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SURFACE TENSION OF DILUTE He^3-He^4 *SOLUTIONS*

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The surface tension α of He^3-He^4 mixtures was measured, on the basis of capillary rise, in the temperature range 0.4–2.0° K for six concentrations $c = N_3/(N_3 + N_4) = 10^{-4}$, 1.2×10^{-3}, 5.6×10^{-3}, 5.2×10^{-2}, 9.6×10^{-2}, and 2.2×10^{-1}. A maximum appeared on the $\alpha(T)$ curve of each mixture except at $c = 10^{-4}$. The results support Andreev's hypothesis[13] that a new type of excitation, with a dispersion law $E = \Delta - \epsilon_0 + k^2/2\mu$, exists near the surface of a liquid He^3-He^4 mixture. The parameters of the spectrum are $\epsilon_0 = 1.7 \pm 0.2°$ K and $\mu = (0.9 \pm 0.1)M$, where M and Δ are the effective mass and the minimum energy of the impurity excitations in the bulk of the liquid.

INTRODUCTION

THE temperature dependence of the surface tension α of the pure helium isotopes has been very well studied.[1-9] As the temperature decreases from the critical point to zero a linear increase of α is observed at first: $\alpha(T) = \text{const} \times (T_{cr} - T)$. This is followed by a region of slower increase approaching saturation in accordance with the general laws of thermodynamics.

Esel'son et al.[10,11] have measured the surface tension of He^3-He^4 mixtures at the higher temperatures (1.2° K to the critical temperature). Their results show the similarity between the temperature dependences of α for the mixtures and the pure isotopes. The absolute surface tension of a mixture decreases as the He^3 concentration is increased, from the value α_4 for pure He^4 to α_3 for pure He^3.

It may be expected, however, that at lower temperatures the surface tension of the mixtures will exhibit a more complex form of temperature dependence than the pure isotopes. Since the addition of He^3 to the solution reduces α, the surface layer adsorbs an extra amount of He^3 (thus exceeding the bulk concentration c).[12] If the solution and adsorption from it are both weak we may write, approximately,

$$\alpha = \alpha_4 - n_s T, \tag{1}$$

where n_s is the excess amount of impurity per unit surface. If as the temperature is lowered n_s increases more rapidly than α_4/T, the $\alpha(T)$ curve should reach a maximum at some temperature. Atkins and Narahara[5] actually observed this maximum for a mixture with c = 0.05% at 0.6°K but attributed it to several extraneous effects. Andreev[13] later derived a function $n_s(T, c)$ theoretically for small amounts of the impurity and accounted for the maximum thereby.

It is known that at low concentrations an impurity atom in He^4 behaves like an excitation with the dispersion law

$$E = \Delta + p^2/2M, \tag{2}$$

where E and p are the energy and momentum of the excitation, while Δ and M are constants. ($\Delta \approx -2.7$°K was calculated by Andreev using the results of Sommers; $M \approx 2.4\, m_{He^3}$ according to the most recent data.[14])

Equation (2) holds true at large distances from the boundary of the liquid. Andreev suggested that for a He^3 atom near the surface of liquid He^4 there exist levels with a minimum energy below Δ. For He^3 atoms located in the surface levels the dispersion law is

$$E = \Delta - \varepsilon_0 + k^2/2\mu, \tag{3}$$

where $\epsilon_0 > 0$, **k** is the two-dimensional momentum in the plane of the liquid boundary, and μ is a constant of the order of M.

