Nonuniformity of the dielectric permittivity and the density profile of adsorbed ^{4}He films

Yu. N. Moiseev, V. I. Panov, and G. I. Salistra

M. V. Lomonosov State University, Moscow

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The thickness dependence of the dielectric permittivity (DP) of thin ^{4}He films has been studied experimentally. The nonuniformity in the distribution of the DP and density of liquid helium near the boundary with a solid wall was determined and agrees qualitatively with theoretical predictions. The physical parameters of the substrate which characterize the van der Waals potential were found for a metallic adsorbing surface. Estimates were obtained for the dielectric permittivity, $\varepsilon_s = 1.0725$, and thickness, $D_0 = 7.1$ Å, for the solid layer of helium separating the substrate from the gas phase.

1. INTRODUCTION

The dielectric permittivity ε and density ρ of thin liquid ^{4}He films have long been studied extensively by different methods (see, for example Refs. 1–3 and the literature cited there). There are a number of reasons for the interest in such investigations.

On the one hand, a detailed experimental investigation of the dependence of ε and ρ on the thickness l of the layer makes possible a comparison with theoretical predictions,[4–6] in which the contribution of the van der Waals forces, decreasing according to the l^{-3} law and dependent on the substrate and solid layer parameters, gives rise to nonuniformity in the distribution of ε and ρ.

On the other hand, the exact form of the distribution of ε and ρ near a solid boundary (below the temperature of the λ transition) is essential for a quantitative study of the effects of superfluidity in systems with finite geometry, since the density profile carries information about the distribution of the superfluid order parameter $\tilde{\Psi}$ and about the boundary condition for $\tilde{\Psi}$ at a solid wall. The existing experimental data confirm qualitatively that ε and ρ of ^{4}He films vary near a solid boundary. However, their quantitative comparison with theoretical relations[5,6] is almost impossible because of the uncertainty in the parameters which characterize the solid film, or as a result of insufficiently accurate experiments.

At the same time the high accuracy now obtainable in capacitance measurements[7–9] makes possible a quantitative observation of the predicted effects and the use of these methods for determining the physical parameters of the substrate, of the solidified helium layer and of the thickness of the adsorbed film. In the present work, therefore, the variation in the distribution of DP of helium near the boundary with a solid wall has been determined to high accuracy on the basis of a detailed experimental investigation of how the dielectric permittivity (DP) ε of thin liquid ^{4}He films depends on the thickness l. The profile of the mean density of the helium layer adsorbed on the substrate has been calculated from the results obtained, using the Clausius–Mosotti relation and the known value of the polarizability of ^{4}He. Numerical values of the van der Waals constant α and of the Casimir–Polder parameter Z, which characterize the potential of the van der Waals forces, were found for the metallic adsorbing substrate (Ag) and an estimate of the thickness D_0 and of the dielectric permittivity averaged over the thickness D_0 was obtained

$$\varepsilon_s = \frac{1}{D_0}\int_0^{D_0} \varepsilon_s(x)\,dx$$

for the solid layer of helium, separating the substrate from the liquid phase. The experimental results were compared with theoretical predictions,[5,6] in which the action of long-wave fluctuations in the electromagnetic field of the substrate on the distribution of the DP of the helium layer was calculated.

It was found that the analytical dependence of the nonuniform part of the DP of the film on its thickness obtained[5,6] for $1 > D_0$,

$$\Delta\varepsilon(l) = A_1 l^{-1} - A_2 l^{-3}(1 - Z^{-1}l - C_1 l^{-3/2}),$$

agrees qualitatively with the experimental results. The best agreement between theory and experiment is obtained for the following values of the coefficients:

$$A_1 \approx 0{,}137\ \text{Å},\ A_2 \approx 2{,}2\ \text{Å}^3,\ C_1 \approx 5{,}5\ \text{Å}^{3/2}.$$

The constant $\alpha = \alpha_{\exp} = 2.62\cdot 10^{-37}$ erg/cm^3, which characterizes the metallic substrate (Ag) is found to be 1.8 times less than the theoretical estimate,[10] but it agrees well with an independent experimental result.[3] The estimates found for the mean dielectric permittivity of the solid ^{4}He layer $\varepsilon_s \approx 1.0725$ and its thickness $D_0 \approx 7.1$ Å at a temperature $T = 1.530$ K indicate that the adsorbed layers in the present experiments are separated from the substrate by two atomic layers of solid helium.

2. EXPERIMENTAL METHOD AND APPARATUS

The determination of the variation in the distribution of dielectric permittivity ε (density ρ) of helium films near the boundary with a solid wall is based on the measurement of the electrical capacity of a plane capacitor on the plates of which helium is adsorbed.

The experimental method is based on recording the difference ΔC in capacity of two plane-parallel capacitors, placed in a chamber into which helium vapor is fed.

