

## Dependence of the Helium-Film Transfer Rate on Pressure Head, Film Height, and Substrate\*

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Helium-film transfer rates  $\sigma$  have been measured as a function of level difference  $z$  and height to the beaker rim  $h$  for filling clean glass and neon-coated beakers. Observed transfer rates as a function of level difference or pressure head are described by:  $\sigma(z) = 1/[A - B \ln z \text{ (cm)}]$ . For  $T=1.65$  K and  $h=7$  cm, we find  $A=(1.80 \pm 0.09) \times 10^4 \text{ sec/cm}^2$  and  $B=(8.9 \pm 1.9) \times 10^2 \text{ sec/cm}^2$ . For  $T=1.28$  K and  $h=7$  cm, we find  $A=(1.35 \pm 0.02) \times 10^4 \text{ sec/cm}^2$  and  $B=(8.3 \pm 1.4) \times 10^2 \text{ sec/cm}^2$ . Dependence of the transfer rate on film height is  $\sigma(h) = 10(\rho_s/\rho)h^{-(0.26 \pm 0.05)} \times 10^{-5} \text{ cm}^3$ . For a glass beaker coated with Ne, the transfer rate is 16% less than the transfer rate for the same uncoated beaker. This is in reasonable agreement with our calculated helium-film thickness for a Ne substrate of  $2.3 h^{-1/3} \times 10^{-6} \text{ cm}$ .

### I. INTRODUCTION

Any surface in contact with a liquid helium II reservoir is covered with a thin mobile helium film. The character of superfluid flow in very narrow geometries can be examined by studying fluid transfer through the film. Typically, transfer into or out of a beaker is measured, and the superfluid velocity  $v$  is related to the transfer rate per unit circumference  $\sigma$  by

$$\sigma = (\rho_s/\rho)vd. \quad (1)$$

Here  $\rho_s/\rho$  is the superfluid fraction and  $d$  is the thickness of the film.

In the first systematic study of film transfer, Daunt and Mendelssohn<sup>1</sup> observed that  $\sigma$  was nearly independent of pressure head, height of the beaker rim above the reservoir, and substrate material. They concluded that  $\sigma$  primarily depended on the temperature. This lack of dependence on pressure head and film length led them to postulate that the flow was taking place at a characteristic or critical velocity  $v_c$ .

Subsequently Atkins,<sup>2</sup> and also independently Esel'son and Lazarev,<sup>3</sup> observed that the transfer rate did depend on pressure head and height. The variation of  $\sigma$  with height arises from the dependence of the film thickness on height

$$d = k/H^{1/3}. \quad (2)$$

Here  $d$  is the film thickness at a height  $H$  from the reservoir and  $k$  is a constant, for a given substrate material, which is equal to  $3.0 \times 10^{-6} \text{ cm}^{4/3}$  for clean glass.<sup>4</sup> However, the actual dependence of  $\sigma$  on  $H$  is less than the dependence of  $d$  on  $H$  because  $v_c$  itself is a function of  $d$ .

The parameter  $k$  depends on substrate material and results in a dependence of  $\sigma$  on substrates. Smith and Boorse<sup>5</sup> investigated the dependence of  $\sigma$  on several glass and metal substrates. They

concluded, however, that the typical 10% background variations in film experiments prevented the systematic resolution of a dependence of the transfer rate on the materials they investigated.<sup>6</sup>

In the present investigation we have studied the dependence of the transfer rate on pressure head and height in more detail. In particular, we are interested in the functional form of these dependences for comparison with theory. To keep background variations to a minimum, the geometry of the beaker is kept simple and a very clean and vibration free chamber is used in these experiments. We have also investigated the substrate dependence of  $\sigma$  for a neon-coated glass breaker.

### II. FILM THICKNESS

If the thickness  $d$  is known, properties of the critical velocity  $v_c$  can be determined from measurements of  $\sigma$ . From the optical measurements of Ham and Jackson,<sup>4</sup>  $k$  in Eq. (2) is taken to be  $3.0 \times 10^{-6} \text{ cm}^{4/3}$  for clean glass. In addition, the dependence of  $d$  on substrate material can be calculated and compared with experimental results.