We thus find that He^3 atoms accumulate in the surface layer of the liquid, forming a kind of two-dimensional Fermi system. Calculating the excess surface density

n_s at temperatures much higher than the degeneracy temperature of this system, Andreev obtained the following expression for α:

$$\alpha = \alpha_4 - c\frac{\hbar\rho}{m}\frac{\mu}{M}\left(\frac{2\pi T}{M}\right)^{1/2} e^{\varepsilon_0/T} \tag{4}$$

with

$$c \ll \frac{m}{\rho}\left(\frac{MT}{\pi\hbar^2}\right) e^{-\varepsilon_0/T}. \tag{5}$$

Here ρ and m are the density and atomic mass of pure He^4, and $c = N_3/(N_3 + N_4)$ is the impurity concentration. The exponentially increasing term on the right-hand side of (4) produces a maximum in the temperature dependence of the surface tension.[1)]

By comparing Eq. (4) with the results given in [10] Andreev obtained approximate values of the parameters in (3) for the impurity excitations at the surface:

$$\varepsilon_0 \approx 3.1^\circ\,K, \qquad \mu \approx M.$$

Our aims in the present work were to investigate the surface tension of dilute He^3-He^4 solutions at low temperatures, to test the validity of Andreev's theory, and to evaluate the spectral parameters of the excitations at surface levels.

DESCRIPTION OF APPARATUS

The surface tension was determined from the height of rise in a capillary. A diagram of the apparatus, which was adapted for optical measurements at low temperatures, is shown in Fig. 1.

From the top of the apparatus three thin-walled stainless steel tubes, each 20 mm in diameter, extended to the lower part. The middle tube 8 was used for pumping from the He^3 reservoir for the purpose of cooling the mixture. The two side tubes 4 served for optical observations and were terminated inside the vacuum jacket of the apparatus by total reflection prisms 6.[2)] The movable mountings of the prisms permitted changes of their inclinations when adjusting the optical system. The lenses 3 formed vacuum seals at the tops of the optical tubes.

The lamp 1, which was located at the focus of a short-focus lens, provided a nearly parallel light beam. The image of the capillaries (in transmitted light) was projected on the rear wall of the camera (lacking an objective lens) by means of a long-focus lens (on the right) and two prisms. The entire optical system was darkened; the inner walls of the optical tubes were coated with soot in order to prevent reflections. Heat flow to the lower part of the apparatus was also reduced in this way.

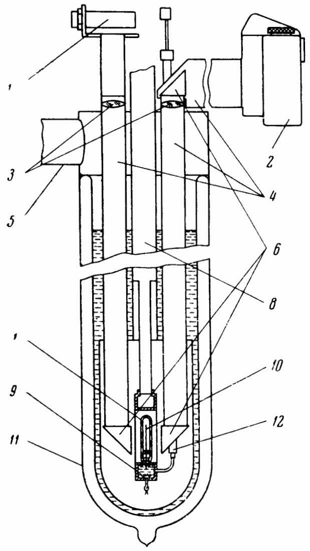

FIG. 1. Diagram of apparatus. 1 - flash lamp, 2 - camera, 3 - lenses of optical system, 4 - tubes of optical system, 5 - tube for pumping out He^4, 6 - prisms of optical system, 7 - ampoule of the reservoir of the mixture, 8 - tube for pumping out He^3, 9 - thermometer in the reservoir of the mixture, 10 - capillaries, 11 - helium Dewar, 12 - valve of mixture system.

In most of the experimental work (except in thermometer calibration) a He^3-He^4 mixture with c ~ 70% was pumped off for cooling, instead of pure He^3. A lowest temperature ~0.4°K was reached by circulation of this mixture. The temperature of the mixture was measured by means of a resistance thermometer made with 30-μ phosphor bronze, calibrated on the basis of pure He^3 vapor pressure, and incorporated in a potentiometer circuit. In all the experiments the maximum error of the mixture temperature was 0.005°K.