One, C_m, is used as the measuring capacitor, the other, C_0, as the reference ($C_m = C_0 = C$). The distance between

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the plates of the measuring d_m and of the reference capacitor d_0 was chosen from the condition $d_m \ll d_0$. Under this condition, when the thickness l of the adsorbed layers is small ($l \ll d_m, d_0$) the recorded quantity ΔC and the parameters of the film studied are connected by the relation

$$\frac{\Delta C}{2C} \approx \frac{\varepsilon_2(\varepsilon_1-\varepsilon_2)}{\varepsilon_1} l \left(\frac{1}{d_m}-\frac{1}{d_0}\right)\left(1+\frac{2l}{\varepsilon_1 d_m}(\varepsilon_1-\varepsilon_2)\right). \quad (1)$$

Here $\varepsilon_1 = \varepsilon_0 + \Delta\varepsilon(l)$ is the mean value over the thickness of the DP of the adsorbed film to be determined, consisting of the DP ε_0 of bulk helium and its nonuniform part $\Delta\varepsilon(l)$; ε_2 is the DP of gaseous helium contained between the plates of the capacitors.

The thickness of the adsorbed film was determined with the help of the barometer formula

$$\left|\ln\frac{P}{P_0}\right| = \frac{\alpha}{\varkappa T}\frac{1}{l^3(1+l/Z)}. \quad (2)$$

Here P is the equilibrium value of the gaseous helium pressure at temperature T in the chamber where the capacitors are; P_0 is the saturated vapor pressure of ^{4}He at the same temperature; α and Z are constants characterizing the properties of the substrate, the magnitudes of which were determined by experiment. In the theory[4] α and Z are related to the parameter $\hbar\tilde{\omega}$ of the substrate:

$$\alpha = \frac{\hbar\tilde{\omega}(\varepsilon_0-1)}{16\pi^2}\frac{m}{\rho}, \quad Z = \frac{3}{2}\frac{\hbar c}{\hbar\tilde{\omega}}, \quad (3)$$

m is the mass of a ^{4}He atom.

The measurements were carried out using a simplified version of the apparatus (Fig. 1) which was used earlier to study the λ transition of ^{4}He in a system of finite geometry.[11] Its cryogenic part consisted of the measuring chamber, filled with the pure ^{4}He being studied, an auxiliary chamber for letting gaseous helium into the measuring chamber in controlled amounts, and the cooled part of the electronic recording system.

The measuring chamber of diameter 60 mm and height 18 mm was made of annealed copper. The chamber was connected to two isothermal massive platforms by thermal links, and the induction coils and leads were placed on these (to reduce the heat flux into the chamber). A heater was wound on the surface of the measuring chamber, which was used to stabilize the temperature of the chamber. A capacitance manometer was housed in the upper part of the chamber, which was used to determine the helium vapor pressure. The indications of the manometer were stabilized by a diaphragm made of annealed beryllium bronze. The diaphragm acted as a mobile electrode of the capacitor recording its deflection for a pressure change in the chamber. A fixed electrode was mounted above the mobile electrode and under it the ring electrode of the capacitor for calibrating the manometer by electrostatic force. The sensitivity of the manometer in the pressure range 0–1 atm was $\Delta C_p/\Delta P = 4.6$ pF/atm. The electronic recording system of the capacitance manometer made it possible to measure pressure with a relative uncertainty of $0.5\cdot 10^{-4}$.

The auxiliary chamber of volume 0.2 cm^3 was connected by a capillary to the measuring chamber. This capillary, with a copper wire inserted into it to reduce its internal cross section, was used to admit ^{4}He into the measuring chamber. A heater wound on an 0.8 cm section of the capillary was used to heat the ^{4}He admitted into the chamber, preventing the formation of a superfluid film when the helium was admitted. Helium was bled into the auxiliary chamber through a superleak and a cut-off valve at a temperature $T \approx 1.4$–1.5 K. A second valve was fixed to the measuring chamber and was used for measuring out the helium admitted into it, entering through a leak. A pressure bellows placed in the upper part of the chamber provided fine mechanical control of the gaseous ^{4}He pressure in the chamber by changing the bellows volume through compression or expansion. The measuring and reference capacitances were mounted in the copper chamber on the horizontal surface of the 10 mm thick massive block, completely made of leucosapphire.

A semiconductor resistance thermometer with a mean sensitivity $\Delta R/\Delta T = 52$ kΩ/K was fixed onto this same block in a hole drilled in it and placed in the space between the capacitors. There were two auxiliary thermometers on opposite side faces of the block, used for control measurements and for revealing circumstances under which temperature differences between the measuring and reference capacitances could arise. All the thermometers, calibrated in advance according to the T_{58} and T_{62} temperature scales, were surrounded by thermally insulating plugs and radiation shields. In the measuring regime the power dissipated in the thermometers was less than 10^{-8} W. Under these conditions the electronic measuring system allowed relative changes in temperature to be recorded with an accuracy of $\sim 10^{-6}$ K. At the same time, long-term temperature stabilization of the chamber for a fixed value of the temperature was achieved to an accuracy of $\lesssim 10^{-4}$ K, with the help of a thermometer on the surface of the chamber and an electronic stabilization system.