In the theoretical treatment of Frenkel,<sup>7</sup> the thickness of the helium film as a function of height, given in Eq. (2), is determined by the balance between the additional attractive van der Waals potential to the solid substrate and gravitational potential energy.<sup>8-10</sup> Using this method, Schiff has calculated the value of  $k$  for some substrates and finds<sup>11</sup>

	Cu	Ag	Glass	Rocksalt
$k(10^{-6} \text{ cm}^{4/3})$	4.3	4.7	$\approx 4$	2.2

The thickness, however, does not depend much on substrate for materials commonly used in experiments, in agreement with the measurements of Smith and Boorse.<sup>5</sup>

However, if a neon substrate is used, the film

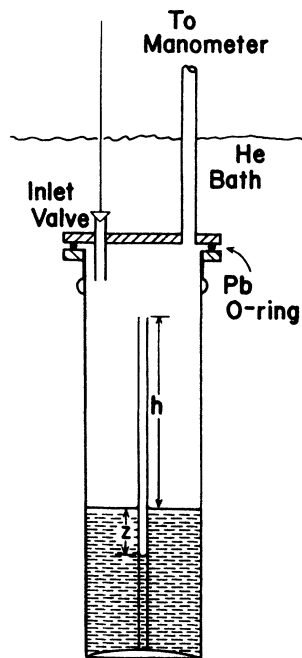


FIG. 1. Schematic drawing of the experimental chamber and beaker. The chamber can be demounted and the beaker removed for thorough cleaning.

will be appreciably thinner. For a rare-gas solid, the van der Waals attraction can be determined from the attractive part of the known Mie-Lennard-Jones potential<sup>12,13</sup>

$$\Phi_{\text{attr}} = -2\epsilon_{12}(r_{12}^*/r)^6, \quad (3)$$

where the subscript 1 on the potential parameters stands for He, and 2 stands for any of the other rare gases.

The potential parameters  $\epsilon_{12}$  and  $r_{12}^*$  are determined from the pure rare-gas parameters using the coupling rules<sup>14</sup>

$$r_{12}^* = (r_{11}^* + r_{22}^*)/2 \quad (4)$$

and

$$\epsilon_{12} = (\epsilon_{11}\epsilon_{22})^{1/2}. \quad (5)$$

Using the method of Frenkel<sup>7</sup> and Schiff<sup>11</sup> and the additional attraction to the substrate as determined by Eqs. (3)–(5), we determine for rare-gas solid substrates

$$k = [(\epsilon_{12}r_{12}^*{}^6n_2 - \epsilon_{11}r_{11}^*{}^6n_1)/3mg]^{1/3}. \quad (6)$$

In Eq. (6),  $n_1$  and  $n_2$  are the respective number densities and  $m$  is the helium atomic mass. The appropriate values of  $k$  are

	Ne	Ar	Kr	Xe
$k(10^{-6}\text{cm}^{4/3})$	2.3	3.0	3.1	3.5

For a neon-coated glass beaker we calculate that the forces are saturated at  $10^{-5}$ -cm neon coverage.

### III. APPARATUS AND PROCEDURE

The apparatus is designed to provide a very clean environment for the beaker. If impurities are condensed on the beaker, much larger transfer rates than normal are seen.<sup>15</sup> This is due to an increased microperimeter for a granular impurity or a thicker helium film. The thick films can arise from a polar impurity such as water which has a much stronger attraction than the usual van der Waals potential.<sup>16</sup>

Therefore, beakers used in these experiments were protected from condensation of impurities by isolating them from the helium bath in the experimental chamber. This is shown schematically in Fig. 1. In addition, the chamber is diffusion pumped during cool down as well as for degassing between runs. Helium is admitted to the chamber from the bath by means of a needle valve rather than by condensing possibly impure gas in the chamber.

The chamber is made from a 2-in. Kovar to glass seal and is demountable by means of a lead O ring. The glass beaker could be removed from the demounted chamber for thorough cleaning in solvents that are not compatible with the metal parts of the chamber.

