

EXPERIMENTAL RESEARCH WITH He³

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METHODS OF SEPARATION OF He³

THE helium isotope of mass 3, He³, occurs under natural conditions as a small impurity to the principal isotope He⁴. Its concentration is at most 10⁻⁴% in helium extracted from the atmosphere and from 10⁻⁶% to 10⁻³% in helium obtained from wells and minerals.¹ There are no other isotopes, with the exception of He⁶, which has a half-life of 0.82 sec.²

The helium obtained artificially from nuclear transformations in atomic reactors (usually from the reaction $Li^6 + n \rightarrow He^4 + H^3 \rightarrow He^4 + He^3 + e^-$) also consists of a mixture of He⁴ and H³; hence, for a study of the properties of He³, we must first separate it from mixtures with He⁴. This problem also appears in subsequent stages of investigation, since it is necessary for the experimenter to be able to prepare mixtures for his own purposes, and to separate them again. McInteer et al.³ applied the thermal diffusion method to the enrichment of He³. Their apparatus consisted of three successively connected thermal diffusion columns of different diameters, with a total length of 10 m. With a supply of 16.6 quarts of helium, containing 1.5 × 10⁻⁵% He³, they obtained 14 cm³ per day of helium (p = 760 mm mercury, T = 0°C) containing 0.21% He³.

After it had been established that He³ does not become superfluid,⁴ a number of low-temperature methods more effective than thermal diffusion were devised for isotopic separation.

Lane, Fairbank, Aldrich, and Nier⁵ proposed a heat flow method for enrichment of He³. Such a method has been used by a number of researchers, who obtain⁶ an enrichment of 4000 in the treatment of 0.2 l of liquid helium per cycle, 3 × 10⁴ in the treatment of 0.7 l⁷ and 10⁵ in the treatment of 0.75 l of liquid helium.⁸ In the last case, a mixture with a concentration of 0.2% of He³ was obtained from an initial mixture containing 10⁻⁶%.

Another more reliable method is also based on the fact that He³ does not take part in superfluid motion. In this case, He⁴ is drawn off as a film or by filters of the superfluid as a result of the thermomechanical effect; that is, use is made of the effect of normal flow. This method was pro-

posed by Pollard and Davidson⁹ and has been used by many authors.¹⁰⁻¹⁴ The method provided an enrichment from several hundred fold up to 3000, and permits one to obtain mixtures with concentrations from 10⁻³ to 10⁻²% He³. Such a method⁸ gave excellent results with the use of the apparatus pictured in Fig. 1.

Liquid helium flows continuously into the outer Dewar from a liquefier and is maintained at atmospheric pressure. Moreover, it enters the inner container through a regulating valve and a copper capillary, being cooled at entrance to 2° K. By thermal flow, the He⁴ is brought down through the filter F from the inner container, in an intermediate container; then the helium vapors are cooled by pumps, as a result of which the temperature is maintained at T = 2° K in the intermediate and inside containers. The fraction remaining in the internal container is enriched in He³ by a factor of 20,000 and is drawn off by the internal tube.

The yield of the apparatus in relation to the initial helium (about 3 to 4 liters of liquid helium per hour) is determined by the power of the pumps

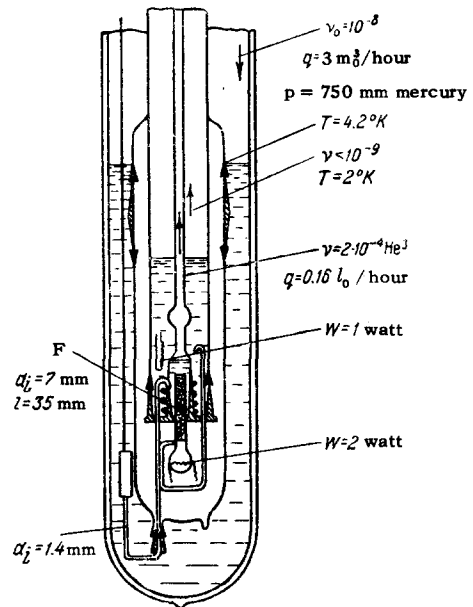


FIG. 1. Apparatus for the enrichment of He³ by means of thermal flow and rectification.

which cool the helium for the intermediate container. The parameters characterizing the operating regime are shown in Fig. 1.

It is significant that it is impossible to enrich He³ to very high concentrations by thermal flow. In the first place, as measurements have shown,^{15,16,17} the λ -transition is displaced with increase in the concentration, in the direction of lower temperatures. Thus, for a 50% solution of He³ in He⁴, superfluidity fails at 1.4° K. Moreover, at not very high concentrations of He³, the relation given by Pomeranchuk¹⁸ for thermal flow,

$$-S dT + \frac{dp}{\rho} - \frac{k}{m_4} d(xT) = 0, \quad (1)$$

is valid, where S is the entropy of one gram of He⁴, ρ the density, p the pressure, m_4 the mass of the He⁴ atom, k Boltzmann's constant, and x the molar concentration of He³. The entropy of He II in the temperature region above 1° K can be expressed by the equation $S = 0.405 (T/2.19)^{5.5}$ cal/gm-deg., i.e., $SdT = (1/6.5) d(ST)$. The density of He II changes but little; therefore the entire expression can be written in the form

$$d \left\{ \left(\frac{kx}{m_4} + \frac{S}{6.5} \right) T - \frac{p}{\rho} \right\} = 0$$

or along the path of thermal flow,

$$\left(\frac{kx}{m_4} + \frac{S}{6.5} \right) T - \frac{p}{\rho} = \text{const.} \quad (2)$$

Since the concentration of He³ in its enrichment by thermal flow is at the same time virtually equal to zero, we have

$$\left(\frac{kx}{m_4} + \frac{S}{6.5} \right) T - \frac{p}{\rho} = \frac{S_0 T_0}{6.5} - \frac{p_0}{\rho}$$

or

$$x = \frac{m_4}{kT} \left(\frac{S_0 T_0 - ST}{6.5} - \frac{p_0 - p}{\rho} \right). \quad (3)$$

The curve of λ -transitions, and also the value of x , calculated from the given formula for different T , are plotted in the graph of Fig. 2. As is seen, thermal flow cannot go to very high concentrations. The theoretical coefficient of separation of He³ for superfluid filtration can be estimated from the following considerations. If we assume that the superfluid flow and the diffusion take place through the same effective cross section of the filter, while the velocity of the superfluid flow is limited by the critical velocity $v_s = 20$ cm/sec, then the value of the superfluid flow across a unit surface area is equal to

$$\omega_1 = \rho_s u_s,$$

and the flow of He³ determined by diffusion will be

$$\omega_2 = \frac{D \rho m_3 x_0}{m_4 l},$$

where D is the diffusion coefficient of He³, l the length of filter, ρ_s the density of the superconducting part of the helium, and x the molar concentration in front of the filter; m_3 and m_4 are the masses of the He³ and He⁴ atoms, respectively.

The molar concentration of the transient helium is determined by the expression

$$x = \frac{\omega_2 m_4}{\omega_1 m_3} = \frac{D \rho x_0}{l \rho_s v_s},$$

i.e., the limiting value of the separation coefficient will be

$$A = \frac{x_0}{x} = \frac{l \rho_s v_s}{\rho D}. \quad (4)$$

The coefficient of diffusion of He³ in liquid helium II was determined experimentally by Beenakker et al.¹⁹ It was shown to decrease rapidly from 10^{-2} cm²/sec at 1.3° K down to 10^{-3} cm²/sec at 1.6° K and 4×10^{-5} cm²/sec at 2.1° K. For a filter length $l = 5$ cm, an estimate of the separation coefficient A by Eq. (4) for various temperatures leads to the following results: at 1.3° K, $A = 10^4$; at 1.6° K, $A = 8 \times 10^4$; at 2.0° K, $A = 6 \times 10^5$ and at 2.1° K, $A = 5 \times 10^5$, i.e., the separation coefficient has a maximum around 2° K. In practice, because of interruptions in the process, when diffusion continues while there is superfluid flow, and also because of incomplete filtration, the separation actually obtained is smaller. Moreover, at high concentrations, the fraction of the superfluid component decreases because of the coupling to it of atoms of He³, the volume velocity of the

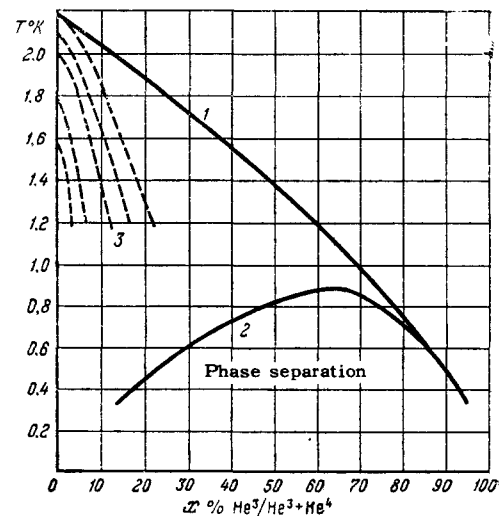


FIG. 2. 1 - Curve of the λ -transitions; 2 - Curve of phase separation, 3 - $\left(\frac{kx}{m} + \frac{S_0}{6.5} \right) T - \frac{p}{P} = \text{const}$ for different T_0 .

superfluid motion falls off, and the effectiveness of the process decreases. The latter circumstance has been confirmed in the research of Kuznetsov.²⁰ In Fig. 3, the dependence of the output of the filter on the concentration of the enriched liquid is plotted at different temperatures. Thus, examination of the method of enrichment of He³ by thermal flow shows that it is very effective for increasing He³ from dilute solutions, but is scarcely suitable for the enrichment of solutions with concentrations of more than 5% He³.

It has been possible to enrich solutions of higher concentrations of He³ by rectification.^{14,20,8,21} Such rectification is particularly effective for separation of isotopes of helium, since the relative difference in the vapor pressures of He⁴ and He³ is very great.

The phase diagram of the liquid-vapor system of the isotopes of helium (He³-He⁴) has been investigated by a number of authors.^{22,23,24,25} The most complete data have been obtained by Esel'son and Bereznyak.²⁵ Their data, shown in Tables I, II, and III, agree within the limits of accuracy of measurement with the results of Sommers²⁴ who carried out measurements in a very narrow range of concentrations, and with the data of Peshkov and Kachinskiy²⁶ for higher concentrations. The phase diagram of the liquid-vapor system of the isotopes of helium at different pressures calculated by Kuznetsov²⁷ from the data of Esel'son and Bereznyak²⁵ has been plotted in Fig. 4.

As has been already remarked earlier, rectification of the isotopes of helium is a very effective method of separation. Estimates of the efficiency of the rectifying column in reference 28 leads to the following results.

The distribution of concentration of helium⁴

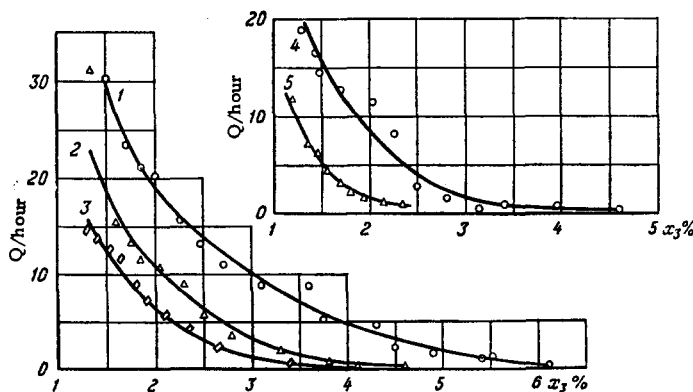


FIG. 3. The dependence of the yield of the filter Q on the concentration of enriched liquid for various temperatures. Curve 1: 1.43° (solution) - 2.15° (He⁴ tank); curve 2: 1.43° - 2.02°; Curve 3: 1.43° - 1.86°; Curve 4: 1.86° - 2.15°; Curve 5: 1.86° - 2.02° K.