All the tubes supporting the vacuum chamber, the rods for adjusting the electronic system and the valves for admitting helium were covered with radiation shields. In addition, while experiments were carried out the rods were raised to eliminate thermal and mechanical contact with the isothermal platform and the chamber. The electrical leads for the

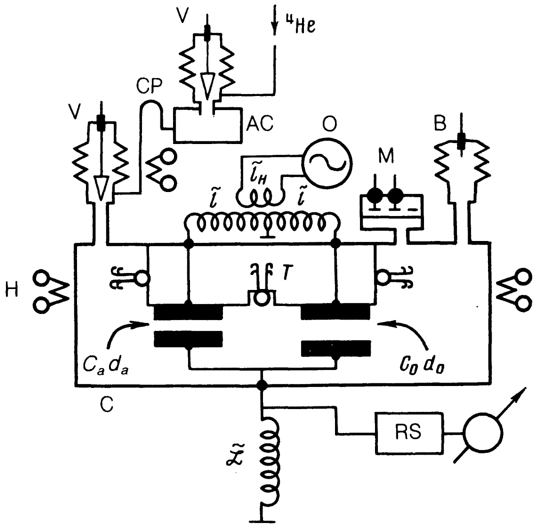

FIG. 1. Circuit of the experimental apparatus: C—measuring chamber, AC—auxiliary chamber, Cp—capillary tube, V—shut-off valve, T—thermometer, H—heater, M—manometer, B—bellows, O—oscillator, RS—recording system.

electronic recording system, the capacitance manometer and for the heaters and thermometers were connected to the chamber by a number of thermal links, reducing the heat influx to the chamber along the leads.

The cryogenic part of the electronic system for indicating the capacity was constructed in the form of a bridge balance circuit, into which the measuring and reference capacitances were connected. In such a system the high sensitivity to a relative variation in capacity (reaching $\lesssim 10^{-9}$) is associated with the small value of the electric field in the gaps of the capacitances, which would introduce undesirable striction effects in the film studied. Under the experimental conditions the amplitude of high-frequency oscillations on the capacitance plates was less than 0.2 V at the oscillation frequency of 3.7 MHz provided by the high-stability quartz oscillator.

The inductances $\tilde{l}$, $\tilde{l}_n$, and $\mathscr{L}$ of the circuits of the bridge system, which were superconducting at helium temperatures, were wound of lead-coated copper wire on cylindrical sapphire casings and covered with a layer of epoxy resin with an Al_2O_3 powder filling in order to obtain maximum temperature stability. The components of the bridge circuit were mounted so as to obtain minimum and equal stray capacitances connected in parallel with the measuring and reference capacitors, and stability to vibrational and acoustic interference.

The main systematic experimental difficulty was in attaining high accuracy in the preparation of the capacitors with a reliably controlled electrode geometry. The measuring and reference capacitances were made by thermally coating an Ag film in ultra-high vacuum on a polished surface of plane-parallel quartz disks of diameter 20–22 mm. In order to obtain a reliable control of the geometry and parallelness of the electrodes of the measuring capacitance, its construction proceeded in several stages (Fig. 2).

Three small support areas (columns) of diameter ~ 4 mm and a central electrode of diameter 1.6 mm were evaporated simultaneously onto the surface of one of the quartz plates. On the other quartz plate (with the second electrode) the height of the small areas was built up by additional evaporation to a height corresponding to the required distance d_m between the electrodes. After the parallelness of the surfaces of the quartz plate and the surface of the silver film evaporated onto it was checked, the plates were connected together. The final parallelness of the electrodes was exhibited and controlled optically by the interference pattern which resulted from light reflected from the parts of the inner surfaces of the quartz plates uncovered by metallic film (Fig. 2). The plates were then glued with low-temperature epoxy resin. The capacitance gap obtained was calculated from the measured capacitance of the capacitor, taking account of the influence of the leads.

Two quartz plates were used for making the reference capacitance, each of which was coated on one side by an Ag film. Coating of an electrode was done over the whole area of the surface of a plate except for a narrow ring of width 1 mm along the perimeter of the plate. The initial value of the gap d_0 of this capacitance was provided by three dielectric washers introduced between the quartz plates. The capacitor C_0 was made equal to the capacitor C_m by squeezing the quartz plates in a special yoke. After the plates were secured with epoxy resin the washers were removed. Measurement of the equality of C_m and C_0 was carried out with a calibrated electronic bridge circuit for determining capacitance. The final equalizing of the capacitors C_m and C_0 to an accuracy $\Delta = \Delta C/C \lesssim 1\%$ was carried out after fixing them to the sapphire block in the chamber and carrying out a cycle of cooling and heating. As a result of using this method for preparing the plane-parallel capacitors with gap $d_m = 3.0\cdot 10^{-5}$ cm and $d_0 = 5.2\cdot 10^{-3}$ cm the difference in the values of the capacitances ($C_m = C_0 = 52$ pF) was, on cooling the apparatus, less than $\Delta = 0.2\%$.