The chamber is immersed in a pumped helium bath the temperature of which is regulated to better than 0.01 K by means of a mechanical pressure regulator. Temperatures in the chamber and the bath are measured using mercury and oil manometers.

Beakers used in these experiments were made of Pyrex tube of 3-mm i.d., 5-mm o.d., and 10-cm height. The beakers were inspected to be free of scratches and were lightly fire polished at the rim. The radius of the beaker was determined volumetrically using distilled water at room temperature. Data taken with three identical beakers agree within experimental uncertainties.

Before each run the beaker was cleaned by the following procedure: It was first rinsed with distilled water and methanol. This was followed by cleaning with detergent in an ultrasonic bath and rinsing ultrasonically in distilled water. The beaker was then rinsed with nitric acid and rerinsed with distilled water. It was then heated to 200 °C in air to remove adsorbed water. Finally, it was mounted in the experimental chamber immediately after heating and diffusion pumped to about  $10^{-6}$  Torr, for one or two days prior to cool down.

The crucial step in the cleaning procedure is heating to remove adsorbed water. If the beaker is not heated, enhanced transfer rates are seen, presumably due to a thicker helium film. Additional heating to about 75 °C while pumping was sometimes used, and was found to be unnecessary. Results

reported here were obtained by cleaning the beaker before each run. Reproducible results were also obtained by pumping the helium away immediately after a run and keeping the beaker under a vacuum at room temperature between runs.<sup>17</sup>

Two different means of illumination gave identical results. The experiment was done with a strip-silvered Dewar either in a darkened room with no special illumination or in a completely dark room using a fluorescent light with infrared filters.

During a run, helium is admitted to the experimental chamber through the inlet needle valve, filling the chamber outside the beaker to 2-cm depth. The position of the inner level is then measured every 30 sec to an accuracy of 0.001 cm. Referring to Fig. 1 we see that the outer level remains a nearly constant distance  $h$  below the rim while the beaker is filling. The level difference  $z$  can then be determined as a function of time for a fixed  $h$ .

After the levels have reached equilibrium, more helium is admitted to the chamber. The level difference  $z$  is then determined as a function of time for this new  $h$ . Figure 2 shows  $z$  as a function of time for three different  $h$ 's in the same run. In this paper, results are reported on 16 runs of typically five beaker fillings at five different  $h$ 's in every run.

#### IV. DATA ANALYSIS

The transfer rate per unit circumference  $\sigma$  is proportional to the time derivative of  $z$ ,

$$\sigma = \frac{r}{2} \frac{dz}{dt}, \quad (7)$$

where  $r$  is the inside radius of the beaker. Derivatives are obtained from the  $z$ -versus- $t$  curves with a computer using the following procedure: A quadratic equation

$$z(t) = A + Bt + Ct^2 \quad (8)$$

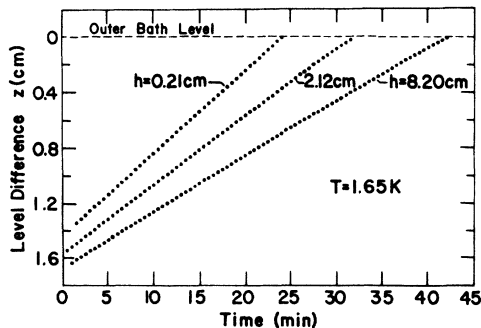


FIG. 2. Level difference as a function of time for three beaker fillings at three different heights  $h$  from the outer level to the beaker rim. The transfer rate is proportional to the slope of these curves.

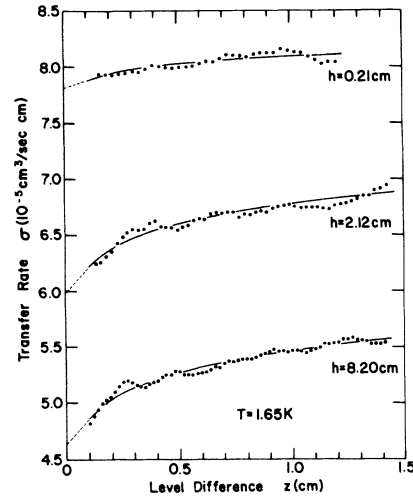


FIG. 3. Transfer rate as a function of level difference for the three beaker fillings of Fig. 2. For  $z > 0.1$  cm, the transfer rate is described by the solid curve which is of the form  $\sigma = 1/(A - Blnz)$ . For  $z < 0.1$  cm, the data are described by the dashed curve which is a linear extrapolation from the curve for large  $z$ . This extrapolation gives the zero-pressure-head transfer rate  $\sigma_c$ .