A cross section of the lower portion of the apparatus is represented in Fig. 2. The He^3 and mixture reservoirs were in good thermal contact through their common copper wall 1. Small columns of 1 × 1 mm cross section and 20 mm high were cut in the bottom of the He^3 reservoir for the purpose of improving temperature equalization. The upper termination of the mixture reservoir consisted of a glass ampoule 2, which was 60 mm

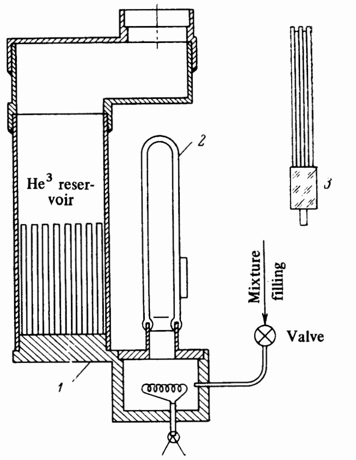

FIG. 2. Cross section of assembly of He^3 and mixture reservoirs (lower part of the apparatus).

[1)] Remark added December 12, 1968. Equation (5) ceases to be valid at the temperature of the maximum. As T decreases further the exponential decrease of α must slow down since $d\alpha/dT \to 0$ for $T \to 0$. Andreev also derived a law whereby α approaches saturation at very low temperatures (when the impurity excitations in the liquid are highly degenerate). In a private conversation Andreev kindly informed us of this result:

$$\alpha = \alpha_4 - \frac{\hbar^2}{2M}N_s\left(3\pi^2 c\frac{\rho}{m}\right)^{2/3} + \left(\frac{\pi m}{3c\rho\hbar^3}\right)^{2/3}\frac{MT^2}{6}N_s$$

where N_s is the total number of He_3 quantum states in the surface levels.

[2)] In [7,10] prisms were used for the first time to reflect a light beam out of apparatus used to investigate α.

high and 5 mm in diameter and held a plug 3 that supported a set of capillaries having different diameters (shown on the right-hand side of Fig. 2). Free entrance of liquid into the ampoule was provided by two holes of 1-mm diameter. The mixture was admitted to the reservoir through a 100-micron capillary which connected the reservoir to a shut-off valve near the bottom of the apparatus. The valve was operated by means of a rod that passed through the cover of the apparatus; the rod was raised after the gas had condensed. This design permitted reduction of the volume of gas and prevented additional heat conduction through the gas and rod. The thermometer was mounted at the bottom of the mixture reservoir, where it was shielded from stray radiation.

In the work with mixtures the entire copper portion of the reservoir and part of the glass ampoule were filled with the liquid phase of the solution. The gaseous phase never exceeded 0.6 cm^3 out of the total 2.5-cm^3 reservoir volume. Because of this volume ratio the relative change $\Delta c/c$ of the liquid phase did not exceed 0.02 below 2°K.

The apparatus included five capillaries ~50 mm long3) having inside diameters of 104.4, 116.2, 147.8, 156.0, and 364 μ and outside diameters from 0.6 to 0.8 mm. Experimental accuracy required that the diameter of each capillary be uniform along its entire length. Nonuniformity of a diameter was determined from the length of a mercury column moving along the capillary and measured to within a few microns by means of a horizontal comparator. In the selected capillaries the variation of the inside diameter was at most 0.3% in 1 cm of length. Absolute values of the diameters were computed to within 0.5% from the combined weight of several mercury columns having known lengths.

The capillaries scattered all light rays except those that traversed them close to the axis. Therefore in the field of view each capillary appeared as a dark band with a brightly illuminated middle stripe. Each meniscus in a capillary was seen as a dark band across the illuminated region, with shadowy "tails" in the darkened portion of the field. For thin capillaries this band was 30–40-μ wide; when the microscope hair was positioned over the middle of the band in a photograph the error of two readings was $\Delta h = \pm 40\ \mu$.

For the purpose of determining the magnifying power possessed by the optical system of the apparatus a glass rod of known length ($l = 8.33 \pm 0.01$ mm) was cemented to the wall of the ampoule; the magnification was found to be 1.56. (Each image then filled the entire camera frame area.)

EXPERIMENT

The surface tension of He^3-He^4 solutions was measured at temperatures from 0.4° to 1.9° K for six concentrations ranging from 1.0×10^{-4} to 0.22. For comparison the surface tension of pure He^4 was also measured. Samples were prepared by mixing known quantities of the pure isotopes. We estimate that the concentrations were determined with not worse than 1% accuracy. The molar concentrations will be mentioned here in values that have been rounded off.