The uniformity of the surface of the substrate plays an important part in the adsorption of a film on it. The microprofile and the distribution of uniformity of work function along the surface of the electrodes which adsorb ^{4}He were monitored with the help of a scanning tunneling microscope.[12] The results of these measurements for three control sections of the surface, of dimensions 1.5×1.5 μm^2, are shown in Fig. 3. The maximum variation in the contour height of the surface of the electrodes was ~ 20 Å on a scale of ~ 200 Å and did not make an appreciable contribution to the effective area of the surface of the electrodes. The distribution of work function for the same sections of the electrodes, measured immediately after deposition and after carrying out the experiment, indicated a small nonuniformity in the electrophysical properties of different sections of the Ag surface, which could lead to variations in the constants α and Z, characterizing the adsorbing surface. It should also be remarked that the Ag film deposition regime was chosen

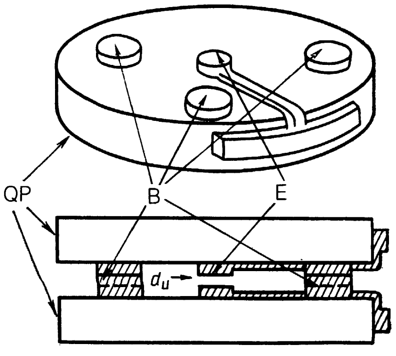

FIG. 2. Design of measuring capacitance: E—electrode, B—bearing surfaces, QP—quartz plate.

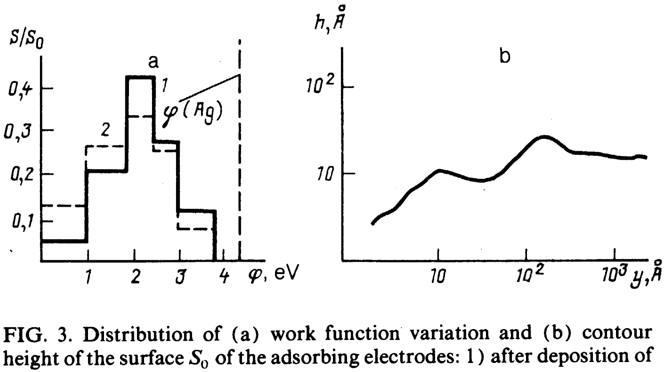

FIG. 3. Distribution of (a) work function variation and (b) contour height of the surface S_0 of the adsorbing electrodes: 1) after deposition of the electrodes; 2) after carrying out the measurements.

from the condition for obtaining maximum uniformity in contour and work function of the surface of the electrodes adsorbing the ^{4}He film.

When analyzing the results of the experiment and estimating the overall sensitivity of the method, we took into account systematic and random errors Δ_i making a contribution to the recorded value $\Delta C/2C$. They were calculated from the relation

$$\sum_i \Delta_i \approx \frac{\delta l}{d_m}(\varepsilon_1-\varepsilon_2)+\delta\varepsilon_1\frac{l}{d_m}+\delta\varepsilon_2\frac{l}{d_m}+\Delta_e+\Delta_s+\Delta_{re}; \quad (4)$$

where $\Delta_e \sim 10^{-8}$ is the error produced by slow variations in the amplification coefficient and the nonlinearity of the electronic recording system;

$$\Delta_s \leqslant \Delta\frac{l}{d_m}(\varepsilon_1-\varepsilon_2)$$

is the systematic error associated with the difference Δ in capacitance between the measuring and reference capacitors.

The largest random error in Eq. (4)

$$\Delta_{re} \approx \frac{l}{d_m}\frac{\delta T}{3T}(\varepsilon_1-\varepsilon_2) \lesssim 5\cdot 10^{-8}$$

is associated with variations in the thickness of the adsorbed ^{4}He film on chamber temperature drifts during the whole cycle of measurements. The systematic error Δ_s, growing linearly with film thickness, was $\lesssim 2\cdot 10^{-8} l$ (Å). For $l \sim 10^2$ Å (which corresponded to the maximum thickness of film studied) it reached the value $\Delta_s \approx 2\cdot 10^{-6}$. At the same time this systematic error gave only a constant increment to the recorded value of ε_1 and did not affect the results of measurement of the nonuniformity in the distribution $\Delta\varepsilon(l)$. The magnitude of this constant addition to ε was taken into account when analyzing the results obtained.

In order to reduce the number of adsorbed molecules of water and of chamber gases, the chamber was pumped for a day while slowly reducing the temperature from ~100 °C to room temperature and only after that was cooled slowly.

Measurement of the variation in the distribution of the DP of adsorbed helium films was carried out after the chamber was filled with pure gaseous ^{4}He, at least an hour after the a steady temperature was established and about 15 min after the pressure stabilized.

After the conclusion of a cycle of measurements the chamber was filled to ~0.8 of its volume with liquid helium and a check was carried out on the reading of the pressure gauge, showing the magnitude of the saturated vapor pressure P_0 of ^{4}He. This quantity was used to calculate the thickness of the adsorbed films, using Eq. (2).