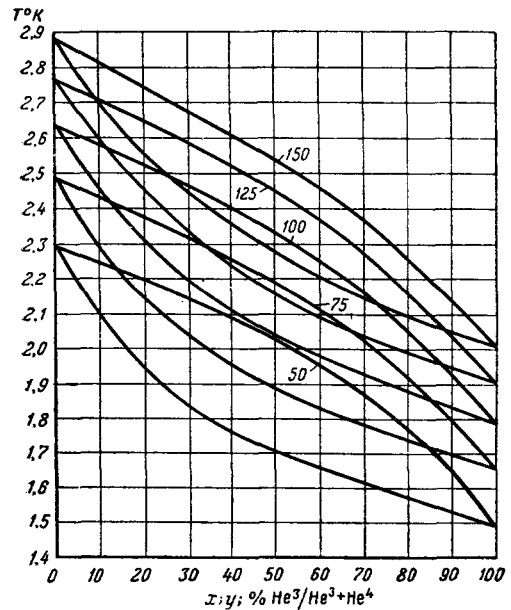


FIG. 4. Liquid-vapor phase diagram of the isotopes of helium at various vapor pressures. The numbers on the curves indicate pressure in mm of mercury.

along the column of liquid phase in the unseparated regime is determined by the formula

$$\frac{x}{(1-x)^{\frac{1}{\alpha}}} = \frac{x_0}{(1-x_0)^{\frac{1}{\alpha}}} e^{\beta z} \tag{5}$$

for the condition $\frac{x_a}{1-x_a} = \frac{\alpha y_a}{1-y_a}$. Or, at the beginning of separation, $x_a = \alpha y_a$, which, for helium in the temperature range 1.5° K to 1.9° K, is valid up to $x = 0.5$ for $\alpha = 3$,

$$x = x_0 \frac{(\alpha-1)e^{\beta z} - \alpha\gamma}{\alpha-1-\alpha\gamma} \tag{6}$$

In these expressions, for turbulent regime in the column,

$$\beta = \frac{\alpha(1-\gamma)-1}{12\alpha(1-\gamma)a} \left(\frac{D_v}{av_0} \right)^{0.2} \tag{7}$$

The entrainment in the column is given by

$$q = \frac{px_1}{\beta l} \tag{8}$$

If the gas with a high concentration of He³ is poured off and the effectiveness of the column is large ($e^{\beta l} \gg 1$), the mean concentration of the extracted gas will be

$$x = \frac{\left(1 - \frac{\alpha\gamma}{\alpha-1}\right) Q e^{-\beta l}}{\alpha p_0} \ln \frac{x_2}{x_1} \tag{9}$$

The time of establishing in the column a condition after which one can begin separation is equal to

$$\tau_0 = \frac{4\alpha p}{(\alpha-1)W} \tag{10}$$

TABLE I. Dependence of the vapor pressure (p, mm mercury) on the temperature (T°, K) for various compositions of the liquid phase (X_l, %)

X _l	T	p	X _l	T	p	X _l	T	p	
0.4	1.377	2.22	1.9	3.073	211.98	8.3	2.144	52.49	
	1.387	2.35		3.217	257.23		2.172	55.67	
	1.489	3.84					2.187	58.40	
	1.594	6.01		3.0	1.377		4.79	2.207	60.45
	1.675	8.26			1.387		4.90	2.248	65.67
	1.755	11.12			1.549		8.36	2.402	87.77
	1.836	14.68			1.572		9.23	2.565	116.39
	1.918	19.04			1.644		11.34	2.720	148.91
	1.993	23.80			1.721		14.23	2.881	189.59
	2.070	29.36			1.836		19.62	3.023	230.39
	2.124	33.67	1.977		28.23	3.206	291.15		
	2.172	38.00	2.053		33.84	3.219	296.01		
	2.201	40.94	2.093		36.94				
	2.301	51.56	2.102	37.80	11.1	1.354	10.32		
	2.406	64.80	2.144	41.16		1.363	10.94		
	2.554	87.05	2.175	44.10		1.461	14.19		
	2.694	112.15	2.197	46.72		1.467	14.58		
	2.793	133.11	2.249	52.30		1.542	17.27		
	2.902	158.09	2.348	64.50		1.635	21.32		
	3.051	197.55	2.460	80.45		1.730	26.16		
	3.104	213.54	2.513	88.93		1.842	32.96		
	3.202	243.60	2.635	110.63		1.936	39.42		
	3.211	246.60	2.752	134.73		2.015	44.96		
	0.8	1.358	2.46	4.0	2.850	157.33	2.020	45.62	
		1.563	5.87		2.979	190.23	2.086	51.88	
		1.634	7.71		3.124	233.17	2.125	56.13	
		1.763	12.21		3.197	256.64	2.148	58.71	
		1.815	14.50		1.358	5.32	2.180	62.49	
		1.927	20.46		1.358	5.31	2.200	65.97	
		2.008	25.76		1.495	8.16	2.298	79.37	
		2.105	33.03		1.559	9.79	2.476	108.64	
		2.126	34.85		1.655	12.91	2.699	154.57	
		2.153	37.15		1.745	16.56	2.796	178.77	
		2.165	38.26	1.855	22.06	2.904	207.62		
		2.171	38.86	1.855	22.06	3.023	243.69		
2.175		39.17	1.939	27.23	3.215	309.33			
2.175		39.17	2.039	34.42					
2.181		39.78	2.039	34.42	13.4	1.342	11.63		
2.188		40.74	2.120	40.89		1.399	12.74		
2.208		42.83	2.169	45.47		1.469	15.99		
2.248		46.89	2.191	48.07		1.553	19.64		
2.380		62.66	2.202	49.34		1.628	23.15		
2.449		72.32	2.254	55.06		1.725	28.48		
2.548		87.58	2.397	73.81		1.807	33.34		
2.748		125.10	2.468	84.34		1.844	35.89		
2.815		139.69	2.485	87.29		1.944	42.87		
2.900		159.38	2.549	97.93		1.995	46.88		
2.902		160.04	2.671	120.46	2.045	51.43			
3.003		186.35	2.799	148.66	2.087	56.30			
3.050		199.43	2.950	186.04	2.122	60.15			
3.052		199.82	3.105	231.36	2.156	64.07			
3.167		234.73	1.358	6.96	2.181	67.19			
3.202		245.97	1.489	10.11	2.199	70.59			
1.9		1.358	3.53	6.3	1.618	14.30	2.301	85.25	
		1.377	3.75		1.718	18.42	2.409	103.12	
		1.495	5.86		1.828	24.08	2.501	119.52	
		1.572	7.73		1.958	32.30	2.600	140.17	
		1.668	10.59		2.008	35.92	2.701	163.41	
	1.743	13.33	2.159		48.91	2.802	189.58		
	1.815	16.61	2.183		51.49	2.884	212.07		
	1.877	19.88	2.194		53.08	3.063	267.83		
	1.978	26.16	2.258		60.30	3.215	321.79		
	2.037	30.26	2.336		70.54				
	2.102	35.36	2.428	84.13	16.7	1.388	14.86		
	2.149	39.39	2.525	100.11		1.463	17.84		
	2.163	40.59	2.603	114.57		1.496	19.23		
	2.177	41.91	2.665	126.88		1.596	23.86		
	2.203	44.98	2.701	134.60		1.701	29.80		
	2.223	47.13	1.352	8.10		1.771	34.01		
	2.298	55.41	1.358	8.88		1.849	39.14		
	2.424	71.83	1.483	12.55		1.896	42.53		
	2.533	88.68	1.598	16.77		1.930	45.06		
	2.608	101.76	1.696	21.05		1.955	47.15		
	2.696	118.59	1.851	29.48	1.980	49.79			
	2.740	127.95	1.983	38.52	2.037	55.60			
	2.790	138.89	2.085	46.53	2.107	63.35			
	2.914	168.08	2.107	48.71	2.138	67.25			
					2.174	71.79			

TABLE I. Continued

X_I	T	p	X_I	T	p	X_I	T	p	
16.7	2.189	75.00	23.9	2.184	85.28	52.7	2.009	97.14	
	2.213	78.15		2.198	88.61		2.054	105.32	
	2.298	90.91		2.279	100.87		2.106	115.41	
	2.384	106.13		2.458	138.48		2.163	127.30	
	2.530	134.41		2.601	171.54		2.193	136.18	
	2.656	162.07		2.754	213.45		2.254	150.86	
	2.805	200.80		2.885	253.67		2.330	171.00	
	2.900	228.16		3.035	305.70		2.390	186.91	
	3.028	269.00		3.206	372.35		2.474	211.68	
	3.155	314.27					2.540	232.01	
	3.219	339.33					2.617	257.47	
19.2	1.360	14.61	30.2	1.316	16.23	56.3	2.255	157.26	
	1.379	15.59		1.341	17.11		2.352	181.33	
	1.474	19.19		1.380	18.70		2.448	208.82	
	1.558	23.60		1.423	21.59		2.535	235.80	
	1.650	28.57		1.485	24.60		2.629	267.32	
	1.706	32.10		1.537	27.80		2.703	294.96	
	1.752	35.06		1.597	31.24		2.774	322.21	
	1.839	41.04		1.649	34.40				
	1.883	44.91		1.722	39.73		73.4	1.400	27.27
	1.930	48.58		1.735	40.71			1.411	26.52
	1.974	53.62		1.784	45.58			1.422	26.89
2.027	59.13	1.830	49.96	1.515	38.52				
2.081	65.43	1.911	58.94	1.596	48.30				
2.128	71.07	1.972	66.74	1.691	61.06				
2.175	77.42	1.992	69.14	1.806	79.06				
2.233	87.67	2.069	79.85	1.897	95.09				
2.278	94.66	2.126	88.03	2.007	117.08				
2.376	111.85	2.170	95.37	2.108	140.19				
2.469	129.86	2.202	102.22	2.152	150.56				
2.571	152.18	2.218	105.97	2.177	157.95				
2.688	180.74	2.302	121.12	2.194	162.98				
2.799	211.32	2.375	135.95	2.257	181.08				
2.900	241.21	2.508	166.50	2.306	195.48				
2.999	273.69	2.655	205.91	2.404	226.82				
3.107	312.17	2.773	241.97	2.499	259.30				
3.202	348.09	2.878	276.70	2.606	298.89				
22.6	1.336	15.14	38.3	1.331	18.99	82.4	1.380	28.56	
	1.353	15.68		1.334	18.95		1.392	30.15	
	1.460	20.74		1.389	21.76		1.490	41.09	
	1.502	22.80		1.422	23.21		1.595	53.71	
	1.554	25.64		1.471	25.80				
	1.646	30.52		1.543	30.11		1.694	68.86	
	1.704	33.99		1.570	32.27		1.808	88.66	
	1.802	40.58		1.641	37.91		1.894	105.38	
	1.831	42.53		1.715	44.94		1.996	128.22	
	1.864	45.14		1.757	49.06		2.085	149.67	
	1.905	48.88		1.804	53.89		2.176	172.34	
1.968	55.53	1.857	60.38	2.199	182.17				
2.007	59.75	1.949	72.69	2.256	199.37				
2.068	66.98	2.046	86.79	2.308	216.63				
2.101	71.32	2.090	94.65	2.400	248.31				
2.126	74.71	2.141	102.24	2.504	287.36				
2.173	81.47	2.170	107.71	2.603	328.42				
2.202	86.68	2.194	113.66	2.695	369.74				
2.294	101.82	2.199	114.78						
2.414	124.29	2.253	125.72	90.8	1.392	31.26			
2.474	136.54	2.306	137.19		1.405	32.49			
2.552	154.02	2.331	142.88		1.507	45.32			
2.611	168.17	2.408	161.34		1.601	58.41			
2.719	196.36	2.488	181.52		1.678	70.71			
2.809	222.64	2.554	199.62		1.809	94.55			
2.906	252.03	2.604	213.13		1.906	115.67			
3.052	301.99	2.680	237.15		2.004	138.61			
3.136	333.87	2.757	262.36		2.097	163.04			
3.211	363.31	2.856	296.94		2.139	175.30			
23.9	1.336	15.54	52.7		1.350	21.06	2.182	186.34	
	1.362	16.39		1.356	21.83	2.193	192.94		
	1.406	18.21		1.437	26.70	2.259	214.74		
	1.498	22.65		1.471	28.94	2.303	229.99		
	1.579	27.15		1.519	34.15	2.395	263.90		
	1.710	35.07		1.562	36.58	2.507	309.00		
	1.806	41.82		1.592	40.29	2.586	343.55		
	1.826	43.10		1.652	45.84	2.648	371.96		
	1.873	47.81		1.686	49.87				
	1.908	51.12		1.728	54.79				
	1.948	54.97		1.803	64.71				
2.038	65.46	1.877	75.24						
2.149	80.07	1.944	85.94						

TABLE II. Dependence of the pressure (p , mm mercury) at the onset of condensation upon the temperature (T° , K) for mixtures with various amounts of helium 3 (X_V per cent)

X_V	T	p	X_V	T	p	X_V	T	p
1.9	2.037	26.4	29.0	1.986	31.5	57.6	1.963	47.8
	2.166	37.2		2.349	78.4		1.998	51.4
	2.369	59.7		2.533	112.7		2.101	66.5
	2.520	81.5		2.676	146.3		2.444	137.4
	2.776	129.3		2.826	187.0		2.652	204.4
	3.033	193.5		3.012	248.3		2.825	267.4
4.0	1.748	10.4	35.4	1.404	3.3	73.4	1.451	9.5
	2.077	30.2		1.836	21.9		1.691	29.6
	2.411	66.4		2.044	40.2		1.865	51.3
	2.732	122.3		2.159	54.4		2.254	127.1
	3.040	198.7		2.298	76.4		2.489	199.3
				2.516	121.1		2.731	294.8
11.6	1.350	1.9	35.4	2.706	167.6	82.4	1.412	16.1
	1.795	14.0		2.808	197.0		1.611	30.3
	2.131	38.0		2.899	225.7		1.818	59.0
	2.309	58.7		3.100	299.7		2.106	119.7
	2.628	111.8		3.355	418.6		2.283	173.7
	2.853	164.2					2.404	210.5
29.0	3.229	281.6	57.6	1.433	5.7		2.640	296.3
	1.410	3.3		1.581	13.8			
	1.768	15.5		1.811	31.5			

In the formulas given above, the following notation has been used:

x_0 , x_y , x_a , x_1 , and x_2 —the concentrations of He⁴ in the liquid phase corresponding to the top of the column, the bottom of the column, the boundary between the liquid and vapor phases, the evaporator, and the beginning and end of the separation.

y_a —concentration of He⁴ in the vapor phase at the boundary with the liquid,

z —the distance measured downward from the top of the column,

γ —the fraction drawn off from the vapor flow at the top of the column,

D_y —diffusion coefficient in the vapor phase,

v_0 —the mean linear velocity of vapor flow,

α —radius of the tube of the column or effective radius, approximately equal to the radius of the packing ring of the column,

q —the number of moles of He⁴ in the column,

p —the total number of moles of helium in the column,

χ —the length of the column,

Q —the number of moles of He⁴ in the column and condenser,

p_0 —the number of moles of helium withdrawn from the column,

W —the rate of flow of vapor in the column (moles per second).

Analysis of the operation of the column leads to the conclusion that it is most suitable to work with maximum flow velocities only a little below the value for percolation.