is fitted to the first 11 data points using the method of least squares. The derivative  $dz/dt = B + 2Ct$  is then evaluated at the midpoint of the segment. Next, data points 2-12 are fitted in the same way and the derivative is obtained from the midpoint of this segment. By taking overlapping segments of the  $z(t)$  curve in this manner, the transfer rate  $\sigma$  can be determined either as a function of  $z$  or  $t$ . Figure 3 shows  $\sigma$  as a function of  $z$  for the three sets of data in Fig. 2. Each point on these curves is obtained by evaluating the derivative at the midpoint of one of the overlapping 11 data-point segments.

The datum-point segment must be chosen to be small enough to follow the actual  $z(t)$  curve rather than the conveniently chosen quadratic equation. On the other hand, the segment must be large enough so that random errors of individual data points do not result in large fluctuations in the derivative. Segments of 11 data points were chosen as a compromise between these two competing effects. From the rms deviation of the 11  $z(t)$  points from the fitted curve, we estimate that there is a 0.6% error in the individual  $\sigma(z)$  points.

In order to relate these observations to the critical velocity, we must consider the dynamics of the flow. The equation of motion of  $z$  can be derived from the two-fluid equations of motion to be<sup>18</sup>

$$M^* \frac{d^2 z}{dt^2} = -\rho g z + P_s, \quad (9)$$

where  $M^* = (\rho^2/2\rho_s) \int_{r_{film}} (r/d) dl$ . Here  $l$  is a coordinate along the path of the film and the line integral is taken from the outside level of the beaker to the inside. The critical velocity has been taken into account by splitting the pressure into two parts:  $P_s$  is the velocity-dependent frictional pressure due to dissipative processes in the film, and  $\rho gz$  is the gravitational pressure driving the flow.<sup>19</sup>

We are primarily interested in the frictional pressure  $P_s$  as a function of superfluid velocity or transfer rate. For  $z > 0.1$  cm, it may be calculated from Fig. 3 that the observed acceleration  $d^2z/dt^2$  is small enough that  $M^* d^2z/dt^2$  is negligible compared to  $\rho gz$ . Then the frictional pressure equals the observed driving pressure, i. e.,  $P_s = \rho gz$ . Hence, for  $z > 0.1$  cm, the curves in Fig. 3 can be interpreted as giving the frictional pressure as a function of  $\sigma$  or the  $I$ - $V$  characteristic of the film.

For  $z < 0.1$  cm, the acceleration term becomes important and  $P_s$  becomes small. In this case inertial effects result in a nonzero transfer rate at zero-pressure head. That is, the inner level overshoots the outer level and subsequently oscillates about it.<sup>18</sup> Our apparatus, however, is not suitable for studying these oscillations.

#### V. PRESSURE-HEAD DEPENDENCE

We notice from the curves on Fig. 3 that  $\sigma$  does depend on  $z$ , decreasing by about 10% over a change in head of 1.5 cm. For  $z > 0.1$  cm, the data give the dependence of the frictional pressure on  $\sigma$  or  $v_s$ . In this region we observe that the data may be described by<sup>20</sup>

$$\sigma(z) = 1/[A - B \ln z(\text{cm})] \quad (10)$$

The solid curves on Fig. 3 are of this form. This functional form is a good fit to all the data observed in 80 beaker fillings. The  $\sigma$ -versus- $z$  data are fitted on a computer by the method of least squares to obtain the parameters  $A$  and  $B$ . For  $T = 1.65$  K, we find  $A = (1.80 \pm 0.09) \times 10^4 \text{ sec/cm}^2$  and  $B = (8.9 \pm 1.9) \times 10^2 \text{ sec/cm}^2$  with  $h = 7$  cm. For  $T = 1.28$  K, we find  $A = (1.35 \pm 0.02) \text{ sec/cm}^2$  and  $B = (8.3 \pm 1.4) \times 10^2 \text{ sec/cm}^2$  with  $h = 7$  cm. The errors given here are the rms scatter in the observed  $A$ 's and  $B$ 's.