3. INVESTIGATION OF THE VARIATION IN THE DISTRIBUTION OF DIELECTRIC PERMITTIVITY (DENSITY) OF THIN ^{4}He FILMS

The variation in the distribution of dielectric permittivity $\varepsilon(l)$ [or density $\rho(l)$] of thin ^{4}He films near the boundary with a solid wall was studied before and was, in part, confirmed qualitatively on analysis of the mass of the adsorbed layer of helium and its thickness.[13]

However, the absence of accurate experimental results for the dependence $\varepsilon(l)$ or $\rho(l)$ and their connection with the parameters of the substrate made comparison with theoretical predictions impossible,[4-6] and thus we could not accurately determine the contribution to $\varepsilon(l)$, $\rho(l)$ of van der Waals forces, which fall off as l^{-3}, by taking into account the thickness D_0 and the DP ε_s of the layer of solid helium, since they are not known in advance.

In addition, below the λ point the form of the $\rho(l)$ dependence requires for its interpretation many effects of the superfluidity of ^{4}He in a limited geometry, since the distribution of the superfluid order parameter $\tilde{\Psi}$ determines this dependence and carries information about the boundary condition at the solid wall, which in the general case has the form $d\tilde{\Psi}/dx = \Lambda^{-1}\tilde{\Psi}$ (Λ is a phenomenological parameter, called the "extrapolation length").[14]

By using this method we could study quantitatively the distribution $\varepsilon(l)$ of DP of thin layers of adsorbed ^{4}He and the mean density profile $\rho(l)$, using the Clausius-Mosotti relation

$$\frac{\varepsilon-1}{\varepsilon+2}=\frac{4\pi\rho\alpha_e}{3\mu}=\frac{4\pi\alpha_e}{3V_\mu}, \quad (5)$$

which has been reliably established[15] for ^{4}He using the value of the polarizability (in cm^3/mol) $\alpha_e = 0.123396 - (0.005604/V_\mu)$.

The dielectric permittivity of adsorbed helium films was studied at a temperature of 1.530 K. In the experiments the change in the capacitance difference ΔC was recorded as a function of the equilibrium pressure P of the gaseous helium in the chamber. The results of the measurements are shown in Fig. 4 in the form of the relation

$$\left(\frac{\Delta C}{2C}\right)^{-3}=\Phi\left(\left|\ln\frac{P}{P_0}\right|\right).$$

The procedure for analyzing these results in relation to Eqs. (1) and (2) was analogous to the method which was used for finding the critical indices and coefficients when studying the λ transition in ^{4}He (Ref. 9). Into it went the evaluation of the van der Waals constant α, the Casimir parameter Z, the determination of the thickness of the adsorbed layer l and the variable part $\Delta\varepsilon(l)$ of the DP of the film. In the calculations the values used were $\varepsilon_0 = 1.05722$ for $T = 1.530$ K

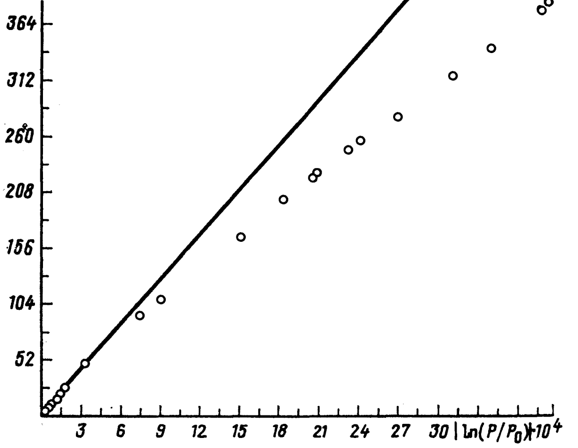

FIG. 4. Capacitance difference as a function of the equilibrium pressure (on $|\ln(P/P_0)|\cdot 10^2$) in the chamber.

(Ref. 9) and $\varepsilon_2(P) = (1 + 6.8\cdot 10^{-5} P/P_0)$, obtained from the equation of state.

The analysis was carried out numerically in several stages. A short description of it amounts to the following.

The first stage consisted of the determination of the constants α and Z. For this purpose we first analyzed the experimental points of Fig. 4, corresponding to massive helium films ($|\ln(P_i/P_0)| \lesssim 2\cdot 10^{-2}$), in which the effects of the retardation $\sim l/Z$ become appreciable, while the contribution of the nonuniform part of the DP can be neglected to a first approximation. Comparison of these points with the analytical dependence

$$\left(\frac{\Delta C}{2C}\right)^{-3} = \Psi\left(\left|\ln\frac{P}{P_0}\right|, \alpha, Z\right),$$

obtained from Eqs. (1) and (2), neglecting the variable part of the DP of massive films, made it possible to find to lowest order $\alpha = \alpha^0$ and $Z = Z^0$ with an uncertainty of 6% (α^0 and Z^0 were determined by standard statistical analysis, for which

$$|\Psi(|\ln(P_i/P_0)|, \alpha, Z) - \Phi_i|$$

is minimized by varying the desired quantities α and Z). The uncertainty in the values of α and Z was reduced to $\sim 2\%$ and their final values were determined after taking account of a small correction to Ψ from the contribution of the variable part of the DP in a massive film $\Delta\varepsilon^0(|\ln(P/P_0)|)$, also obtained in this stage to lowest order. The $\Delta\varepsilon^0$ dependence used for this purpose came from an analysis of the result for the difference between Ψ, when we neglect the contribution of the variable part of the DP and the remaining experimental points Φ_i for thin films ($|\ln(P_i/P_0)| \gtrsim 2\cdot 10^{-2}$) in which the effect of variation in the DP dominates. The values of α_{exp} and Z_{exp} found by this analysis were used to calculate l_i according to Eq. (2). [Examination of the spread in experimental points Φ_i in the region of massive films from that calculated by taking the correction in $\Psi(|\ln(P_i/P_0)|, \alpha_{\text{exp}}, Z_{\text{exp}})$ into account showed that the dispersion was less than two standard deviations.]