The combined apparatus, shown in Fig. 5, was very effective for the enrichment of He³ from mixtures with concentrations above 0.1 per cent. The rectifying column 1 and filter 2 were used in it. The column consisted of a tube of stainless steel of diameter 9.6×10 mm and length 200 mm, and had a packing in the ring of diameter 1.5 mm, pre-

TABLE III. Distribution of He^3 between vapor and liquid (X_V/X_L) for different compositions of the liquid phase X_L and different temperatures

X_L	X_V/X_V																		
	1.4° K	1.5° K	1.6° K	1.7° K	1.8° K	1.9° K	2.0° K	2.1° K	2.2° K	2.3° K	2.4° K	2.5° K	2.6° K	2.7° K	2.8° K	2.9° K	3.0° K	3.1° K	3.2° K
0.004	39.75	29.25	22.25	16.75	13.00	10.25	8.75	7.50	6.75	6.00	5.25	4.75	4.25	3.75	3.50	3.00	2.75	2.62	2.50
0.008	33.75	25.62	20.00	15.87	12.62	10.12	8.37	6.87	6.00	5.37	4.75	4.37	4.00	3.62	3.25	3.00	2.75	2.62	2.50
0.019	24.53	19.63	16.05	13.26	11.00	9.21	7.79	6.68	5.74	5.05	4.42	4.05	3.68	3.37	3.05	2.84	2.63	2.47	2.26
0.030	19.17	16.03	13.53	11.57	9.93	8.43	7.10	6.00	5.10	4.43	4.03	3.70	3.30	3.10	2.87	2.67	2.47	2.27	2.07
0.040	16.10	13.82	11.92	10.22	8.67	7.47	6.42	5.50	4.80	4.25	3.97	3.62	3.20	3.07	2.82	2.57	2.40	2.22	—
0.063	11.67	10.43	9.24	8.11	7.11	6.35	5.75	5.21	4.59	3.98	3.75	3.44	3.17	3.00	—	—	—	—	—
0.083	9.34	8.52	7.76	7.04	6.24	5.70	5.22	4.77	4.30	3.87	3.54	3.25	3.04	2.89	2.73	2.54	2.37	2.20	2.06
0.111	7.32	6.76	6.29	5.79	5.33	4.90	4.57	4.19	3.85	3.61	3.31	3.05	2.86	2.71	2.60	2.45	2.31	2.17	2.04
0.134	6.22	5.79	5.42	5.04	4.68	4.36	4.07	3.79	3.51	3.33	3.11	2.90	2.72	2.59	2.48	2.36	2.25	2.14	2.04
0.167	5.12	4.82	4.53	4.24	3.96	3.74	3.53	3.32	3.12	2.99	2.84	2.66	2.52	2.43	2.33	2.19	2.10	—	—
0.192	4.53	4.28	4.03	3.80	3.58	3.39	3.22	3.04	2.87	2.79	2.66	2.50	2.39	2.33	2.23	—	—	—	—
0.226	3.91	3.71	3.52	3.34	3.15	3.01	2.88	2.73	2.61	2.54	2.44	2.33	2.24	2.19	2.10	—	—	—	—
0.239	3.72	3.53	3.36	3.19	3.02	2.89	2.76	2.64	2.53	2.46	2.37	2.27	2.19	2.14	2.05	—	—	—	—
0.302	3.00	2.87	2.75	2.64	2.53	2.44	2.35	2.27	2.20	2.15	2.08	2.03	1.96	1.93	1.87	—	—	—	—
0.383	2.39	2.31	2.24	2.17	2.11	2.05	1.99	1.94	1.90	1.86	1.81	1.78	1.73	1.71	1.68	—	—	—	—
0.527	1.78	1.73	1.69	1.66	1.63	1.61	1.58	1.55	1.53	1.51	1.48	1.47	1.44	1.43	1.41	—	—	—	—
0.563	—	—	—	—	—	—	—	—	1.46	1.44	1.42	1.41	1.39	1.37	1.36	—	—	—	—
0.734	1.31	1.29	1.28	1.26	1.26	1.24	1.23	1.22	1.21	1.20	1.20	1.19	1.18	1.17	—	—	—	—	—
0.824	1.18	1.17	1.16	1.15	1.15	1.14	1.14	1.13	1.12	1.12	1.12	1.11	1.11	1.10	—	—	—	—	—
0.908	1.09	1.08	1.08	1.07	1.07	1.07	1.06	1.06	1.06	1.06	1.06	1.05	1.05	1.05	1.05	—	—	—	—

pared of constantan wire of 0.2 mm. The filter consisted of a copper tube of diameter 2×4 mm, length 45 mm, solidly packed with rouge.

As a rule, the apparatus worked in two cycles, the extraction cycle and the enrichment cycle. The yield and the data characterizing the extraction cycle are given in Fig. 5 on the right side of the instrument. Helium with a velocity of $100 l_0/\text{hour}$ (at 760 mm and 0°C) was condensed in the coil and flowed into the tank 3, from which He^4 was drawn off into the collector 4 by means of thermal flow. Simultaneously, separation of the mixture was carried out from the upper part of the column; this mixture was enriched by rectification to a concentration of the order of 0.4 He^3 . In a second cycle, data on which are given in Fig. 5 on the left of the apparatus, the mixture with an original concentration of about 0.4 He^3 was condensed in the tank 3 and then separation of He^3 was carried out from the top part of the column with a rate of $10 l_0/\text{hour}$. Along with the separation, the temperature of the tank 3 increased from 2 to 2.5°K , while the rate of separation decreased to $4 l_0/\text{hour}$. In one of the experiments, the mean concentration of $4 l_0$ of the withdrawn gas contained 0.99995 He^3 . In the column there remained $0.8 l_0$ of the untreated mixture, of which $0.25 l_0$ were He^3 . For this apparatus, $e\beta l = 4000$, i.e., $1/\beta = 2.5$ cm, and the

time of establishment $\tau_0 = 10$ sec. The apparatus described above makes possible the obtaining of

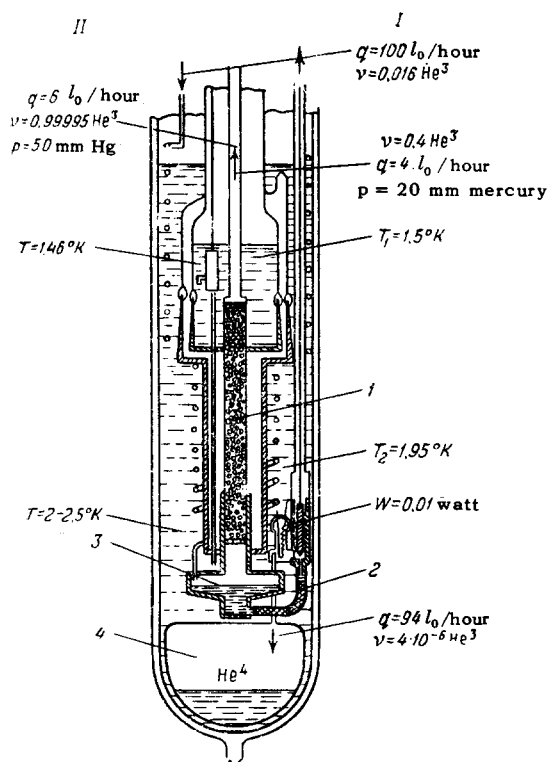


FIG. 5. Apparatus for concentration of He^3 .

He³ of suitably high purity and suitable for re-search with mixtures of He³ and He⁴.

SATURATED VAPOR PRESSURE. CRITICAL POINT

Investigation of the proper use of pure He³ began about ten years ago, when the available supply of He³ was measured in tens of cubic centimeters of the gas under normal conditions. Interest in this liquid was so great that although negligible amounts of He³ were available, researchers nevertheless attempted to ascertain its basic properties.

The first He³ was liquefied in 1948 by Sydoriak, Grilly and Hammel²⁹ who had a total of 20 cm³ of He³ under normal conditions. The He³ was condensed in a steel capillary of internal diameter 1.2 mm, which was immersed in a bath of liquid helium to a depth of 5–10 mm. The other end of the capillary outside of the cryostat was connected to a mercury manometer and a Toepler pump. By variation of the mercury level in the pump, the pressure of He³ in the capillary and manometer was increased. The beginning of condensation was established by the cessation of increase of equilibrium pressure upon further decrease in the volume of the system. In spite of the predictions of many authors,^{30,31} who doubted the possibility of liquefaction of He³ because of its great zero-point energy, Sydoriak, Grilly and Hammel obtained typical Van-der-Waals liquid curves on the diagram of He³. Furthermore, replacing the steel capillary with glass, the authors could see for themselves that liquid was present in the capillary. From the character of the isotherms they established the parameters of the critical point: $T_c = 3.34^\circ\text{K}$, $p_c = 875$ mm of mercury. The same authors determined the normal boiling point of He³, equal to 3.20°K , and the saturated vapor pressure curve in the temperature range from 1.2° to the critical point.

Later, the vapor pressure curve and also the parameters of the critical point were more carefully measured by Abraham, Osborne, and Weinstock,³² who investigated the temperature range from 1.0 to 3.35°K . The authors had at hand 30 cm³ of He³ under normal conditions with a content of He⁴ of $0.03 \pm 0.03\%$. The He³ was concentrated in a cavity in a copper block of volume 50 mm³, connected with an external volume by a monel tube of internal diameter 0.5 mm. The block with the He³ was cooled by the pumping off the helium vapor from the cryostat. Controlling the velocity of flow by means of diaphragms located at the entrance to the pump, the authors

could determine the temperature in the region under investigation with an accuracy up to 0.001°K . The vapor pressure curve, according to the data of this research, is in good agreement with the empirical relation

$$\log p_{\text{mm}} = \frac{0.97796}{T} + 2.5 \log T + 0.00302T^3 + 1.91594, \quad (11)$$

$$1.0^\circ < T < 3.3^\circ\text{K},$$

where T is determined from the consistent temperature scale of 1948³³ with the corrections of Kistemaker.³⁴ The normal boiling temperature computed from this equation is $T = 3.195 \pm 0.001^\circ\text{K}$. The parameters of the critical point, according to the data of this research, are: $T_c = 3.35 \pm 0.02^\circ\text{K}$ and $p_c = 890 \pm 20$ mm mercury. In the temperature range below 1°K (from 0.45 to 1.0°K) the saturated vapor pressure was measured with great accuracy by Sydoriak and Roberts,³⁵ who pumped off the vapors of He³ to cool the liquid. The apparatus of Sydoriak and Roberts is pictured in Fig. 6. The He³ was condensed in a copper sphere of internal diameter 1.2 cm, which was suspended from a thin tube (70 Cu, 30 Ni) of diameter 3 mm in a copper vacuum jacket. The upper end of the tube was fastened to a brass block as shown in the drawing, and the He³ was led outside to the pump. The temperature of the liquid is measured from the magnetic susceptibility of iron ammonium or chromium-potassium alum, located inside the sphere. The salt was

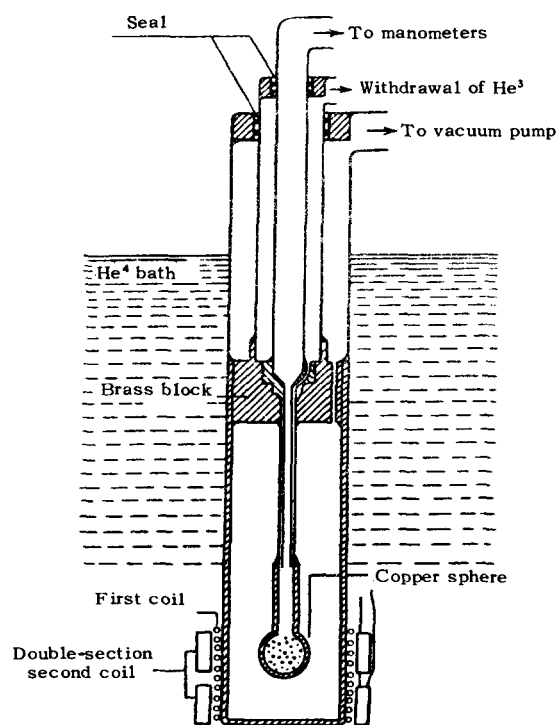


FIG. 6. He³ cryostat of Sydoriak and Roberts.³⁵

first calibrated by the vapor pressure of He³ in the temperature region of 1° K. The vapor pressure was measured by a mercury and oil manometers with high accuracy. The saturated vapor curve below 1° K according to the measurements of Sydoriak and Roberts can be represented by the equation

$$\log p_{\text{mm}} = \frac{1.10371}{T} + 2.3214 \log T + 2.0936 - 0.08976 T + 0.03756 T^2 - 0.004 T^3, \quad 0.45^\circ < T < 1.0^\circ \text{ K}, \quad (12)$$

where T is determined by the E₅₅ scale.^{36,37}

In addition to the saturated vapor pressure, Sydoriak and Roberts also observed the temperature and pressure at the critical point, which were respectively equal to

$$T_c = 3.329^\circ \text{ K}, \quad p_c = 875 \text{ mm mercury}$$

We note that from the measurements of the density

of He³ by the optical method, Peshkov³⁸ obtained for the triple point values $T_c = 3.38 \pm 0.03^\circ$, $p_c = 930 \pm 20$ mm mercury, which differ somewhat from the preliminary data.

The empirical equation (12) of Sydoriak and Roberts for the saturated vapor pressure of the low 1° K agrees very well with the equation of Abraham, Osborne, and Weinstock¹¹ for much higher temperatures. Table IV lists the values of the saturated vapor pressure of He³ in the temperature range 0.3 to 3.34° K according to the data of references 32 and 35. The temperature was determined according to the E₅₅ scale. It is not difficult to see that the vapor pressure of He³ in the region of low temperatures often exceeds the corresponding pressure for He⁴. This makes it possible to apply He³ successfully as a working substance in condensation thermometrics for the measurement of temperatures below 1° K.

