 in the temperature range of our experiment. The range of Z over which observations were made is too small to make this experiment a good one for determining B .

Since the transfer rates predicted by Eq. (8) are appropriate to small level differences, where $v_s \approx v_{s,c}$, we deduce from the fitted curve for each beaker filling the transfer rate corresponding to 1-mm level difference. We choose this level difference because it does not involve extrapolating the fitted curve for σ beyond the range of our data. Figure 5 is a plot of our results for beaker filling experiments at 1.66 K for σ at 1-mm level difference versus the height H from the outer He

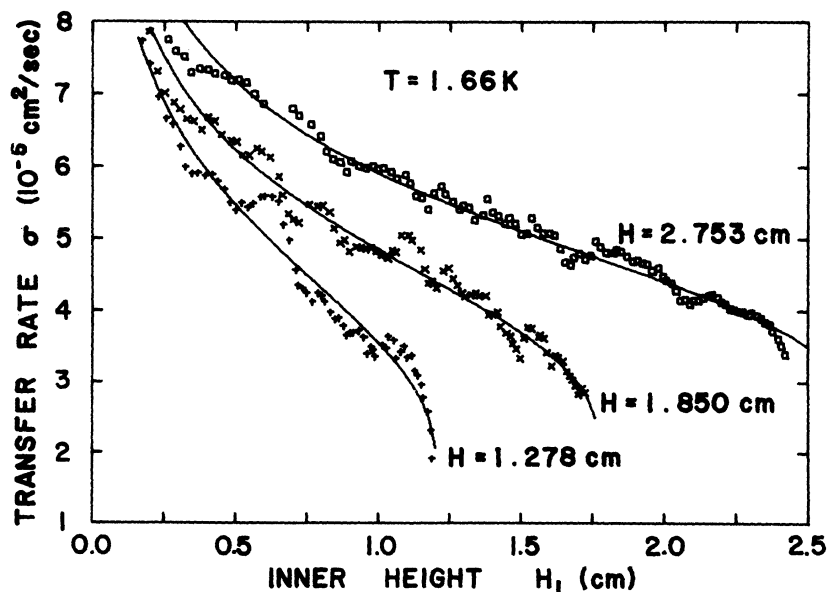


FIG. 6. Transfer rate as a function of the height H_I from the inner He level to the beaker rim at three different heights H from the outer He level to the beaker rim. The solid curves are fits to the data of the form $\sigma = H_I^{-0.21} / (A - B \times \ln Z)$.

TABLE I. Comparison of the experimental results for beaker emptying and filling rates with the theoretical predictions. The transfer rates σ_0 correspond to 1-mm level difference and 1-cm film height.

T (K)	Theory [Eq. (8)]		Filling experiments		Emptying experiments	
	$10^5 \sigma_0$ (cm ² /s)	$10^5 \sigma_0$ (cm ² /s)	n	Number of fillings	$10^5 \sigma_0$ (cm ² /s)	Number of emptyings
1.99	2.28	3.2	0.41	6
1.77	3.54	3.9	0.26	6	3.1	2
1.66	3.99	4.8	0.21	21	4.3	12
1.47	4.59	4.7	0.22	11	4.3	3

level to the top of the beaker. The solid curve is a fit of the form $\sigma = \sigma_0 H^{-n}$ to the data points. The values of the parameters for the curve are $\sigma_0 = 4.8 \times 10^{-5}$ cm²/s and $n = 0.21$. The exponent $n = 0.21$ is in good agreement with Eq. (8), the theoretical result. The rms deviation of the data points from this curve is 0.5×10^{-5} cm²/s.

We collected beaker-filling data at several other temperatures. The results are summarized in Table I. The magnitudes of σ_0 as calculated from Eq. (8) are also displayed for comparison.

The transfer rates from three beaker-emptying experiments are plotted in Fig. 6. The ordinate H_l is the height from the He level inside the beaker to the lip, which increases as the beaker empties. Both the level difference and the height relevant to the film thickness change simultaneously. Therefore, we fit each beaker emptying to the two-parameter form

$$\sigma = H_l^{-0.21} / (A - B \ln Z). \quad (9)$$

Equation (9) contains the dependence on both height, as in Eq. (8), and level difference, as in Eq. (5). We also make an additive correction of 0.07 cm to the level difference Z for the fitted curves for both emptyings and fillings to compensate for the observed capillary rise of the He inside the beaker. The solid curves in Fig. 6 are fitted curves of Eq. (9) to the data.

In order to compare the beaker-emptying rates to the beaker filling rates, we calculate, using the parameters A and B from each emptying, the transfer rate σ_0 for 1-mm level difference and 1-cm film height. The mean values of σ_0 for the beaker-emptying runs are displayed in column six of Table I. They are consistently smaller than for beaker fillings, shown in column three of Table I. This is probably a result of the geometry of the beaker, especially of the flare in the i.d.

at the top.² The best fits result in a value of B for the beaker emptyings of $(4.8 \pm 2.0) \times 10^3$ s/cm², somewhat higher than, but overlapping with, the value for the beaker fillings.