The second stage of the statistical analysis consisted of finding for each value of the film thickness l_i the magnitude of its DP: $\varepsilon_i(l) = \varepsilon_0 + \Delta\varepsilon_{\text{exp}}(l)$. The DP of the film was evaluated from the measurements of the value of $(\Delta C/2C)$, using Eq. (1).

The following values of the parameters of the substrate (characterizing the potential of the van der Waals forces) were obtained as a result of the analysis: $\alpha_{\text{exp}} = (2.62 \pm 0.06)\cdot 10^{-37}$ erg/cm^3 and $Z_{\text{exp}} = (280 \pm 5)$ Å. The value of α_{exp} differs from the theoretical estimate $\alpha \approx 4.9\cdot 10^{-37}$ erg/cm^3, given earlier[10] for adsorbed ^{4}He atoms on Ag, but agrees well with a previous[3] experimental result $2.6\cdot 10^{-37}$ erg/cm^3. The difference between α and α_{exp} averaged over the surface of the substrate, may be produced both by oxidation processes of the Ag in separate sections and by a difference in the adsorptive properties of the faces of the microcrystals in the polycrystalline substrate. The results of measurements of the variation in work function along the substrate surface (Fig. 3) point to disturbances of this sort.

In addition, the reliability of the mean values of α_{exp} and Z_{exp} obtained in the experiment which were used to calculate the mean thickness of adsorbed layers of helium, is confirmed by their agreement with Eqs. (3) to an accuracy of $\sim 6\%$.

The variable part of the DP $\Delta\varepsilon(l)$ of a ^{4}He film makes its main contribution to the relation

$$\left(\frac{\Delta C}{2C}\right)^{3} = \Phi\left(\left|\ln\frac{P}{P_0}\right|\right)$$

for large $|\ln(P/P_0)|$ (small l), which is illustrated by the noticeable departure of the experimental points (Fig. 4) from a straight line:

$$\left(\frac{\Delta C}{2C}\right)^{-3} = \left[\frac{\varepsilon_2(P_0)(\varepsilon_0 - \varepsilon_2(P_0))}{\varepsilon_0}\right]^{-3} \times \frac{\varkappa T}{\alpha_{\text{exp}}}\left|\ln\frac{P}{P_0}\right| = B\left|\ln\frac{P}{P_0}\right|.$$

The DP of a ^{4}He film thus depends on its thickness l and the effect is perfectly clear with the accuracy achieved in our experiment.

TABLE I.

№	P/P_0	l, Å	$\Delta\varepsilon_{\text{exp}}\cdot 10^8$	$\Delta^{\text{exp}}\cdot 10^8$	$\Delta\rho/\rho_0$, %
1	0,67703	14,4	8,950	4,65	15,35
2	0,68178	14,5	8,896	3,96	15,26
3	0,71101	15,1	8,588	2,07	14,73
4	0,73212	15,5	8,355	0,79	14,33
5	0,76312	16,3	7,994	−1,15	13,71
6	0,78513	16,9	7,724	−2,31	13,25
7	0,792	17,1	7,637	−2,63	13,10
8	0,81109	17,7	7,385	−3,59	12,67
9	0,8144	17,8	7,340	−3,87	12,59
10	0,82981	18,4	7,132	−3,84	12,23
11	0,86139	19,8	6,656	−4,60	11,41
12	0,91288	23,2	5,733	−3,38	9,83
13	0,9264	24,6	5,426	−3,39	9,30
14	0,9651	31,5	4,310	0,92	7,39
15	0,98128	38,5	3,555	2,92	6,10
16	0,98363	40,2	3,419	3,55	5,86
17	0,98828	44,8	3,101	5,98	5,32
18	0,99307	53,0	2,636	5,95	4,52
19	0,99838	83,5	1,697	5,76	2,91
20	0,99868	88,9	1,598	5,90	2,74