TABLE IV. Vapor pressure of He³ in millimeters of mercury at intervals of 0.01° K.
T_E — temperature scale

T°K	0.00	0.01	0.02	0.03	0.04	0.05	0.06	0.07	0.08	0.09
0.3	0.00150	0.00213	0.00296	0.00404	0.00542	0.00717	0.00935	0.01205	0.01533	0.01930
0.4	0.02405	0.02969	0.03634	0.04411	0.05314	0.06355	0.07551	0.08915	0.10462	0.12210
0.5	0.1418	0.1638	0.1883	0.2155	0.2456	0.2789	0.3154	0.3554	0.3991	0.4468
0.6	0.4985	0.5546	0.6152	0.6806	0.7510	0.8267	0.9077	0.9945	1.0872	1.1860
0.7	1.291	1.403	1.522	1.647	1.780	1.921	2.069	2.225	2.390	2.563
0.8	2.744	2.935	3.135	3.344	3.563	3.792	4.031	4.280	4.540	4.810
0.9	5.092	5.385	5.689	6.005	6.333	6.673	7.026	7.391	7.769	8.160
1.0	8.564	8.981	9.413	9.858	10.318	10.792	11.280	11.783	12.301	12.835
1.1	13.384	13.948	14.528	15.124	15.737	16.366	17.012	17.674	18.354	19.051
1.2	19.765	20.497	21.247	22.015	22.801	23.606	24.429	25.271	26.132	27.013
1.3	27.913	28.833	29.772	30.732	31.711	32.711	33.732	34.774	35.837	36.920
1.4	38.03	39.15	40.30	41.47	42.66	43.88	45.12	46.38	47.66	48.97
1.5	50.30	51.65	53.03	54.43	55.85	57.30	58.77	60.27	61.79	63.34
1.6	64.91	66.51	68.13	69.78	71.46	73.16	74.88	76.64	78.42	80.23
1.7	82.06	83.92	85.81	87.73	89.67	91.64	93.64	95.67	97.73	99.81
1.8	101.93	104.07	106.24	108.44	110.67	112.94	115.23	117.55	119.90	122.28
1.9	124.69	127.14	129.61	132.12	134.66	137.23	139.83	142.46	145.12	147.82
2.0	150.55	153.31	156.11	158.94	161.80	164.70	167.63	170.59	173.59	176.62
2.1	179.68	182.78	185.92	189.09	192.30	195.54	198.81	202.13	205.47	208.86
2.2	212.28	215.74	219.23	222.76	226.33	229.93	233.58	237.26	240.97	244.73
2.3	248.52	252.36	256.23	260.14	264.08	268.07	272.10	276.17	280.27	284.42
2.4	288.60	292.83	297.10	301.41	305.75	310.14	314.57	319.04	323.56	328.11
2.5	332.71	337.35	342.03	346.75	351.52	356.33	361.18	366.08	371.02	376.00
2.6	381.02	386.09	391.21	396.37	401.57	406.82	412.11	417.45	422.83	428.26
2.7	433.73	439.25	444.81	450.42	456.08	461.78	467.53	473.33	479.17	485.06
2.8	491.09	496.99	503.02	509.10	515.23	521.40	527.63	533.90	540.22	546.59
2.9	553.01	559.48	566.00	572.56	579.18	585.84	592.56	599.32	606.13	613.00
3.0	619.92	626.89	633.90	640.97	648.09	655.26	662.48	669.76	677.08	684.46
3.1	691.88	699.36	706.89	714.48	722.11	729.80	737.54	745.34	753.18	761.08
3.2	769.04	777.04	785.10	793.21	801.38	809.60	817.87	826.20	834.58	843.01
3.3	851.50	860.05	868.64	877.30	886.00					

EQUATION OF STATE OF He³ GAS

The P-V isotherms of He³ gas (with purity of 99.75%) were obtained experimentally by Keller³⁹ for five values of the temperature in the interval 1.5 to 3.8° K. The second virial coefficient B computed from these measurements and deter-

mined by the equation

$$\frac{PV}{N} = a + b \left(\frac{N}{V} \right) + c \left(\frac{N}{V} \right)^2 + \dots \quad (13)$$

$$a = A = RT; \quad b = aB; \quad c = aC,$$

is plotted in Table V, where the temperatures are given in the Kistemaker scale.³⁴

TABLE V

T° K	B
3.782	-65.4
2.993	-86.3
2.150	-117.9
1.817	-142.3
1.516	-168

These data are accurately described by the empirical relation

$$B \text{ cm}^3/\text{mole} = 10.26 - 299.90 T + 44.27/T^2. \quad (14)$$

The second virial coefficients found experimentally by Keller agree sufficiently well with the values computed in references 40, 41, and 42 on the basis of the Lennard-Jones potential

$$V(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right].$$

The equation of state of He³ gas was also determined by Peshkov,³⁸ who measured the density and the coefficient of isothermal compressibility of gaseous He³ by an optical method. The apparatus of Peshkov consisted of a rectangular glass cell with plane parallel sides and filled with He³. Bands of equal thickness were observed on the surface of the cell in its irradiation by monochromatic light. To determine the coefficient of isothermal compressibility, a constant-temperature bath was maintained and the displacement of the band was observed upon the change in pressure inside the cell. The results obtained are given in Fig. 7, where, for comparison purposes, the Keller data at 3° K are plotted with crosses. The other results of Keller, which are not shown in the figure, also agree well with the measurements of Peshkov. The solid lines on Fig. 7 are curves computed by

Peshkov from the equation

$$\frac{p}{\rho} = 27.35 T - 2.3 \cdot 10^3 \rho + 1.8 \cdot 10^4 \rho^2. \quad (15)$$

As is seen from the drawing, the experimental results are satisfactorily described by these curves which represent the equation of state of the He³ gas. In contrast to the Keller equation (14), the second virial coefficient B is determined in the Peshkov equation (15) by the relation

$$B = -243 \frac{1}{T}. \quad (16)$$

within the limits of experimental error.

THERMOMOLECULAR PRESSURE DIFFERENCES IN He³

Thermomolecular pressure differences in He³ were measured over a wide temperature range (from room temperature to that of liquid helium) by Roberts and Sydoriak.⁴³ The results of measurement for the ratio of the pressure p_{cold} to p_{hot} are given in Table VI, where the product R p_{hot} is expressed in cm of mercury (R = radius of the tube in cm and p_{hot} is the pressure at the "hot" end in microns of mercury).

MELTING AND PHASE TRANSITION CURVE OF SOLID He³

Experiments with He³ at very low temperatures have shown that He³ (like He⁴) cannot be solidified by cooling under pressure of the saturated vapor. To obtain solid He³, it is necessary to raise the pressure to several tens of atmospheres.

The first solid He³ was obtained by Osborne, Abraham, and Weinstock⁴⁴ in 1951. The authors studied the melting curve of He³ in the temperature range 1.02 - 1.51° K, using the blocked capillary method for this purpose. The apparatus sys-

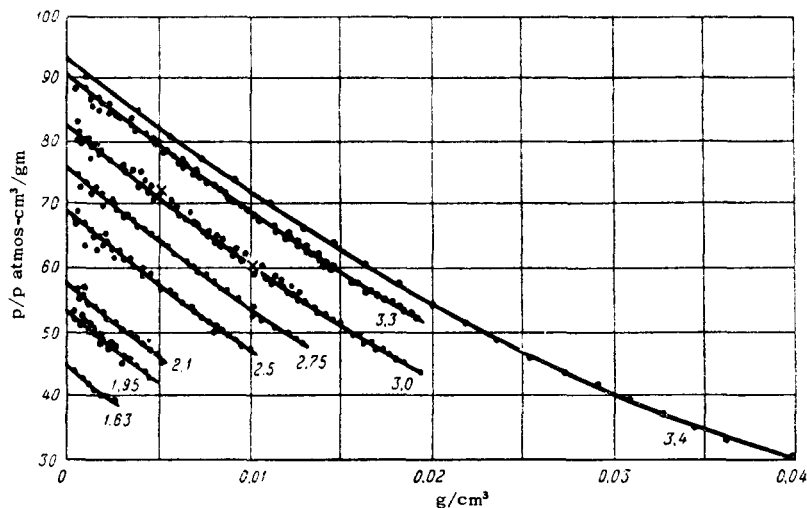


FIG. 7. Compressibility curves for gaseous He³. The temperature in °K is shown on the curves.

TABLE VI

$R p_{\text{hot}}$ cm $\cdot\mu$ mercury	$(p_{\text{cold}}/p_{\text{hot}})_{\text{comp}}$	$(p_{\text{cold}}/p_{\text{hot}})_{\text{meas}}$ not corrected for absorption	$(p_{\text{cold}}/p_{\text{hot}})_{\text{meas}}$ corrected for absorption	$\frac{(3)-(1)}{(1)} \cdot 100$
Experiment I: $T_{\text{cold}} = 2.15^\circ\text{K}$; $T_{\text{hot}} = 301.2^\circ\text{K}$				
4.87	0.4730	0.4783	0.4719	-0.2%
8.12	0.5917	0.5939	0.5857	-1.0
10.94	0.6608	0.6683	0.6619	+0.2
13.67	0.7096	0.7155	0.7101	+0.1
18.97	0.7781	0.7834	0.7794	+0.2
24.2	0.8225	0.8206	0.8174	-0.6
29.0	0.8527	0.8573	0.8546	+0.2
33.9	0.8756	0.8804	0.8781	+0.3
43.8	0.9074	0.9079	0.9061	-0.1
53.1	0.9272	0.9347	0.9332	+0.6
63.0	0.9419	0.9448	0.9435	-0.2
89.2	0.9648	0.9654	0.9645	0.0
115.3	0.9761	0.9758	0.9752	-0.1
167.2	0.9870	0.9832	0.9828	-0.4
291	0.9950	0.9944	0.9943	-0.1
409	0.9973	(0.9973)	(0.9973)	
Experiment II: $T_{\text{cold}} = 1.90^\circ\text{K}$; $T_{\text{hot}} = 305.7^\circ\text{K}$				
8.56	0.6000	0.6004	0.5955	-0.8%
14.8	0.7235	0.7142	0.7095	-2.0
20.25	0.7897	0.7967	0.7930	+0.4
24.7	0.8227	0.8370	0.8338	+1.3
29.0	0.8492	0.8646	0.8620	+1.5
36.4	0.8820	0.8953	0.8932	+1.3
46.8	0.9122	0.9196	0.9179	+0.6
55.5	0.9303	0.9414	0.9400	+1.0
66.4	0.9457	0.9482	0.9470	-0.1
95.6	0.9682	0.9734	0.9726	+0.5
155	0.9846	0.9842	0.9836	-0.3
244	0.9930	0.9908	0.9903	-0.4
390	0.9971	0.9934	0.9933	
646	0.9990	(0.9990)	(0.9990)	

tem is pictured in Fig. 8. The authors had available 190 cm³ of gaseous He³. To decrease the volume of the system, a U-shaped tube, submerged in a liquid-helium bath, and the connecting tubes were made of steel capillaries of very small internal diameter, 0.1 mm for the U-shaped tube and 0.5 mm for the attached tubes. The Bourdon gauges B and G were filled with mercury to decrease the free volume. To prevent stopping of the U-shaped capillary with mercury, the traps C and F were prepared and immersed in liquid nitrogen. The process of measurement consisted of the following. By means of the mercury piston H, the pressure in the system at a certain constant temperature of the cryostat was raised uniformly. The increase in the pressure was recorded by the manometers B and G. The readings of the manometers were identical so long as the He³ did not solidify in the U-shaped capillary, after which the readings of the manometer B remained constant. The measurements showed that, just as in He⁴, the melting pressure of He³ rose monotonically with temperature, changing from 40 atmos at 1°K to 57 atmos at 1.5°K. The same authors soon extended their measurements to 0.16°K,⁴⁵ making use of the method of adiabatic demagnetization for this purpose. For this purpose a U-shaped capillary (Cu-Ni) with an internal diameter of 0.16

mm, containing the He³, was placed in the chamber and filled with iron ammonium alum. To improve the thermal contact between the capillary and the salt, copper bands were attached to the surface of the capillary while the chamber was filled with He⁴ at a pressure of 1 atmos at the temperature of liquid nitrogen. The measurements were carried out in the same order as in the previous case. Experiment showed that in the tem-

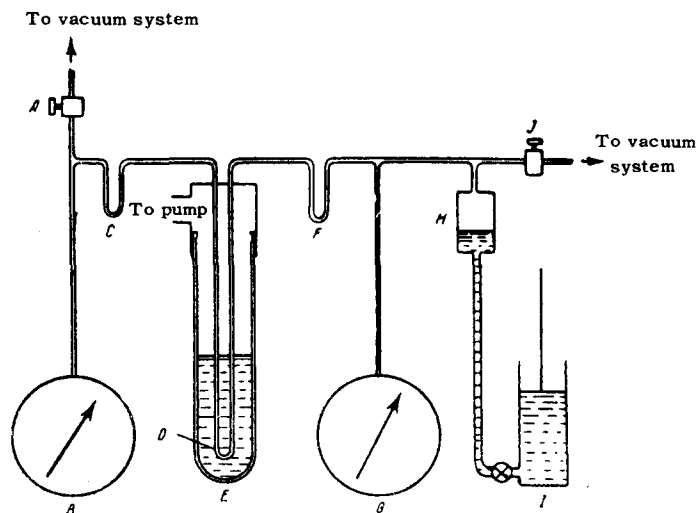
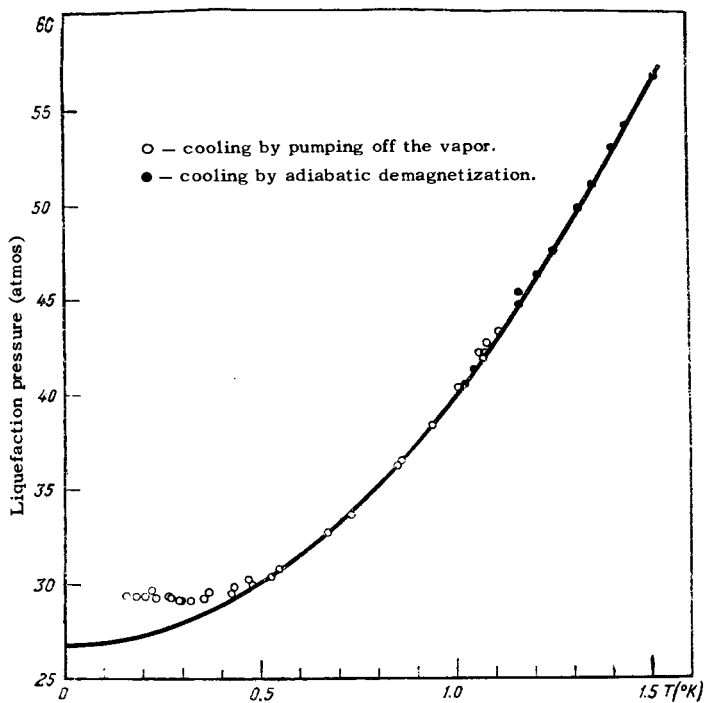


FIG. 8. Apparatus for the determination of the solidification curve (schematic representation).