CONCLUSIONS

As can be seen from Figs. 4 and 6, the level-difference dependence of the transfer rates we have measured is described well by fits to Eq. (5). This is especially apparent in Fig. 6, since without any level-difference dependence the emptying rates should be independent of the height H from the outer level to the beaker lip.

From Table I, the temperature dependence of the observed transfer rates is somewhat weaker than Eq. (8) predicts, although the observed rates do decrease substantially at the higher temperatures. The agreement is quantitatively best around 1.66 K, where we collected most of the data.

The He II film transfer rates that we have observed using Ar beakers are, to the best of our knowledge, the lowest that have been observed for the same temperatures and film heights. Good quantitative agreement is obtained between our results and the usual theory that has been successful in describing transfer rates over glass. In using this theory, one must be careful to use the correct calculation, including retardation effects, for the van der Waals attractive potential between the He and the Ar substrate.

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Bilinear Hydrodynamics and the Stokes-Einstein Law*

T. Keyes[†] and Irwin Oppenheim

Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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The autocorrelation function of the density of a tagged particle is studied using the Mori formalism. The variables used are the collective conserved variables, the tagged-particle density, and bilinear products thereof. The case of point particles is considered in two dimensions, and, in three dimensions, self-diffusion by a particle of arbitrary size is treated. It is found that the bilinear-hydrodynamic approach automatically separates the self-diffusion coefficient of the tagged particle into a nonhydrodynamic part, and a hydrodynamic part which resembles the Stokes-Einstein law. In two dimensions, it is found that the mean-square displacement of a particle increases as $t \ln t$, and that certain natural redefinitions of the diffusion and friction coefficients leave Einstein's law invariant. In three dimensions, for a large particle, the Stokes-Einstein law is reproduced. The relation between the well-known $t^{-3/2}$ "tails" on correlation functions, and the Stokes-Einstein law, is discussed.

I. INTRODUCTION

Stokes¹ has calculated the force on a sphere in uniform motion through a continuum which obeys the laws of linearized Navier-Stokes hydrodynamics, and which sticks to the sphere at its surface. The result is

$$\vec{F} = -6\pi\eta R\vec{p}/m, \quad (1)$$

where \vec{p} is the momentum of the sphere, m is the mass, and R is the radius, while η is the coefficient of shear viscosity of the continuum. The friction coefficient of Brownian-motion theory ξ is defined by the relation

$$\vec{F} = -(\xi/m)\vec{p}; \quad (2)$$

thus, we have Stokes law

$$\xi = 6\pi\eta R. \quad (3)$$

Einstein's law² relates the diffusion coefficient D of a Brownian particle to the friction coefficient

$$D = k_B T / \xi, \quad (4)$$

where k_B is Boltzmann's constant and T is the absolute temperature. The combination of Eqs. (3) and (4) yields the Stokes-Einstein law²

$$D = k_B T / 6\pi\eta R, \quad (5)$$

which relates the diffusion coefficient of a Brownian sphere to the viscosity of the solvent, the radius of the sphere, and known constants. From a molecular point of view, the Brownian-motion conditions for which Eqs. (1)–(5) ought to be valid correspond to the motion of a large heavy particle through a dense solvent of small light molecules. More precisely, the mass of the heavy particle must be large compared to the mass of a solvent particle and its radius must be large compared to the distance between solvent particles. It is indeed true³ that solutions of macromolecules in small-molecule solvents appear to obey the Stokes-Einstein law.

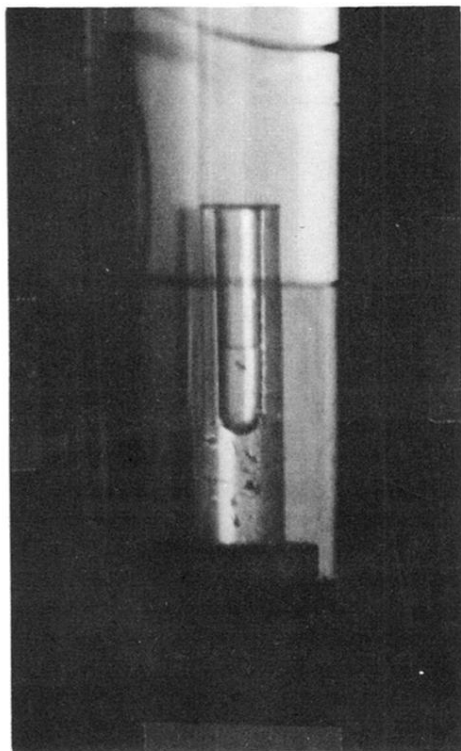


FIG. 2. Photograph of one of the Ar beakers. The beaker is filling, and the liquid He levels inside and outside the beaker can be seen. Some defects are visible in the solid Ar, especially below the end of the bore.