Each mixture was fed to our apparatus from Töpler pumping apparatus. The filling of the mixture reservoir was monitored both with a manometer and visually by sighting through the frame opening of the camera. After the ampoule had been filled to the required level the valve of the mixture system was closed, the rod was lifted, and the connecting tubes were evacuated with the mercury piston and the diffusion pump of the Töpler apparatus.

In each experimental run $\alpha(T)$ was registered for the particular mixture while the temperature was reduced slowly from 2° to 0.4° K. Prior to each measurement the temperature of the reservoir was held constant for the time required to establish both temperature and concentration equilibrium of the mixture. This time varied from 20–30 min in the 1.4–1.8° K range to 2–3 min at the lowest temperatures, because of the different diffusion rates existing in He^3-He^4 mixtures at high and low temperatures[15] and the consequently different speeds with which equilibrium was established in the reservoir. A few photographs served to monitor the heating of the apparatus. When the heating rate did not exceed 0.02 deg/min the measurements obtained during heating and cooling were in agreement.

Two independent experimental runs were completed for each concentration of the mixture. All points for a given concentration were well fitted by a single curve.

Several control experiments were performed to evaluate the extraneous heating of the capillaries by radiation from the lamp and from bodies at room temperature. Illumination was furnished by an IFK-20 xenon flash lamp (20-J, 200-μsec flashes). This type of lamp caused no illumination effect; there was no perceptible change in the capillary level during a flash. At 0.4° K the thermometer registered an increase of only 0.002° K, and the previous temperature was restored within a few seconds. At higher temperatures the thermometer indicated no reaction to the flashes. For comparison, measurements were obtained for pure He^4 illuminated continuously by a 4-W incandescent lamp. At high temperatures the liquid rise h was found to be reduced due to heating of the vapor, while below 0.9° markedly elevated measurements were obtained in narrow capillaries because of the thermomechanical effect.

We did not completely prevent heating due to radiation from bodies at room temperature. On the basis of the thermomechanical effect in pure He^4 we estimated that at the lowest temperature (0.44° K) the maximum temperature increase was 0.008° for a 116-μ capillary and 0.003° for a 148-μ capillary. This extra heating of the mixtures reduced the He^3 concentration in the capillaries by an amount ΔC determined from the equation $(kT/m_3)\Delta c = -\sigma_0 \Delta T$, where m_3 is the mass of a He^3 atom, k is Boltzmann's constant, and σ_0 is the entropy of pure He^4. A calculation for the worst case indicated $\Delta c \approx 3 \times 10^{-6}$, which is insignificant for all the experimental concentrations.

To determine whether steady thermal flow in the reservoir had affected the calibration of the thermometer, a special calibration was performed. Both optical tubes were sealed with copper plugs that were in contact with a helium reservoir. This calibration agreed completely

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3)The central portion (20 mm long) of each capillary was in the field of view; the maximum rise of the liquid was 10 mm.

with the calibration performed under the usual conditions.

TREATMENT OF DATA. DISCUSSION OF RESULTS

Each measurement (each photographic frame) furnished four independent values of the surface tension. (In most instances the level in the 104.4-μ capillary was not observed because of poor illumination.) For the widest (364-μ) capillary the results were in most cases less accurate than for the remaining three capillaries, because of the small rise h and the large dark band at the location of the level. We therefore considered as the true value of α the arithmetic mean of the three values for the narrower capillaries (116.2, 147.8, and 156.0 μ); the spread of these values was at most ± 0.5%.