FIG. 9. Liquefaction curve of He³.

perature range from 0.5 to 1.5° K, the liquefaction curve is accurately described by the empirical formula $P = 26.8 + 13.1 T^2$, while below 0.5° K the experimental points deviated from this dependence, lying on a horizontal line corresponding to a pressure of 29.3 atmos. The liquefaction curve, based on reference 45, is plotted in Fig. 9.

The authors first assumed that the horizontal portion of the curve below $T = 0.5^\circ \text{K}$ is a consequence of a sudden thermal contact between the liquid He³ and the salt which did not guarantee cooling of the liquid below 0.5° K. However, it soon became clear that the result obtained could be explained by the existence of a minimum in the melting curve predicted by Pomeranchuk⁴⁶ on the basis of his theory of the Fermi liquid. At temperatures below the temperature of the minimum, blocking of the capillary will take place each time, at a pressure corresponding to a minimum on the liquefaction curve. The capillary in this case could be blocked not in the cold place itself but in a warmer part above, where the temperature corresponds to the minimum pressure. In 1957 the prediction of Pomeranchuk was indirectly confirmed by Walters and Fairbank⁴⁷ who observed an increase in temperature at 0.2° K upon decrease in the pressure on the solid He³. For a temperature of 1.2° K, the effect has an opposite sign, i.e., the decrease in the pressure is accompanied by cooling. These observations indirectly supported the predictions of Pomeranchuk, since only in this

case will the heat of melting be negative.

A study of the liquefaction curve for much higher temperatures was carried out by a number of authors. In the range of temperatures above 1.5° K (from 1.5 to 2.3° K) the solidification curve of He³ was indirectly determined by Esel'son and Lazarev⁴⁸ by extrapolation of their data on the solidification of mixtures of He³-He⁴. In 1958, Sherman and Edeskuty, carefully measuring the P-V-T diagram of He³, determined the liquefaction curve in the range from 1.07 to 3.10°, also using the method of a blocked capillary. In this temperature range, the liquefaction curve is described by the equation

$$P_{\text{atm}} = 23.29 + 2.955 T + 14.66 T^2 - 1.1871 T^3. \quad (17)$$

with an accuracy of not less than 0.2% (± 0.05 atmos). The liquefaction curve was obtained by Mills and Grilly⁵⁰ for a significantly wide range of temperatures from 2 to 31° K, which corresponds to a change in pressure from 76 to 3500 kg/cm².

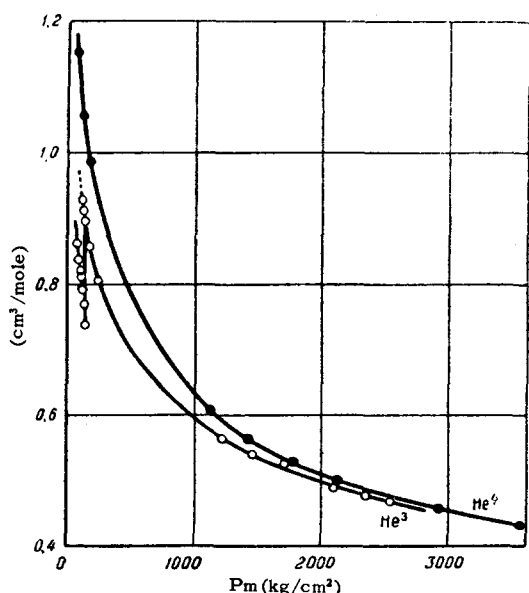
The measurements were also carried out by the method of the blocked capillary. The liquefaction curve obtained by these authors was a smooth continuation of the previous curve. In the range from 2 to 31° K, it is satisfactorily described by the equation

$$P = a + bT^c, \quad (18)$$

where $a = 25.16$, $b = 20.082$ and $c = 1.517$.

In 1957 experiments were carried out by the same authors⁵¹ on the determination of the change in the molar volumes upon liquefaction ΔV_M , the molar volumes of the liquid V_f , and the coefficient of thermal expansion of the liquid α_f along the liquefaction curve over a wide range of pressures from 80 to 3500 kg/cm² (or from 2 to 31° K). The change in the volume of solid helium on liquefaction was determined from the mass of gas absorbed by the container during the process of liquefaction of He³ in order to maintain constant pressure. The authors obtained an interesting and unexpected result, namely: at pressures below 141 kg/cm²,¹² two branches were observed in the curve of change in volume upon liquefaction vs. pressure: an upper and a lower, displaced parallel to one another about 15%. The results of these measurements are shown in Fig. 10. The branch obtained depends upon the initial temperature of the solid helium and bears witness to the existence in it of some sort of transformation.

Careful measurements of the liquefaction curve in steps of 0.1° K in the range from 2 to 4.5° K, carried out by the authors for the purpose of discover-

FIG. 10. Change in volumes He³ and He⁴ upon liquefaction.

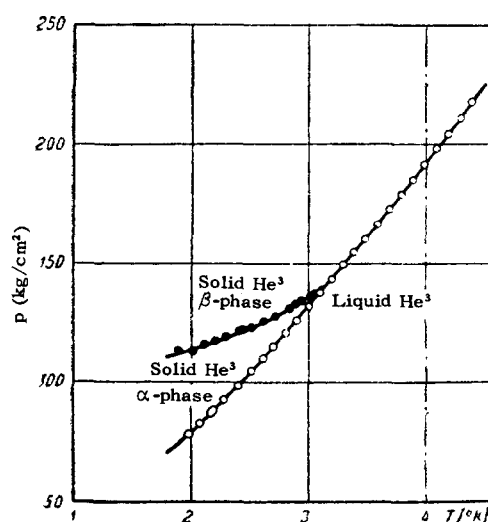
ing discontinuities in the derivatives at any point in the curve, showed that for $p = 141 \text{ kg/cm}^2$ and $T = 3.15^\circ \text{ K}$ a small discontinuity actually takes place in the slope of the curve. Further investigations succeeded in determining part of the transition line from one solid phase to the other. The transition curve was determined by careful observations on the rate of increase or, conversely, of decrease in pressure on the solid He³. At the transition point, the pressure changes discontinu-

TABLE VII. Characteristics of the α - β transition in solid He³

$p \text{ kg/cm}^2$	$T^\circ \text{ K}$	$\Delta V \text{ cm}^3/\text{mole}$	$\Delta S \text{ cal/mole-deg}$
110	1.78	0.138	0.053
120	2.35	0.142	0.063
130	2.82	-0.150	0.093
141	3.15	0.157	0.138

TABLE VIII. Characteristics of the melting curve of He³ and He⁴

$P_m \text{ kg/cm}^2$	$\Delta V_m \text{ cm}^3/\text{mole}$		$\Delta S_m \text{ cal/mole-deg}$		$V_f \text{ cm}^3/\text{mole}$		$\alpha_f \times 10^3 \text{ deg}^{-1}$	
	He ³	He ⁴	He ³	He ⁴	He ³	He ⁴	He ³	He ⁴
80	0.854	1.150	0.94	1.37	21.92	19.74	7.5	12.0
100	0.815	1.106	0.99	1.38	20.80	19.10	7.0	10.7
141	0.737	1.036	1.06	1.43	19.72	18.18	6.5	9.5
141	0.894		1.15					
200	0.834	0.955	1.26	1.49	18.42	17.22	5.9	8.3
500	0.688	0.762	1.44	1.62	15.56	14.74	4.4	5.7
1000	0.590	0.628	1.58	1.70	13.03	13.03	3.4	4.1
1500	0.536	0.554	1.65	1.73	12.42	12.05	2.9	3.3
2000	0.494	0.506	1.69	1.75	11.71	11.38	2.6	2.8
3000	0.446	0.452	1.74	1.80	10.78	10.46	2.1	2.3

FIG. 11. Liquefaction curve and phase transitions of solid He³.

ously, which is evidence of the sudden change in molar volumes. Figure 11 shows the phase diagram of solid He³, where the different modifications of the solid state are denoted by the authors⁵¹ as α and β phases. Table VII gives the basic characteristics of the α - β transition, obtained by Mills and Grilly. In the light of these data, the presence of two peaks on the curve of change of volume of liquefaction (Fig. 10) becomes understandable. In the case in which the solidification of He³ takes place at a temperature corresponding to the α phase, the melting is accompanied by a change in volume of ΔV_m . If the same solidification takes place at a temperature much lower than the transition temperature, the volume change upon liquefaction increases to a value equal to the volume change in the α - β transition. Data are given in Table VIII on the volume and entropy changes upon liquefaction, and also on the volume and the coefficient of bulk compressibility of the liquid phase along the melting curve of He³.

To make clear the structure of the α and β

phases, Schuck, Grilly and Mills⁵² carried out x-ray analysis of solid He³. The authors investigated the diffraction of x-rays by solid He³ contained in a beryllium capillary. The studies were carried out by the method of Laue and Hull-Debye-Scherrer. It was found that He³, like He⁴, formed large homogeneous crystals much more easily than fine crystalline powders. It was established by x-ray experiments that the α phase possesses a cubic lattice. At a temperature of 1.9° K and pressure 100 kg/cm², the length of the cube edge was $A = 4.01 \pm 0.03$ Å. The density of the α phase determined on the basis of these experiments was $\rho_\alpha = 0.154 \pm 0.004$ g/cm³, which agrees well with the direct measurements of the density of Mills and Grilly.⁵¹ The structure of the β phase was found to be hexagonal with close packing. At $T = 3.3^\circ$ K and $p = 183$ kg/cm², the axes of the crystal were given by $A = 3.46 \pm 0.03$ Å and $C = 5.60 \pm 0.03$ Å, which corresponds with a density $\rho_\beta = 0.172 \pm 0.004$ g/cm³. Thus, it was shown by x-ray experiments that the α and β phases possess different crystallographic structure.

It is clearly seen from the phase diagram of He³ that the melting curve does not show the slightest tendency toward intersection with the vapor pressure curves. Thus He³, like He⁴, does not have the ordinary triple point, where the liquid, vapor, and solid phases are found in equilibrium.

DENSITY OF LIQUID, GASEOUS, AND SOLID He³

The approximate determination of the density of the liquid and its saturated vapor as a function of temperature was performed in the first experiments on liquid helium by Sydoriak, Grilly, and Hammel.²⁹ However, direct measurements of the density of the liquid and vapor with high accuracy (0.1% for the liquid and 1.3% for the vapor) were completed by Kerr⁵³ in the temperature range from 1.3 to 3.2° K only as recently as 1954. The apparatus for the measurement of the density⁵³ consisted of a calibrated 50-cc container filled with gas at room temperature and of a small Toepler pump with a manometer for the measurement of the pressure. The gas whose amount was measured by a calibrated vessel was passed through a monel capillary of 0.31 mm internal diameter into a pycnometer, immersed in a tank of liquid helium. To decrease the fluctuations of the amount of gas contained in the capillary (because of temperature gradients) the latter was surrounded by a copper vacuum jacket, immersed at the lower end in the helium bath and at the upper end in liquid nitrogen. The pycnometer was prepared of pyrex glass and had a thin calibrated neck with a recessed mark.

The volume of the pycnometer up to the mark (0.11148 ± 0.00006 cm³) was determined by suspending it both empty and filled with mercury. The pycnometer for the measurement of the gas density had a volume of 0.7465 ± 0.0003 cm³. In the calculations, a correction was introduced which took into account the change in the volumes of the pycnometers from room temperature to liquid helium temperatures. The results of the measurements of Kerr are shown graphically in Fig. 12. The

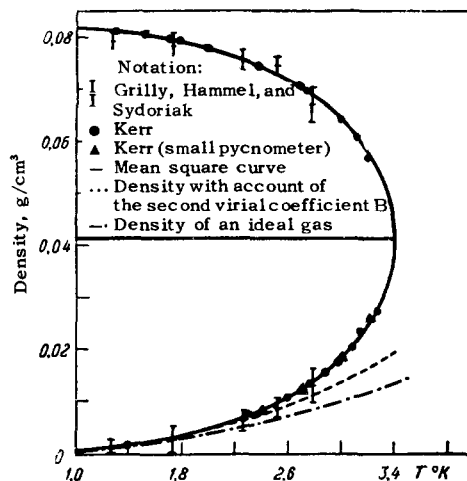


FIG. 12. Density curve of He³.

values of the density of the liquid and the vapor in the 1.0–3.3° K range are also given in Tables IX and X, where the temperature was determined according to the Kistemaker scale.³⁴ In the tables the values are also given of the density computed by the formula $\rho = 0.04117 + 1.14 \times 10^{-5} T \pm [(0.04117)^2 - 3.3015 \times 10^{-6} T^2 - 4.1876 \times 10^{-5} T^3]^{1/2}$. The plus sign corresponds to the density of the liquid and the minus to that of the vapor. The density of liquid helium at the critical point, according to the measurements of Kerr, is equal to

TABLE IX. Density of liquid He³

T_c °K	Density (meas) g/cm ³	Density (comp.) g/cm ³	Deviation (per cent)
1.304	0.08121	0.08121	0.00
1.516	0.08051	0.08053	+0.02
1.709	0.07964	0.07971	+0.09
1.787	0.07935	0.07931	-0.05
1.998	0.07803	0.07803	0.00
2.013	0.07792	0.07792	0.00
2.378	0.07470	0.07472	+0.03
2.393	0.07444	0.07458	+0.16
2.698	0.07072	0.07054	-0.26
2.710	0.07040	0.07035	-0.07
2.748	0.06971	0.06973	+0.03
3.010	0.06434	0.06437	+0.05
3.020	0.06423	0.06412	-0.17
3.135	0.06067	0.06086	+0.31
3.223	0.05681	0.05775	+1.62

TABLE X. Density of He³ vapor

T_c , °K	Density (meas) g/cm ³	Density (comp.) g/cm ³	Deviation (per cent)
1.395	0.00146	0.00152	+3.94
1.689	0.00269	0.00268	-0.37
2.305	0.00713	0.00706	-0.99
2.596	0.01059	0.01051	-0.76
2.767	0.01315	0.01317	+0.15
2.890	0.01529	0.01550	+1.35
2.991	0.01756	0.01776	+1.13
2.994	0.01774	0.01783	+0.50
3.097	0.02030	0.02058	+1.36
3.157	0.02293	0.02247	-2.04
3.160	0.02297	0.02257	-1.77
3.282	0.02727	0.02752	+0.91

$\rho_c = 0.0413 \text{ g/cm}^3$. The results of Kerr were confirmed in the subsequent work of other authors. In 1937, Peshkov³⁷ measured the temperature variation of the density of liquid and gaseous helium by an optical method in the interval 1.4 – 3.2° K.