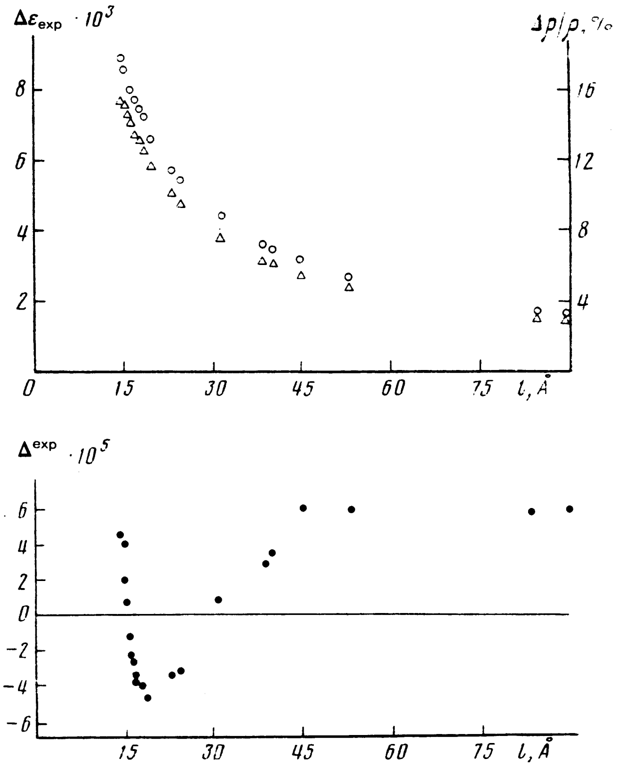

FIG. 5. Distribution of the variable part of the DP and of the density of the adsorbed ^{4}He film: △) $\Delta\rho/\rho$, ○) $\Delta\varepsilon_{\rm exp}$, ●) $\Delta^{\rm exp}$.

The measured values of $\Delta\varepsilon_{\rm exp}(l)$ are given in Table I and in Fig. 5.

4. DISCUSSION OF THE RESULTS

To compare the results of the experiment with theoretical predictions[5,6] we consider a plane film of liquid helium with the surfaces of separation $x = 0$ (metal-liquid helium) and $x = l$ (liquid helium-gas). The variable part of its DP

$$\Delta\varepsilon(l)=\frac{1}{l}\int_0^l\Delta\varepsilon(x)\,dx \qquad (6)$$

is determined by the local density profile $\rho(x)$. It is usual to assume[1,16] that the film is separated from the substrate by a layer of solid ^{4}He of thickness D_0, which according to various measured data lies within the limits 4–8 Å, and can be estimated from the solidification pressure of helium $\tilde P_s$ using the expression[4]

$$\tilde P_s=\frac{\hbar\bar\omega(\varepsilon_0-1)}{16\pi^2 D_0^3}. \qquad (7)$$

Following others[5,6] we will assume that for $x > D_0$ the local DP of a film $\varepsilon(x)$ is mainly determined by the van der Waals forces, and we can take the following model for it:

$$\varepsilon(x)=\varepsilon_0+\Delta\varepsilon^{W}(x),\quad D_0\leqslant x\leqslant l. \qquad (8)$$

The general expression for the DP of a film $\Delta\varepsilon^{W}(x)$, resulting from van der Waals forces is known,[5,6] but it is difficult to make use to obtain numerical estimates and a comparison with experiment. Furthermore, on the basis of the experimental conditions, we can limit ourselves to terms of second order in $\varepsilon_0 - 1$, and also neglect the contribution of temperature corrections and the contribution to the DP of the liquid-gas surface of separation, since its value is $\lesssim 10^{-6}\varepsilon_0$. (Under the experimental conditions the contribution of the retarded potential should be taken into account for all film thicknesses). In this case to the accuracy indicated

$$\Delta\varepsilon^{W}(x)=\frac{(\varepsilon_0-1)^2\hbar\bar\omega\beta(P_s)}{16\pi^2\varepsilon_0 x^3(1+x/Z)}. \qquad (9)$$

We have here taken into account the pressure dependence of the isothermal compressibility $\beta(P_s)$, the value of which changes from $5.5\cdot 10^{-8}$ CGSE units at a pressure $P_s\sim 20$ atm (i.e., at a distance $x = D_0 \approx 6$ Å from the substrate) to $\beta = 12.5\cdot 10^{-8}$ CGSE units at $P_s\to 0$ (i.e., at a distance $x > 30$ Å)

In this pressure range, using earlier results[17] and the fact that x and P_s are connected by the condition of hydrostatic equilibrium, $\beta(P_s)$ can be expressed in the form

$$\beta(P_s)\approx\beta(1-\Delta\beta(x)/\beta), \qquad (10)$$

$\Delta\beta(x)/\beta = 9.64x^{-1/2}$, where x is in Å. As a result, substituting Eqs. (8)–(10) into Eq. (6) yields the following dependence of the DP of a film on its thickness:

$$\Delta\varepsilon(l)=\frac{A_1}{l}-\frac{A_2}{l^3}\left(1-\frac{2l}{Z}-\frac{4}{7}\frac{\Delta\beta(l)}{\beta}\right). \qquad (11)$$

Here

$$A_1[\text{Å}]=D_0(\varepsilon_s-\varepsilon_0)+\frac{A_2}{D_0^2}\left[1-\frac{2D_0}{Z}-\frac{4}{7}\frac{\Delta\beta(D_0)}{\beta}\left(1-\frac{7}{5}\frac{D_0}{Z}\right)\right], \qquad (12)$$

$$A_2[\text{Å}^3]=\frac{(\varepsilon_0-1)^2\beta\hbar\bar\omega}{32\pi^2\varepsilon_0}.$$