In calculating α from the difference between the capillary rises of a liquid in cylindrical tubes it is customary to use the equation

$$g(\rho_l - \rho_v)h = 2\alpha(1/b - 1/B), \tag{6}$$

where g is the acceleration of free fall, ρ_l and ρ_v are the densities of the liquid and vapor, h is the distance between the lowest points of the menisci, b and B are the radii of curvature at the lowest points of the menisci in two tubes of radii r and R. Corrections for the discrepancies between values of b or B and r or R, respectively, were calculated from Sugden's tables.[16]

In our experimental work the widest tube was the ampoule (R = 2.5 mm) within which the five capillaries were mounted. The presence of the barrier created in this way induced an additional rise of the liquid in the ampoule, which must be taken into account. The correction γ was determined in a special experiment by comparing the heights of the menisci in a U-shaped tube with a set of capillaries mounted in one of the arms. For this purpose ethyl alcohol and acetone were used at room temperature; the linear dimensions of the tubes were enlarged accordingly. The formula for α including the correction is

$$\alpha = \frac{1}{2} g(\rho_l - \rho_v)\left(\frac{H}{K} + \gamma\right)(r + \delta). \tag{7}$$

Here H is the height of the capillary rise on the negative, K is the magnification of the optical system, and δ was calculated from the relation $1/(r + \delta) = 1/b - 1/B$.[4]

The density of the liquid was calculated from

$$\rho_l = c\rho_3 + (1 - c)\rho_4, \tag{8}$$

where the densities of the pure isotopes, ρ_3 and ρ_4, were taken from [18]. The experimental densities of the solutions[19-21] are somewhat lower than those calculated from (8); however, for the concentrations used in the present work the difference was at most 1% (and was considerably less for low concentrations). The calculations based on (8) yielded values of ρ_l that are practically independent of temperature below 1.9° K; these values of ρ_l are given in the table. A density change could be evaluated from the change of the level in the ampoule. In this way it was shown that the densities actually remained unchanged to within ± 0.5%.

T, °K	He^4, $\rho_l = 0{,}145$ *		$c = 1{,}2\cdot10^{-3}$, * $\rho_l = 0{,}145$		$c = 5.6\cdot10^{-3}$, $\rho_l = 0.145$		$c = 5{,}2\cdot10^{-2}$, $\rho_l = 0{,}142$		$c = 9{,}6\cdot10^{-2}$, $\rho_l = 0{,}139$		$c = 2{,}2\cdot10^{-1}$, $\rho_l = 0{,}131$	
	$\rho_v\cdot10^3$	α	$\rho_v\ 10^3$	α	$\rho_v\cdot10^3$	α	$\rho_v\cdot10^3$	α	$\rho_v\cdot10^3$	α	$\rho_v\cdot10^3$	α
0,45		(0,375) **		0,367		0.346		0,270		0.230		—
0,5		(0.375)		0,371		0.352		0.278		0,239		0.176 ***
0,6		(0.375)		0.373		0.359		0.292		0,253		0.194
0,7		0,374		0.373		0.363		0.301		0,264		0,207
0,8		0,373		0.372		0,364		0.309		0.273		0.218
0,9		0.371		0.370		0,364	0.5	0,314	0,5	0,281	0.5	0,225
1.0		0.369		0,368	0.5	0.364	0,5	0.318	0,5	0,286	0.5	0,230
1.1		0,367		0,366	0,5	0,363	0,5	0,320	0.5	0,290	0,5	0.235
1.2		0,364		0.363	0.5	0,360	1,0	0,321	1.0	0,292	1,0	0,238
1,3		0,361		0.360	0.5	0.357	1,0	0,320	1,0	0,293	1.0	0,240
1.4		0,357		0,356	0.5	0.353	1,0	0.319	1,5	0,293	1.5	0,240
1,5		0.353		0.352	0,5	0,349	1,0	0,317	1,5	0,291	1.5	0,240
1,6		0,348		0,347	0.5	0,344	1,5	0,314	1.5	0,288	1.5	0,239
1,7	0.5	0.343	0.5	0,342	0,5	0,339	1,5	0,309	2.0	0,285	2,0	0,238
1,8	0,5	0,337	0,5	0,336	0,5	0,333	1,5	0,304	2,0	0,280	2,0	
1,9	0,5	0,330	0,5	0,329	1,0	0,326	1,5	0,298	2,0	0,274	2,5	

*The densities ρ_l and ρ_v are given in g/cm^3, and α in erg/cm^2.