These results coincided with the Kerr curve. The value of the density at the critical point according to the data of Peshkov was equal to $\rho_c = 0.041 \pm 0.001 \text{ g/cm}^3$. The results of the measurements of Kerr and Peshkov were also confirmed in the work of Ptukha who determined the density of pure He³ in the range 1.3 – 2.7° K. The value of the density close to absolute zero was obtained by Kerr by extrapolation of his data in the region of low temperatures. However, it has recently become known from the experiments of Fairbank and Lee,⁵⁴ on the determination of the thermal conductivity of He³, that such an extrapolation is not valid, since in the temperature range around 0.5° K the density curve has a maximum. The presence of the maximum was established by the sharp increase in the heat transmission in liquid He³ close to 0.5° K at high thermal flows directed from top to bottom. The effect was absent at very high temperatures. The increase in the heat transmission was explained by the author by an additional convective transfer of heat, which is possible in the case when $\partial\rho/\partial T > 0$. We found out⁵⁵ that this assumption was confirmed, by measurements made by the same authors with other apparatus, in which the heat was propagated from bottom to top. In the new apparatus the effect observed previously was discovered at $T > 0.5^\circ \text{ K}$.

To explain the temperature variation of the density below 1° K, Taylor and Kerr⁵⁶ carried out measurements on the coefficient of thermal expansion of liquid He³, $\alpha = (1/V)(dV/dT)$, in the temperature range from 0.54 to 1.01° K. The measurements were made with a pycnometer of volume of about 3 cm³, attached to a tube with a small internal bore. For a constant mass of He³ con-

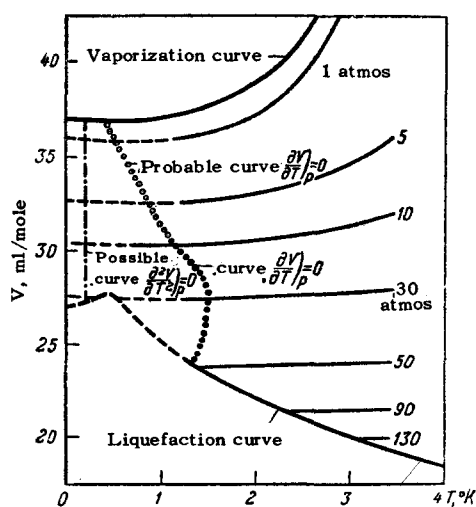
densed in the pycnometer, the location of the meniscus of the liquid was determined at various temperatures. In the entire interval under investigation, the coefficient of expansion was a positive quantity. The values of α for three temperatures are given in the following short table. Thus

T , °K	α
0.55	0.0036
0.7	0.0082
0.9	0.0164

the authors did not reach any anomalies in the density curve. Since the increase in the thermal conductivity in the first experiments of Fairbank and Lee⁵⁴ was observed only for very large heat flows, the maximum on the density curve in all probability is not large. To make clear the temperature dependence of the density in the vicinity of absolute zero further experiments are necessary.

Measurements of the density of liquid helium under pressure over a wide range of temperatures and pressures were completed by a number of investigators. The V-T diagram of He³ computed⁵⁷ in accord with the data of Sherman and Edeskuty,⁴⁹ Taylor and Kerr,⁵⁶ and Kerr⁵³ is plotted in Fig. 13.

In this drawing the solid line refers to the experimental curves, the dashed lines to those curves obtained by extrapolation. The geometric location of the minima of isobars (maxima of density) is indicated by the points. Since, in accordance with the Nernst theorem, $(\partial V/\partial T)_p = 0$ at $T = 0$, the minimum on each curve below must be a point of inflection, where $(\partial^2 V/\partial T^2)_p = 0$. For lack of experimental data, this curve (dash-dot) is drawn arbitrarily. So far as the density of

FIG. 13. V - T diagram of He³.

solid He³ is concerned, as has been pointed out above, values at two points have been obtained from x-ray analysis:

α -phase

$$T = 1.9^\circ \text{K}, \quad p = 100 \text{ kg/cm}^2, \quad \rho_\alpha = 0.154 \pm 0.004 \text{ g/cm}^3$$

β -phase

$$T = 3.3^\circ \text{K}, \quad p = 183 \text{ kg/cm}^2, \quad \rho_\beta = 0.172 \pm 0.004 \text{ g/cm}^3.$$

HEAT OF VAPORIZATION

The heat of vaporization can be computed from the Clausius-Clapeyron equation:

$$\Delta Q_{\text{evap}} = T(V_g - V_{\text{liq}}) \frac{dP}{dT} \quad (19)$$

on the basis of experimental data on the specific densities of the liquid and the vapor as a function of the pressure of saturated vapor and temperature. Such calculations were carried out by Abraham, Osborne and Weinstock³² and later by Kerr⁵³ and Sydoriak and Roberts.⁵⁸ Direct measurements of the heat of vaporization at a single point for $T = 1.5^\circ \text{K}$ ($\Delta Q_{\text{vap}} = 10.39 \pm 0.02 \text{ cal/mole}$) were carried out by a calorimetric method with great accuracy by Osborne and Weinstock.⁵⁹ The temperature dependence of the heat of vaporization ΔQ_{vap} according to the data of the authors just mentioned is shown in Fig. 14. It is easy to see that it is similar to the temperature dependence of ΔQ_{vap} for He⁴; however, it is less in absolute magnitude by a factor of about 2. The values of ΔQ_{vap} , obtained by the different authors, are given in Table XI.

SPECIFIC HEAT

A great interest for investigators lies in the measurement of the specific heat of He³. The discovery of an anomaly in the curve of the specific heat would certainly give evidence toward the

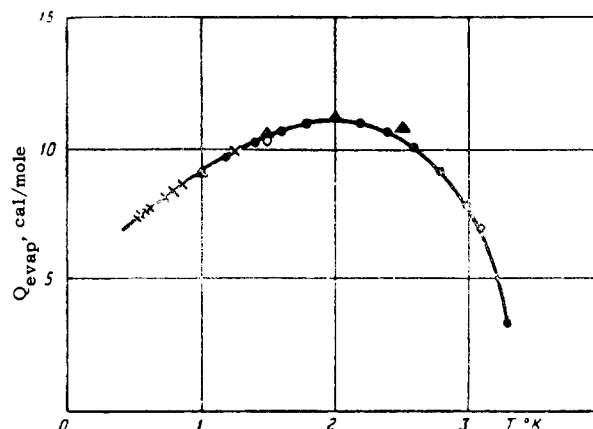


FIG. 14. Heat of vaporization of He³, \blacktriangle — Abraham, Osborne, and Weinstock,³² \bullet — Kerr,⁵³ \times — Sydoriak and Roberts,⁵⁸ \circ — Osborne and Weinstock.⁵⁹

existence of some sort of transformations in liquid He³ similar to the anomaly in the specific heat of He⁴ bears witness to the phase change of non-superfluid helium into the superfluid.

The specific heat of He³ (96% He³, 4% He⁴) was first measured by de Vries and Daunt^{60,61} in the temperature range $0.57 - 2.3^\circ \text{K}$ at the saturated vapor pressure. The authors possessed a negligible amount of He³, about 8 cm^3 of gas under normal conditions; this determined the construction of the apparatus. The He³ was condensed through a thin-walled capillary of internal diameter 0.25 mm into an adiabatic calorimeter of 16 mm^3 volume. The temperature was recorded automatically. Below a temperature of 1.3°K , the calorimeter was cooled by adiabatic demagnetization. Thermal contact with the reservoir created a superconducting heat switch, which was controllable by a magnetic field.⁶² As a consequence of the small amount of He³ contained in the calorimeter, corrections to the heat of vaporization, specific heat of the vapor, etc., amounted to a substantial amount — up to 17%. According to the

TABLE XI. Heat of vaporization of He³

Abraham et al ³²		Kerr ⁵³		Sydoriak, Roberts ⁵⁸		Osborne, Weinstock ⁵⁹	
T° K	ΔQ_{evap} cal/mole	T° K	ΔQ_{evap} cal/mole	T° K	ΔQ_{evap} cal/mole	T° K	ΔQ_{evap} cal/mole
0	(4.47)	1.0	9.10	0.54	7.36	1.5	10.39 ± 0.02
0.5	(6.95)	1.2	9.76	0.566	7.47		
1.0	9.12 ± 0.12	1.4	10.32	0.598	7.60		
1.5	10.65 ± 0.24	1.6	10.71	0.629	7.72		
2.0	11.34 ± 0.41	1.8	11.02	0.748	8.19		
2.5	10.81 ± 0.69	2.0	11.15	0.804	8.41		
		2.2	11.05	0.872	8.66		
		2.4	10.71	1.256	9.90		
		2.6	10.11				
		2.8	9.9				
		3.0	7.88				
		3.1	7.01				
		3.2	5.91				
		3.3	3.36				

estimate of the authors, the accuracy of measurement did not exceed 10%. In spite of the great scatter of individual measurements, the results quite clearly showed a smooth decrease in the specific heat over the measured temperature range, from 3.6 to 0.9 cal/deg-mole, and demonstrated the complete absence of any sort of anomaly. In these measurements, attention was turned toward the temperature interval below 1.0° K, where the specific heat had a constant value, equal to 0.9 cal/deg-mole. Subsequently, Roberts and Sydor-
iak,^{63,58} and simultaneously, Abraham, Osborne and Weinstock^{64,65} in their measurements of specific heat expanded the temperature range, to 0.37° and 0.23° K, respectively. The measurements of Roberts and Sydor-
iak⁵⁸ were completed in the temperature interval from 0.37 to 2.36° K on the apparatus used previously by them for the determination of the saturated vapor pressure (Fig. 6). For the determination of the specific heat, the authors employed an original calorimetric method. He³ with a content of He⁴ less than 0.1% was condensed in a copper sphere of internal diameter 10 mm, which was suspended in a vacuum from a thin-walled tube (Cu-Ni). Fifty per cent of the volume of the sphere was filled with a paramagnetic salt. The calorimeter was cooled by pumping out the He³ vapors to a temperature of 0.5° K. Much lower temperatures were achieved by adiabatic demagnetization of the salt. The temperature of the liquid was determined by the magnetic susceptibility of the salt. The process of measurement consisted in the determination of the rate of heating the liquid (\dot{T}) for a known heat supply \dot{Q} as a function of the amount of He³ contained in the calorimeter. It is easy to see that the slope of the line \dot{Q}/\dot{T} as a function of N the number of moles of He³ (after introduction of corrections for the heat of vaporization of the liquid and the specific heat of the vapor in the calorimeter) gives the specific heat of the liquid directly in cal/deg-mole. In such a method of measurement, the specific heat of the empty calorimeter is automatically eliminated. However, the method is ill-suited for measurement at high temperature, where the corrections become very great. In the temperature range 0.5 – 1.7° K, the accuracy of the measurement, according to the estimate of the authors, amounted to 1.5 or 2%.

The measurements of Abraham, Osborne, and Weinstock were carried out in the temperature range 0.23 – 2° K. The adiabatic calorimeter had a volume of 1.87 cm³. The He³ was cooled by adiabatic demagnetization of the salt which was in contact with the liquid. The values of the spe-

cific heat in reference 65 were determined with an accuracy of not less than $\pm 5\%$.

The results of the researches of all three groups of authors agreed with each other within the limits of experimental measurement, with the exception of the data of de Vries and Daunt⁶¹ in the region below 1° K. In correspondence with the more accurate measurements,^{58,65} the specific heat of He³ in the region below 1° K falls from 1.0 cal/deg-mole to 0.7 cal/deg-mole at 0.25 – 0.3° K. The results of the measurement of Sydor-
iak and Roberts⁵⁸ and Abraham, Osborne and Weinstock⁶⁵ are shown in Fig. 15.