We observe that  $B$  is independent of  $h$  for  $h > 4$  cm. For  $h < 4$  cm,  $B$  decreases so that  $\sigma$  is nearly independent of  $z$ , as can be seen on the top curve of Fig. 3. This decrease of  $B$  at small  $h$  may not be a property of the helium but may be a result of the rapidly changing film thickness when  $z$  is comparable to  $h$ . However, for  $h \approx 7$  cm the thickness changes sufficiently slowly with  $h$  so that pure pressure-head dependence can be seen. Most of the height dependence of  $\sigma$  is contained in  $A$  which is the subject of Sec. VI.

For  $z < 0.1$  cm, the acceleration term in Eq. (9) is important, and as a result the frictional pressure cannot be measured directly. In this region  $\sigma$  has been approximated by a straight-line extrapolation from the curve for large  $z$ . These are shown as the dashed curves on Fig. 3 from which the zero-pressure-head transfer rate is given as

$$\sigma_c = \sigma(0.1 \text{ cm}) - B[\sigma(0.1 \text{ cm})]^2 \quad (11)$$

Here the zero-pressure-head rate is taken to be the critical rate  $\sigma_c$  at that  $h$ .

We have also looked for abrupt changes in the transfer rate as reported by Harris-Lowe *et al.*<sup>21</sup> and Allen and Armitage.<sup>17</sup> As the level difference decays it appears that the transfer rate has a stepped structure as reflected in the departure of the data from the fitted curves in Fig. 3.

The deviations of the  $\sigma(z)$  data from the fitted curves are larger than would be expected from the measurement errors of the individual points. The average rms deviation of the  $\sigma(z)$  points from the fitted curves is found to be 1.2%, which is twice the 0.6% deviation expected from the errors of the individual  $\sigma(z)$  points.

The data have also been examined for abrupt changes in the transfer rate using the more direct method of Harris-Lowe *et al.*<sup>21</sup> Although the results of this analysis are again suggestive of steps, the data are not sufficiently accurate to give more quantitative information.

These deviations result in an approximately 1.2% random error in  $A$  and  $\sigma_c$ , and an approximately 16% random error in  $B$ , for a given beaker filling. In addition, there are comparable run-to-run background variations in  $A$  and  $\sigma_c$  presumably resulting from differing substrate conditions. These two sources of error limit the total run-to-run accuracy of  $A$  and  $\sigma_c$  to about 4%.

These  $\sigma(z)$  results for the film are in qualitative agreement with Atkin's<sup>2</sup> original experiment and with recent results of Martin and Mendelssohn.<sup>22</sup> Similar results were also obtained by Keller and Hammel<sup>23</sup> for flow through a slit.

The functional form of the pressure-head dependence agrees with Notarys's<sup>24</sup> observations of superfluid flowing through narrow pores. However, there are several disagreements with the application of Notarys's results and his extension of the Langer-Fisher<sup>25</sup> theory to the film. The most serious disagreement is in the temperature dependence. The Langer-Fisher theory gives  $A(T), B(T) \propto T/(\rho_s/\rho)^2$ . We observe  $A(1.65 \text{ K})/A(1.28 \text{ K}) = 1.33$  and  $B(1.65 \text{ K})/B(1.28 \text{ K}) = 1.1$ . Hence, for the narrow temperature range we investigated, we find that  $B$  is nearly temperature independent and  $A(T) \propto \rho/\rho_s$ . Within experimental uncertainties this gives a critical transfer rate which agrees with the usual:  $\sigma_c(T) \propto \rho_s/\rho$ . These observed temper-

ature dependences also give an increasing  $d\sigma/dz$  with decreasing  $T$  in agreement with Martin and Mendelssohn.<sup>22</sup>