In order to compare the theoretical relation (11) with the results of the experiment, we find from Eq. (12) the theoretical estimate of the magnitude of the coefficients A_1 and A_2. For example, for $D = 6$ Å, $Z = 280$ Å, $\varepsilon_s = 1.072$, $\hbar\bar\omega = 1.6\cdot 10^{-11}$ CGSE units, we obtain $A_1\approx 0.12$ and $A_2\approx 1.9$ Å^3. However, it must be said that the accuracy of these estimates is not high due to the indeterminacy in the quantities D_0 and ε_s (D_0 and ε_s depend on the parameters of the substrate and on temperature and will, consequently, differ for different experimental conditions). For this reason we used numerical values of the coefficients to verify the relation of Eq. (11), $A_{1\,\rm exp} = 0.13697$ Å and $A_{2\,\rm exp} = 2.20$ Å^3, which were determined from actual experiment by analyzing the results in Table I (the second and third columns) and for which the best agreement between theory and experiment was obtained. The values of $\Delta\varepsilon(l)$ calculated in this way according to Eq. (11) (for $A_1 = A_{1\,\rm exp}$ and $A_2 = A_{2\,\rm exp}$) were compared with the measured $\Delta\varepsilon_{\rm exp}(l)$. The small difference ($\sim 1.5\%$) between the experimental values $\Delta\varepsilon_{\rm exp}(l)$ and the theoretical estimates $\Delta\dot\varepsilon(l)$, expressed in the form $\Delta^{\rm exp}(l) = \Delta\varepsilon_{\rm exp}(l) - \Delta\varepsilon(l)$ in Table I (fourth column) and in Fig. 5, indicate the sufficiently good agreement of the experimental results with the theoretical dependence of Eq. (11). At the same time, a small non-monotonic variation in $\Delta^{\rm exp}(l)$ is observed in the range 14–35 Å, which requires further analysis.

The distribution of the variable part of the mean density $\Delta\rho(l)$ near the boundary with a solid wall is calculated from the value of $\Delta\varepsilon_{\rm exp}(l)$ using the Clausius–Mosotti relation.[5]

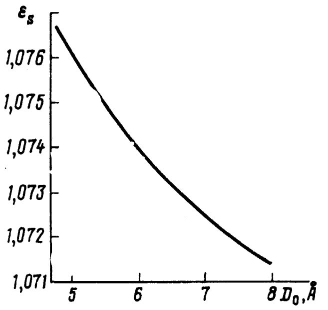

FIG. 6. The $\varepsilon_s(D_0)$ relation.

The results of these calculations are shown in Fig. 5 and can be used to analyze experiments to study superconductivity in thin ^{4}He films.

It was shown above that $\Delta\varepsilon(l)$ (for $l\to D_0$) is determined by the thickness D_0 and DP ε_s of the solid layer of helium. These quantities can not be measured separately by the capacitance method to the necessary accuracy, since the capacity of a capacitor depends in the same way on D_0 and ε_s. Besides, a unique relation between D_0 and ε_s can be found from the experimental results [using Eq. (12), $A_{1\,\mathrm{exp}}$ and $A_{2\,\mathrm{exp}}$] according to which, knowing one of these parameters suffices to calculate the magnitude of the other. A plot of $\varepsilon_s(D_0)$, obtained from the present experiments, is shown in Fig. 6 for the range 5 Å$\leqslant D_0 \leqslant$8 Å. Experimental results available in the literature (see, for example, Refs. 13, 18, 19) and our independent measurements enable us to estimate the DP, averaged over the thickness, of the solid layer

$$\varepsilon_s=\frac{1}{D_0}\int_0^{D_0}\varepsilon_s(x)\,dx,$$

adsorbed on the metallic surface at $T=1.5$ K. If we take account of the estimate $\varepsilon_s = 1.0725 \pm 6\cdot 10^{-4}$ obtained in this way, the thickness of the solid layer D_0 should lie within the limits 6.7 Å$\leqslant D_0 \leqslant$7.4 Å, i.e., in the conditions of the present experiments it amounts to about two atomic layers of ^{4}He.

5. CONCLUSIONS

On the basis of these results we can conclude that a film of adsorbed ^{4}He is nonuniform and its DP depends on its thickness and on the parameters of the substrate. Theoretical estimates of the variable part of the DP of a film agree qualitatively with the experimental results, while the relations obtained [Eq. (11)] can be used to measure the thickness of a film and the substrate parameters by the capacitance method.

The variation in the distribution of the component of the DP normal to the metallic surface and the density of the helium film, and also the value found for the thickness of the solid layer are characterized by the values of α and Z averaged over the electrode surfaces. The small variations of α_{exp} and Z_{exp} along the surface, which could occur in all similar experiments, do not make a noticeable contribution to the measured values of $\varepsilon(l)$ and $\rho(l)$ for film thicknesses $l > D_0$. At the same time the effects of local variation in the substrate parameters must be taken into account in experiments to study the hydrodynamics of superfluid liquid in thin slits and films, since they can lead to hysteresis phenomena as a result of pinning of vortex lines and a shift in the temperature for nucleating vortices.

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