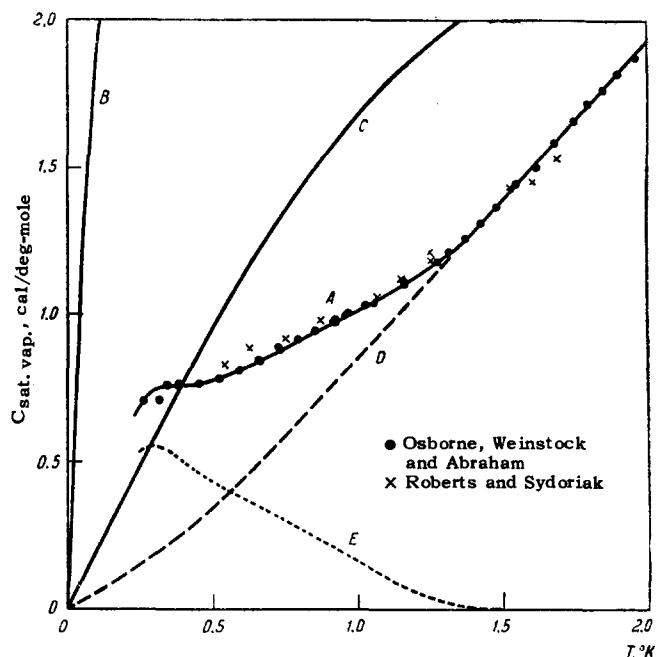
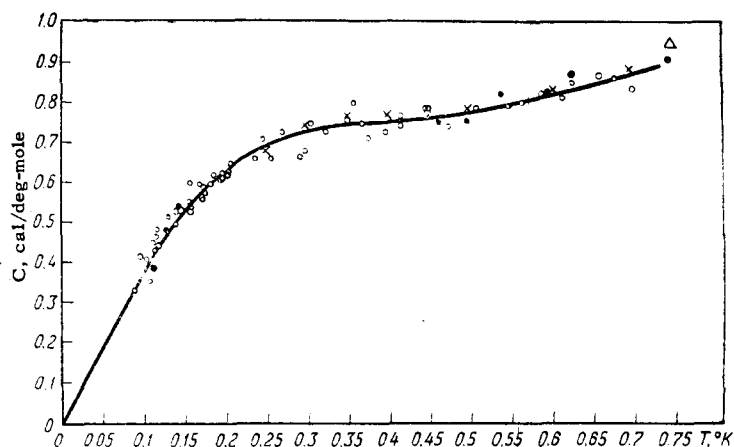


FIG. 15. Specific heat of liquid He³ under pressure of the saturated vapor. A – experimental results; B and C represent the C_V of an ideal Fermi-Dirac gas with a degeneracy temperature of 0.45 and 4.98°K, respectively; D – the computed curve of the non-spin specific heat; E – the computed curve of the spin specific heat (the curve A minus the curve D).

In 1958, Brewer, Sreedhar, Kramers, and Daunt⁶⁶ measured the specific of He³ under pressures from 12 to 15 cm of mercury in the temperature range from 0.7 to 0.085° K.

The He³ was condensed in an adiabatic calorimeter of volume 0.5 cm³. The calorimeter was cooled by adiabatic demagnetization of a salt located in contact with the He³. Thermal contact with the bath existed by means of a superconducting lead switch, which was controlled by a magnetic field.⁶² A 100-ohm manganin coil served as an electric heater. The temperature was measured by the magnetic susceptibility of cerium-magnesium nitrate. This salt was chosen because of its strong adherence to Curie's law in the in-

FIG. 16. Molar heat capacity of He³ at a pressure of 12–15 cm of mercury. O – Brewer, Sreedhar, Kramers and Daunt,⁶⁶ Δ – de Vries and Daunt,⁶¹ ● – Roberts and Sydoriak,⁵⁸ × Abraham, Osborne and Weinstock.⁶⁵



terval under investigation and also because of its low specific heat.

The results of the measurements of Brewer, Sreedhar, Kramers and Daunt are shown in Fig. 16, where for comparison the data of Abraham, Osborne, and Weinstock⁶⁵ and Roberts and Sydoriak⁵⁸ are also given. It is seen from the drawing that the results of reference 56 are a smooth continuation of the earlier curve, pictured in Fig. 15 (the difference in specific heats which is due to the pressure difference on the liquid, is very small here). The characteristic in this drawing is the linear extrapolation of the results to 0° K. The research of reference 56 confirms the existence of a linear term in the heat capacity predicted for the Fermi liquid by Pomeranchuk⁴⁶ and obtained on the basis of the Landau theory⁵⁷ by Khalatnikov and Abrikosov.⁶⁸ At the same time, any doubts were eliminated of the possibility of the transition of He³ to the superfluid state above 0.1° K, since in this region the heat capacity curve has no anomalies.

ENTROPY

Data on the specific heat permitted the authors of reference 56 to calculate the entropy of liquid helium in the interval 0–0.7° K (the specific heat below 0.08° K was assumed to be proportional to T). The values of the entropy for higher temperatures were computed by Roberts and Sydoriak,⁵⁸ Abraham, Osborne, and Weinstock,⁶⁵ and de Vries and Daunt⁵¹ on the basis of their data on specific heat, and also on existing experimental data on the vapor pressure and heat of vaporization of He³. In the temperature range 0.085–2.5° K, the entropy of the liquid according to the calculations of these authors gradually increases from the value 0.3 to 9.5 cal/deg-mole. Below 0.1° K, the curve has a linear portion with a slope of 3.75 cal/mole-deg.²

The dependence of the entropy of liquid and solid

He³ on temperature has been considered theoretically by Pomeranchuk.⁴⁶ According to Pomeranchuk's theory, the entropy of the liquid at a temperature of the order of 1° K must fall to zero as $T \rightarrow 0$ much more rapidly than the entropy of the solid. The latter circumstance is explained by the fact that the forces of exchange interaction in the liquid reduce to the ordering of the spin orientations in parallel or anti-parallel states at temperatures below 1° K while in the solid body, because of the small amplitude of the zero-point vibrations in comparison with the interatomic distances, such as ordering sets in at much lower temperatures (10^{-6} – 10^{-7} deg K), where T becomes of the order of the magnetic energy of interaction of two neighboring atoms. For this reason, the entropy of solid He³ in the temperature range from 10^{-7} to 1° K is fundamentally determined by the chaotic distribution of spin orientations of the nuclei and must be equal to $R \ln 2 = 1.377$ cal/deg-mole (the amount of entropy, corresponding to thermal excitation is practically equal to zero), since in the liquid the entropy falls off rapidly with decrease in temperature, in accordance with the increase of the degree of order. In Fig. 17 we have plotted the variation of the entropy for the liquid and solid He³ to be expected from the Pomeranchuk theory.

As a result of the temperature dependence that has been described for the entropy of solid and liquid He³, there must be a minimum on the melting point curve following from the Clausius-

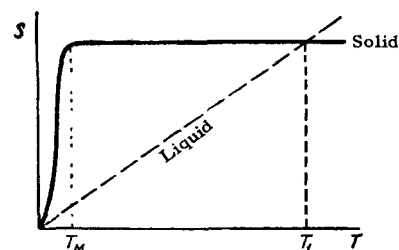


FIG. 17. Entropy of solid and liquid He³ according to Pomeranchuk.⁴⁶

Clapeyron equation, namely;

$$\left. \begin{aligned} \frac{d p_{\text{melt}}}{d T} &= \frac{S_{\text{liq}} - S_{\text{sol}}}{V_{\text{liq}} - V_{\text{sol}}} < 0, & T < T_{\text{min}}, \\ \frac{d p_{\text{melt}}}{d T} &= \frac{S_{\text{liq}} - S_{\text{sol}}}{V_{\text{liq}} - V_{\text{sol}}} > 0, & T > T_{\text{min}}, \end{aligned} \right\} \quad (20)$$

As is easily from (20), the position of the minimum on the melting point curves corresponds to a temperature at which the entropies of the liquid and solid phases of the He^3 in equilibrium with one another are equal. It also follows from (20) that there is a change inside of the heat of melting in the transition through the minimum:

$$\left. \begin{aligned} Q_{\text{melt}} &= T(S_{\text{liq}} - S_{\text{sol}}) < 0, & T < T_{\text{min}}, \\ Q_{\text{melt}} &= T(S_{\text{liq}} - S_{\text{sol}}) > 0, & T > T_{\text{min}}. \end{aligned} \right\} \quad (21)$$

Equation (21) shows that for temperatures below T_{min} , the solidification of He^3 must be accompanied by the absorption of heat; consequently, compression of He^3 under adiabatic conditions, which brings about a solidification, must lead to a decrease in temperature. This effect is known as the "Pomeranchuk effect." The variation of the entropy of solid He^3 of low temperatures has also been considered by Primakoff.⁶⁹ According to the Primakoff theory, the spins of the nuclei in solid helium are established in antiparallel states at temperatures much lower than the above pressure. Thus, for a pressure of 30 atmos, the orientation of spins begins at 0.5°K while at a pressure of 10^3 atmos the ordering begins at a temperature of $T = 10^{-3}^\circ \text{K}$. In this connection, the entropy of solid He^3 increases with pressure and under adiabatic compression, the He^3 must experience a cooling. However, in contrast to Pomeranchuk, who predicted a very strong cooling suddenly to $T = 10^{-6} - 10^{-7}^\circ \text{K}$, the cooling according to Primakoff under the same conditions is much less.

The inclination of investigators to explain the character of the entropy diagram of He^3 is made clear from all of the above. Unfortunately, at the present time there are not sufficient direct measurements of a specific heat of the liquid at various pressures as also for the specific heat of solid He^3 which would permit a direct verification of the predictions of the theory. However, on the basis of the existing experimental data, it is now possible to make some preliminary calculations of the entropy of the solid He^3 and liquid He^3 under pressure. An entropy diagram of He^3 is shown in Fig. 18. This was calculated by Hammel, Sherman, Kilpatrick, and Edeskuty⁵⁷ on the basis of a generalization of all the experimental data known at the present time. In this diagram, the entropy of the liquid at $T = 1.5^\circ \text{K}$ and pressure of saturated vapor, $S = 2.614 \pm 0.03$ cal/deg-mole,

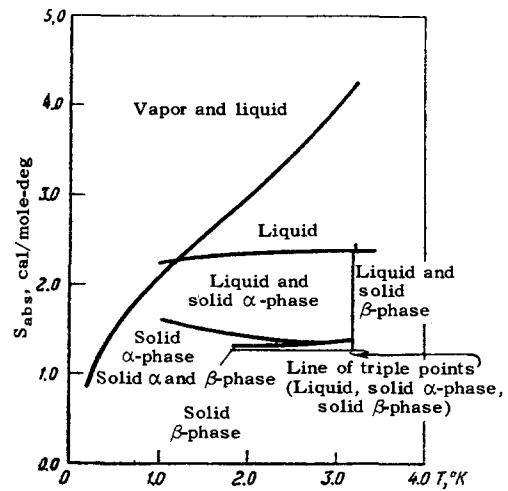


FIG. 18. Entropy diagram of He^3 .

was determined from the value of the heat of vaporization of He^3 , measured calorimetrically with great accuracy for this purpose by Osborne and Weinstock,⁵⁹ and from the entropy of the vapor. The calculations were made according to the formula

$$S_{\text{liq}} = S_{\text{vap}} - \frac{\Delta Q_{\text{evap}}}{T} \quad (22)$$

Throughout the entire temperature interval, the entropy of the liquid under the saturated vapor pressure was determined from the data on the specific heat of Roberts and Sydoriak⁵⁸ and Brewer, Sreedhar, Kramers, and Daunt.⁶⁶ The change in the entropy of the liquid with pressure was taken into account by the formula

$$\Delta S_{\text{comp}} = - \int_{p_{\text{sat}}}^p \left(\frac{\partial V}{\partial T} \right)_p dp, \quad (23)$$

where the value of $(\partial V / \partial T)_p$ as a function of p was taken from the data of Sherman and Edeskuty.⁴⁹ In this way separate isobars were obtained (not shown in Fig. 18) throughout the whole region of existence of the liquid from the pressure of saturated vapor to the solidification curve. The entropy of the liquid in equilibrium with the solid phase was computed from the data of Weinstock, Abraham, and Osborne,⁴⁵ Sherman and Edeskuty,⁴⁹ and Grilly and Mills.⁵¹ The entropy jump in the transition of the liquid to the solid state in the region of high temperatures (from 1.9°K and higher) was computed from the data of Grilly and Mills,⁵¹ who measured $d p_{\text{melt}} / d T$ and the change of the molar volumes upon melting. At the lowest temperatures, because of lack of experimental data, the curves of the change of molar volumes and the corresponding entropy were obtained by extrapolation. The data of Grilly and Mills⁵¹ also permit one to compute the entropy jump in the

solid He³ in the transition from the α to the β phase.

Consideration of the entropy diagram of He³ shows that with decrease in temperature the entropy along the solidification curve decreases, while along the melting curve in the region of comparatively high temperatures it remains constant and even increases slightly. Extrapolating the curves in the direction of lower temperatures, the authors established the fact that the curves intersect at $T = 0.35^\circ\text{K}$, which, according to the thermodynamic relations (20), must be a minimum on the melting curve. The entropy diagram after Hammel, Sherman, Kilpatrick and Edeskuty⁵⁷ is shown in Fig. 19. This contains a number of isobars and additions by means of extrapolation of curves in the direction of lower temperatures. The extrapolation was carried out on the basis of known experimental data and theoretical assumptions.^{69,70} Some doubt on this diagram exists for the region of the curve for the entropy of solid He³ below 0.3°K . The curve was carried out in accordance with the experimental data of Fairbank and Walters⁷¹ on the magnetic susceptibility of He³ and the theory of Primakoff.⁶⁹ However, there is some basis for assuming that the measurements of Fairbank and Walters, which give some evidence to the phenomenon of ordering of the spins in solid He³ beginning at 0.3°K , are not reliable. We shall give further consideration to the experiments of Fairbank and Walters below. We note that the present entropy diagram, constructed in the region of low temperatures by means of rather arbitrary extrapolation, is only a certain approximation to reality, and therefore does not provide a serious basis for making a choice in the use of

this or that theory. For solution of the problem new reliable experimental data are necessary.