### VI. HEIGHT DEPENDENCE

The critical velocity occurs in the region near the top of the beaker.<sup>28</sup> Using the dependence of the film thickness of height given in Eq. (2), we can determine the dependence of the critical velocity on thickness from a measurement of  $\sigma_c$  versus  $h$ . This can be compared with theory. Several theories using Feynman-Onsager<sup>19,27</sup> vortices give the dependence  $v_c d \propto \ln d$  which results in  $\sigma_c$  only weakly dependent on  $h$ . From the examination of many different experiments spanning several decades of channel width, van Alphen *et al.*<sup>28</sup> have proposed the empirical relation  $v_c \approx 1d^{-1/4}$  cm/sec. For  $d = 3h^{-1/3} \times 10^{-6}$  cm, this gives  $\sigma_c \approx 7(\rho_s/\rho)h^{-1/4} \times 10^{-5}$  cm<sup>3</sup>/sec cm.

The dependence of  $\sigma_c$  on  $h$  is determined from a log-log plot of the  $\sigma_c$ -versus- $h$  data. Figure 4 shows a log-log plot of the observed zero-pressure-head transfer rates  $\sigma_c$  versus  $h$  for the data at 1.65 K. For  $h$  in the range 2–9 cm the data are described by a straight line. A least-squares fit gives  $\sigma_c \propto h^{-0.19}$  for the data in this range.<sup>29</sup>

For  $h < 2$  cm, the transfer rate is nearly independent of  $h$  as well as  $z$ . The departure of the data from the straight line occurs near  $h = 2$  cm, i. e., when the initial level difference  $z$  is comparable to  $h$ . This suggested performing an experiment with an initial level difference of 0.3 cm instead of the usual 1.5 cm. It was then observed that the departure occurred at 0.6 cm instead of 2 cm as in Fig. 4. For  $h > 0.6$  cm, we have in this case  $\sigma_c \propto h^{-0.20}$ .

These observations indicate that the thickness at the rim is not exactly determined by the distance

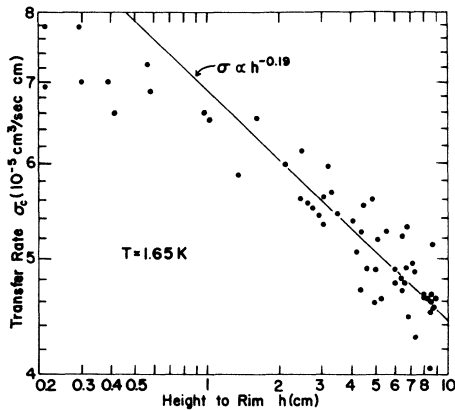


FIG. 4. Log-log plot of the zero-pressure-head transfer rate at 1.65 K versus height to the beaker rim from the outer level. For  $h > 2$  cm, the data are described by  $\sigma_c \propto h^{-0.19}$ .

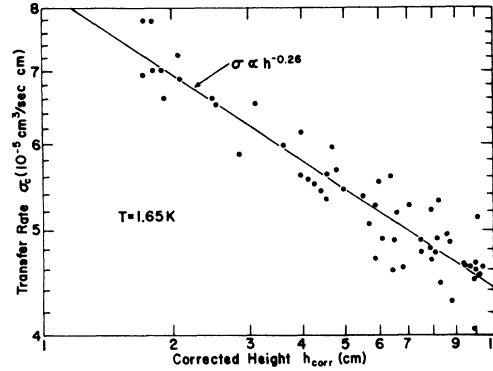


FIG. 5. Log-log plot of the zero-pressure-head transfer rate at 1.65 K versus corrected height,  $h_{\text{corr}} = h + 1.5$  cm. The corrected data are described by  $\sigma_c = 8.3h^{-1/4} \times 10^{-5}$  cm<sup>3</sup>/sec cm.

$h$  to the source reservoir and that further corrections must be made.<sup>2</sup> It is unlikely that the thickness is determined by the instantaneous distance to the inner level from the rim, because in this case  $\sigma$  would increase with decreasing  $z$  for small  $h$ . From the top curve on Fig. 3, we see that this is not observed.