**Extrapolated values are given in parentheses.

***At T = 0.505°K.

The calculations also took into account the vapor density above the solution. Above 1.4° K the vapor density was calculated from the known vapor densities of the pure isotopes[22,23] assuming that these are additive. (The He^3 concentration in the vapor was calculated by interpolations in the data of [24].) Below 1.4° K the vapor consists practically entirely of He^3; consequently the vapor pressure of pure He^3 was taken for the correction. Since the correction is very small the accuracy required for the calculation of α is satisfied by this crude approximation.

Our results are shown in Fig. 3. Above 0.7° K the surface tension of pure He^4 coincided completely with that of a mixture having the concentration $c = 1.0 \times 10^{-4}$; the identical points are indicated by small squares. Below 0.7° K the results in these two cases were distorted by the thermomechanical effect (the capillaries were heated somewhat by radiation along the optical tubes). These results were not taken into account, and the curve of $\alpha_4(T)$ below 0.7° K was based on extrapolations of the high-temperature data (the dashed segment of the curve). Extrapolation to T = 0 yielded $\alpha_4(0) = 0.375$ erg/cm^2, agreeing with the results in [5] within error limits. For mixtures with higher concentrations the phonon mean free path decreases through scattering by the impuriities and the thermomechanical effect vanishes. For all the other mixtures the thermomechanical effect was not observed experimentally down to the minimum temperatures.

The figure shows that for all concentrations beginning with $c = 1.2 \times 10^{-3}$ the maximum surface tension is temperature dependent. The dependence of α on T and c is represented in the table on the basis of smooth curves plotted through the experimental points. The table also mentions the liquid and vapor densities used in the calculations. The deviations from the smooth curves are at most ± 0.5%; the possible systematic error is estimated to be 2%.

For a comparison with Andreev's theory it is convenient to use the logarithmic coordinate

$$\ln\frac{\alpha_4 - \alpha}{\sqrt{T}} = A + \ln c + \frac{\varepsilon_0}{T}, \tag{9}$$

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[4] In our report at the XIth International Conference on Low Temperature Physics the corrections γ and δ were not included in the calculation. Consequently the absolute values of the surface tension were slightly too low (from 1.5 to 3%, depending on the value of α.)

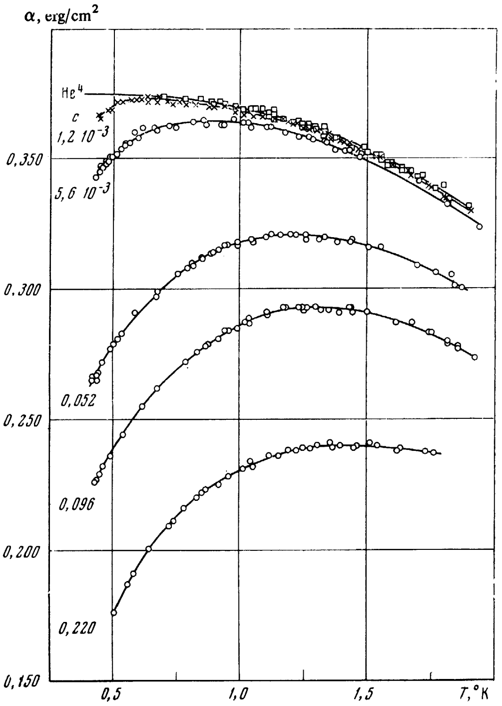

FIG. 3. Surface tension of He^3 - He^4 mixtures as a function of temperature. The concentration c is indicated at the beginning of each curve.

where A = ln ($\hbar\rho\mu/mM$) is a constant that depends on the effective mass μ of an excitation. The theoretical dependences in the coordinates ln $[(\alpha_4 - \alpha)/\sqrt{T}]$ and 1/T are a family of parallel straight lines.