The data in Fig. 4 suggest that  $h$  is displaced by a constant factor. This leads us to make the plausible assumption that the proper distance to use for determining the thickness is a distance  $h_{\text{corr}} > h$ . This correction appears to be a result of the non-zero initial level difference except that the observed slope on the log-log plot did not increase for small  $z_{\text{initial}}$ . This is similar to metastable behavior that has been seen for beakers that were filled by submersion.<sup>30</sup> Abrupt changes in the transfer rate as reported by other investigators<sup>17,21</sup> are also suggestive of metastable behavior of the film.

If we make the correction,  $h_{\text{corr}} = h + 1.5$  cm, we get a straight line on a plot of  $\log \sigma_c$  versus  $\log h_{\text{corr}}$  and also agree with the empirical relation  $\sigma_c \propto h^{-1/4}$ . This is shown in Fig. 5. A least-squares fit to the corrected data at  $T = 1.65$  K gives  $\sigma_c = (8.3 \pm 0.2) h_{\text{corr}}^{-(0.26 \pm 0.05)} \times 10^{-5}$  cm<sup>3</sup>/sec cm. Although we have less data at  $T = 1.28$  K, if these data are treated in the same way we get  $\sigma_c = (10) h_{\text{corr}}^{-1/4} \times 10^{-5}$  cm<sup>3</sup>/sec cm. Combining these results we have

$$\sigma_c = 10(\rho_s/\rho)h_{\text{corr}}^{-1/4} \times 10^{-5} \text{ cm}^3/\text{sec cm}. \quad (12)$$

This agrees with the results of Allen and Armitage<sup>17</sup> and also with the empirical  $h^{-1/4}$  behavior. The magnitude of  $\sigma_c$  at  $h = 2$  cm agrees with many other experiments,<sup>31</sup> although it is larger than the results of Allen and Armitage<sup>17</sup> and the proposed value of van Alphen *et al.*<sup>28</sup> discussed above.

### VII. SUBSTRATE DEPENDENCE

We have investigated also the dependence of the transfer rate on substrate for neon substrates.

This was done by coating a clean glass beaker with Ne taking care to provide a smooth surface and also to exclude polar impurities from the condensed gases. In this way we were able to observe a decrease in  $\sigma$  going from a clean glass substrate to the neon substrate in the same apparatus.

The apparatus used in the substrate study is similar to the clean glass apparatus shown in Fig. 1. In order to raise the temperature of the experimental chamber during the Ne condensation, it was enclosed in a vacuum chamber made from a second glass to metal seal. Also, the experimental chamber was wound with a heating coil and provided with a platinum resistance thermometer to control and measure the temperature.

To coat the beaker, a dilute mixture of Ne gas in He gas was admitted to the chamber with the initial temperature at 25 K. Neon was then condensed on the beaker by lowering the temperature at a rate of 0.2 K/min from 25 to 4 K. The He gas serves both to distribute the Ne and to maintain thermal equilibrium within the chamber. We assume that the Ne condenses uniformly on all surfaces of the chamber and beaker. Reproducibility and magnitude of transfer rate data indicate that this procedure results in a smooth surface. However, no independent test of the surface was made.

We have calculated that the He-Ne forces are saturated at 1000-Å coverage so that at this coverage we have an effective Ne beaker. One-half saturation  $d = (d_{\text{Ne}} + d_{\text{glass}})/2$  occurs at 100-Å Ne coverage. Here  $d_{\text{Ne}}$  and  $d_{\text{glass}}$  are the thicknesses of the helium film for a neon and glass substrate, respectively.

The minimum observed transfer rates occurred for neon coverage in the range 500–5000 Å. At 100-Å coverage, we observed a larger  $\sigma_c$  than the

minimum, as expected because the forces are not saturated. At 10000-Å coverage, we observed that  $\sigma_c$  was again larger than the minimum. This is possibly due to unavoidable rough substrate conditions. All the observed transfer rates over Ne were greater than or equal to the theoretically expected Ne rates, also possibly due to slightly rough substrates. However, the rates were less than the clean glass results.

For neon coverage in the range 500–5000 Å, the lowest observed rates are  $\sigma_c = 4.1 \times 10^{-5}$  cm<sup>3</sup>/sec cm with  $h = 7$  cm and  $T = 1.65$  K. This was observed in four separate experiments. Two other experiments at this coverage gave  $\sigma_c = 4.5 \times 10^{-5}$  cm<sup>3</sup>/sec cm, presumably due to a slightly rough substrate. The rate for clean glass in the same apparatus at the same  $h$  and  $T$  was  $\sigma_c = 4.86 \times 10^{-5}$  cm<sup>3</sup>/sec cm.

We can now compute the ratio of the thicknesses of the helium film on a Ne substrate and on a glass substrate. This may be done by using the relation  $\sigma_c \propto d^{3/4}$ , obtained from Eqs. (1) and (2) and  $v_c \propto d^{-1/4}$ .<sup>28</sup> The ratio of the thicknesses obtained in this manner is

$$d_{\text{Ne}}/d_{\text{glass}} = (\sigma_{\text{Ne}}/\sigma_{\text{glass}})^{4/3} = 0.79. \quad (13)$$

Using  $d_{\text{glass}} = 3.0h^{-1/3} \times 10^6$  cm, we get  $d_{\text{Ne}} = 2.4h^{-1/3} \times 10^6$  cm. This agrees very well with our value of  $d_{\text{Ne}} = 2.3h^{-1/3} \times 10^6$  cm previously calculated above.

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PHYSICAL REVIEW A

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## Cross Sections for Electron Capture and Loss by Fast Bromine and Iodine Ions Traversing Light Gases\*

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Bromine and iodine ions, accelerated to energies between 6 and 15 MeV, were passed through thin gas targets of H<sub>2</sub> and He. Nonequilibrium charge distributions were measured, from which cross sections for capture and loss of one or more electrons by the heavy ions were determined by means of a least-squares fitting technique. The cross sections are discussed with regard to their functional dependence on the charge and velocity of the ions. Owing to the unprecedented precision of the results, characteristic irregularities were revealed in the charge dependence of the cross sections for both capture and loss of one and two electrons. These effects are attributed to the influence of shell structure and residual excitation of the ions.

### I. INTRODUCTION

Atomic collisions between energetic ions and matter have been studied for a number of years, both for the purpose of understanding the fundamental physics of atomic interactions and for a more practical reason, the necessity of producing intense high-energy beams from heavy-ion accelerators. In the past, the lighter ions have been studied intensively.<sup>1</sup> Since only a few measurements have been reported<sup>2-6</sup> of charge-exchange cross sections for ions with a nuclear charge greater than 18, additional investigations are of the greatest importance. For example, systems of highly accurate cross sections for the capture and loss of one or more electrons in a single collision by fast heavy ions with many adjacent charge states have never been reported in detail. Although it is believed that existing theories describe the essentials of the charge-changing mechanisms, it is not yet possible to predict values, often as large as  $10^{-15}$  cm<sup>2</sup>, of the many cross sections involved in the interaction.

In this experiment, Br and I ions have been accelerated to energies between 6 and 15 MeV, passed through targets of He and H<sub>2</sub> gas of varying thick-

ness, and the distributions of ionic charge states in the emerging beam were detected. The experimental procedure<sup>7</sup> has been described previously, as well as the technique of analysis<sup>6,8</sup> which has been employed to obtain charge-changing cross sections with uncertainties of 5 to 10%. Additional details of both the experimental procedure and the data treatment, which yielded these high accuracies, are included in the body of this paper. Results confirm the existence of shell effects previously reported,<sup>6</sup> but a thorough analysis of the data showed that these shell effects can be strongly influenced by ions which are in excited states before the charge-changing collisions occur. This indicates that residual ion excitation is of greater importance than previously believed and cannot be neglected in many heavy-ion experiments. A method of analysis is discussed to take into account residual ion excitation in order to obtain consistent sets of charge-changing cross sections.

In the present investigation, shell structures were identified in the double-capture cross sections, as well as in the single- and multiple-loss cross sections. The shell effects in electron-loss probabilities are small and seem to support qualitative theoretical assumptions about single electron loss.