Figure 4 shows the results in the new coordinates. The values of α_4 were taken from the smooth curves in Fig. 3. Vertical bars indicate the mean statistical error for some points. Arrows designate the degeneracy temperatures of impurity excitations in surface levels. The degeneracy temperature is below 0.4° K for a mixture having the concentration 1.2×10^{-3}.

The same figure shows that the predicted dependence holds true, within experimental error limits, from the highest temperatures to the degeneracy temperature of a surface impurity. For the highest concentration (c = 0.22) all the experimental points lie below the degeneracy temperature, although a few of the last points lie on a straight line parallel to the straight lines for the other concentrations in the region where Andreev's theory is applicable.

The slope of the curves in Fig. 4 enables us to calculate $\epsilon_0 = 1.7-0.2°$ K, and the vertical separation of the straight lines leads to the excitation effective mass μ. The last calculation requires that we know the concentration dependence of the effective mass M for a bulk impurity excitation, in a large range of concentrations. The requisite data were recently obtained in Kharkov.[25] Using this dependence for the first four concentrations we obtained $\mu = (0.9 \pm 0.1)$M. For c = 0.22 this calculation yields $\mu = 0.6$ M, presumably because this is not a dilute solution.

Our results confirm Andreev's hypothesis that in dilute He^3-He^4 solutions there exist localized impurity excitations near the surface with a dispersion law that differs from the dispersion in the bulk of the solution. When we know the spectral parameters ϵ_0 and μ of these excitations we easily calculate their density n_s. The calculations show that at temperatures greatly exceeding the degeneracy temperature only an insignificant amount of He^3 is adsorbed by the surface. For example, at 2° K, which is four times higher than the degeneracy temperature, for $c = 5.6 \times 10^{-3}$ we have $n_s \sim 10^{13}/\text{cm}^2$, which is only 2.5% of the density of a monomolecular He^3 film. However, with decreasing temperature n_s increases exponentially and near the degeneracy temperature it becomes comparable with the density of a monomolecular He^3 film. With further reduction of T we find that the increase of n_s is slowed, as is evident from the slower decrease of the surface tension (Fig. 4). At still lower temperatures a monolayer of He^3 evidently is formed on the surface of the liquid and n_s approaches the saturation value N_s. An investigation of α in this region is of considerable interest, since we are thereby able to determine the number of N_s of He^3 quantum states in surface levels.

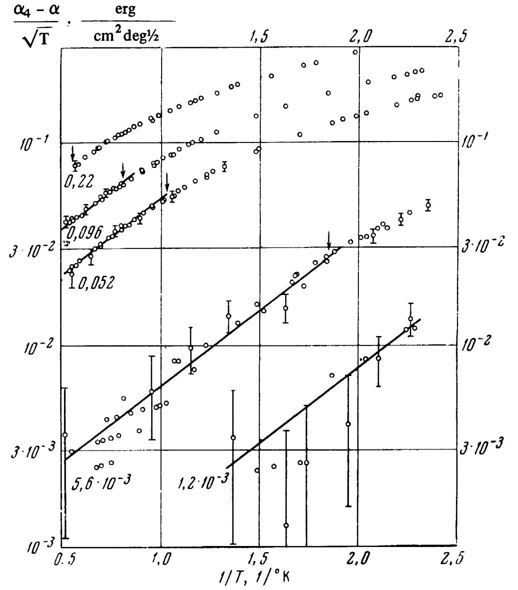

FIG. 4. Inverse temperature dependence of log $[(\alpha_4 - \alpha)\sqrt{T}]$.

In conclusion, we wish to thank P. L. Kapitsa for his attention and interest, A. F. Andreev and V. P. Peshkov for valuable discussions and comments, and V. N. Krutikhin for experimental assistance.

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