MAGNETIC PROPERTIES OF He³

The magnetic properties of He³ are of special interest, inasmuch as, in contrast to He⁴, the He³ atom possesses a magnetic moment that is different from zero and a spin of one half. The magnetic moment of He³, according to the measurements of Anderson and Novik,⁷² is equal to 0.763 ± 0.007 of the magnetic moment of the proton, or 1.07×10^{-23} cgs. According to the expressions of Pomeranchuk,⁴⁶ an ordering of spins in parallel or anti-parallel states has already begun at temperatures below 1°K because of exchange effects in the liquid He³. In solid He³, where the amplitude of the zero point vibrations is less than the distance between atoms, the exchange interaction is practically non-existent and the ordering of the spins begins only as a consequence of the interaction of the magnetic moment of the nuclei at temperatures of the order of 10^{-6}°K . Primakoff,⁵⁹ in contradiction to Pomeranchuk, assumed that the atoms in solid He³ have already begun to be ordered by their spins in anti-parallel arrangements at temperatures below 0.6°K . It should be noted that a considerable amount of theoretical research has been devoted to the problem of the ordering of nuclear spins. The criterion of their validity can be found by the investigation of the magnetic properties of He³.

The first experiments on the determination of the magnetic properties of He³ were undertaken by Sydoriak and Hammel,⁷³ and Hammel, Laquer, Sydoriak and McGee.⁷⁴ Measurements were carried out by two methods. In the first method the displacement of the frequency of an oscillator op-

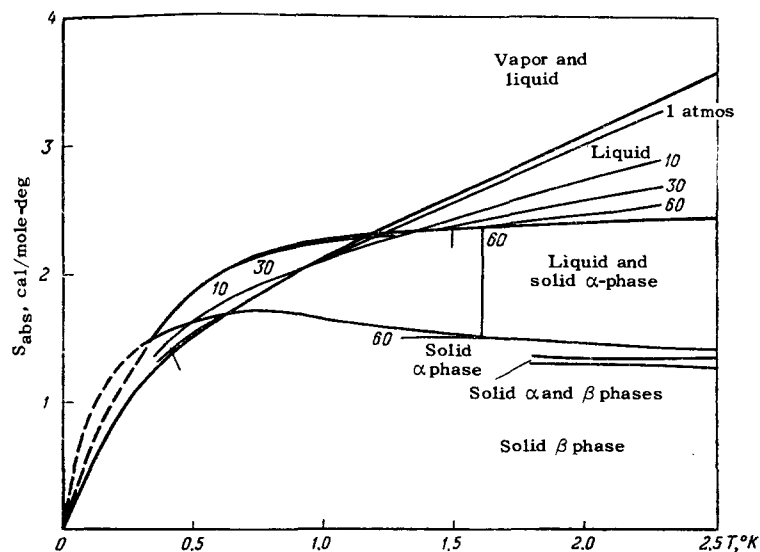


FIG. 19. Entropy diagram of He³.

erating at a frequency of 1.47 Mc was observed in the condensation of He^3 inside its coils, and under a change of temperature of the helium from 2.08 to 1.27° K. No such displacement was observed, with accuracy up to 5 cps; this meant that the volume magnetic susceptibility of liquid helium was less than 5×10^{-6} .

Such a method would not give positive results even in the case in which the He^3 would be ferromagnetic and oriented by the magnetic field of the earth while the variable field, because of the large relaxation time, would not be able to orient the domains. In order to exclude this possibility, the authors undertook experiments with a frequency of 60 cps. Resonance was observed between the two coils when the He^3 in one of them was condensed at $T = 0.904^\circ \text{K}$, or condensation took place at $T = 2.51^\circ \text{K}$ while thereafter the temperature dropped to 1.35°K in a period of 45 minutes. In both cases, with accuracy limited by experimental errors ($\sim 5\%$), the effect was not observed. Thus, although the experiments carried out did not possess sufficient sensitivity to measure the magnetic susceptibility of liquid He^3 , they did permit it to be established that He^3 did not become ferromagnetic suddenly, down to $T = 0.9^\circ \text{K}$.

Fairbank and his co-workers^{76,77,78} developed a more sensitive apparatus, making use of the method of paramagnetic resonance discovered by Zavoiskii.⁷⁵ The most complete description of the experiments is contained in the material of the conference which took place at Ohio State University.⁷⁶ A block diagram of the Fairbank apparatus is shown in Fig. 20. The specimen was placed inside a coil which connects to a tuned resonant circuit. A magnetic field of about 9000 Gauss was applied perpendicular to the axes of the coil. An alternating current was introduced into the coil from the oscillator, which worked at a stabilized frequency of 30 Mcs.

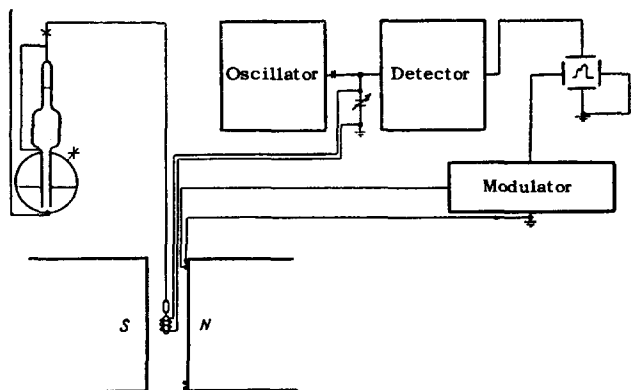


FIG. 20. Simplified circuit for the measurement of paramagnetic resonance in He^3 .

The loading of the resonant circuit was brought about by a high impedance circuit which guarantees the constancy of the amplitude of the driving current. Under these conditions the signal taken from the coil is proportional to the Q factor of the resonance circuit. With the aid of a modulator, the signal of which is simultaneously applied to the output of the oscillograph and to an additional coil on the magnetic poles, the magnetic field is easily changed by about 9000 Gauss. In the time of onset of paramagnetic resonance, a strong absorption appeared in the form of high frequency vibrations with corresponding decrease in Q of the circuit. The signal obtained from the circuit was amplified, attenuated, and delivered to the oscillograph where at the time of arrival of resonance it underwent a clearly marked maximum. The area under the maximum on the oscillogram was proportional to the magnetic susceptibility of the specimen. Inasmuch as the characteristic width of the resonance line of He^3 was less than the width produced by the inhomogeneity of the magnetic field along the specimen, the form of the resonance curve of the oscillograph was an unknown at the time of the experiment and therefore the comparative change in the magnetic susceptibility of the specimen was directly proportional to the amplitude of the maximum. For measurement of the susceptibility of the liquid He^3 above 1.2°K ⁷⁷ and in the gas at 4.2°K a small glass sphere was used just as shown in the circuit of Fig. 20 with a glass capillary from the Toepler pump. The coil of copper wire was fastened directly on the sphere.

For measurement of susceptibility for temperatures below 1°K ,⁷⁷ the apparatus pictured in Fig.

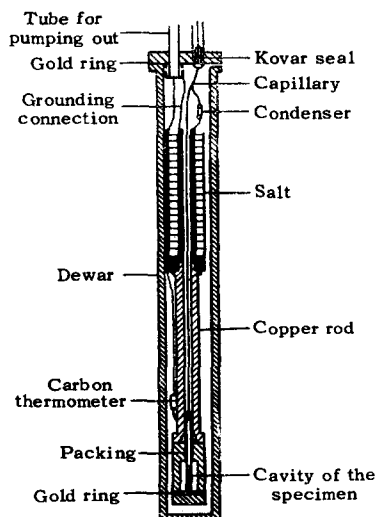


FIG. 21. Apparatus for the measurement of the susceptibility of He^3 at temperatures below 1°K .

21 was used. The liquid He³ was placed inside the glass tube of volume 15 mm³, which was placed in a copper cavity. The coil for nuclear resonance was placed inside a glass tube. Lower temperatures were obtained with the help of the demagnetization of 25 g of potassium-chrome alum, pressed, as is seen from the drawing, around the copper cavity at some distance from the measuring volume. For the experiments He³ was used which was concentrated by the authors by fractional distillation to a purity of better than 99%.

The volume magnetic susceptibility of He³ was directly measured with the aid of this apparatus in arbitrary units. On the basis of these data and the data of Kerr⁵³ for the density of liquid He³, the relative molar magnetic susceptibility of liquid He³ was computed. Below 1.2° K, the density of liquid helium became constant, inasmuch as the estimates⁷⁸ of the density of liquid He³ according to the change in the volume of the compressed vapor upon heating of the specimen from 4.2 to 1.2° K showed that for all pressures and temperatures from 0.2 to 1.2° K, the density of liquid helium changes by less than 1%.

The results of the measurement are shown in Fig. 22. The lower curve corresponds to measurement of the magnetic susceptibility of liquid He³ at saturated vapor pressure. It should be noted that the lower curve agrees within the limits of error of measurement with the theoretical curve for the ideal Fermi-Dirac gas with a degeneracy temperature $T_0 = 0.45^\circ\text{K}$. The measurements for solid helium are also part of the same drawing; however, at low temperatures, the results correspond better to liquid than to solid helium. The He³ could melt in the apparatus of the authors, inasmuch as the pressure of its melting at $T \approx 0.3^\circ$ increases with decrease in tem-

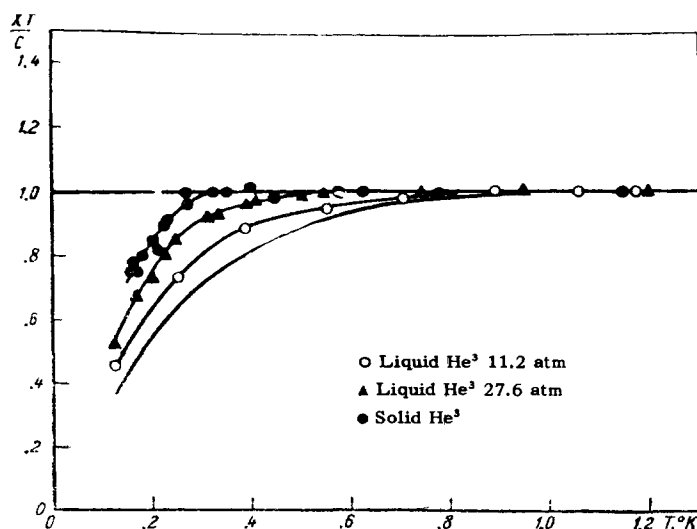
perature. Moreover, the same authors show that their results below 0.4° sometimes lead not to negative but to positive deviations of the magnetic susceptibility of solid He³ from the Curie law.⁷⁹ It is true that the lack of definition can also be explained by the fact that in this temperature region the possibility is not excluded of the existence of one of the two phases α or β observed⁵¹ in solid He³.

Besides the measurements described, Walters and Fairbank⁷⁸ determined the dependence of the volume magnetic susceptibility and the density of liquid helium on pressure at $T = 1.2^\circ\text{K}$, using apparatus similar to that pictured in Fig. 21. Their results are given in Table XII. The authors assume that the magnetic susceptibility under these conditions is proportional to the density; with accuracy of the order of errors of measurement (5%) this is confirmed by the change in the amount of He³ condensed in the vessel, and also

TABLE XII. Dependence of the magnetic susceptibility and density of liquid helium on pressure at 1.2° K

Pressure (atmospheres)	Volume susceptibility (relative units)	Density (g/cm ³)
0	1.00	0.0815
0.62	1.02	0.0831
0.97	1.04	0.0847
3.75	1.12	0.0913
6.89	1.20	0.0978
10.8	1.24	0.101
21.7	1.34	0.109
32.8	1.40	0.114

FIG. 22. Dependence of $\chi T/C$ on temperature for liquid and solid He³ (χ = molar susceptibility, C = Curie constant).



by the agreement of these data with the results of the measurement of the density of helium carried out by Sherman and Edeskuty.⁴⁹

W. Fairbank and Walters⁷⁶ also undertook measurements of the relaxation time for the establishment of equilibrium of the magnetic state of liquid He³ after a significant decrease in the amplitude of the variable field, which was initially maintained at a level which saturates the nuclear resonance. As the relaxation time T_1 they use the time during which the signal increases from zero to a value $1/e$ of its final value. In liquid helium at the saturated vapor pressure, the value of T_1 appeared to be irreproducible from experiment to experiment and varied from 20 to 200 seconds. The latter circumstance lead the authors to the idea that T_1 is determined by the effect of the walls. However, investigation of the dependence of T_1 on pressure during a single experiment showed that T_1 does not increase with pressure as would be the case for a wall effect, but rather decreases.

The decrease of T takes place, at first glance, in correspondence with the theory of Bloembergen, Purcell and Pound⁸⁰ who, taking into account Brownian motion, obtained the expression $T_1 \sim Da/\rho$, where D is the self-diffusion coefficient, D is the density of He³ and A is the effective radius of an atom of He³. Fairbank and Walters, making use of experimental data for density⁵³ and diffusion⁸¹ and setting $a \sim 1 \text{ \AA}$, obtained a value $T_1 \sim 200 \text{ sec}$. However, the irreproducibility of the experiments rendered the latter conclusion doubtful. One should rather recognize that the relaxation time is determined by the impurities, the content of which per unit volume is proportional to the density of the liquid He³. The measurements of these authors also showed that the relaxation time T_1 in the solid phase amounts to about 2 sec. Garwin and Reich⁸² reported at the Low Temperature Conference in Leyden (June, 1958) that measurements of the diffusion D and the relaxation time T_2 , which corresponds to the interaction of magnetic nuclei of He³, have been carried out by them. Making use of the spin echo method, they determined that D always decreases from 3 to 1.8° K and, extrapolating from 1.1° K to 0° they obtained a final value $D = 2 \times 10^{-5} \text{ cm}^2/\text{sec}$. The value of D changes with the volume of the liquid V as V^3 , increasing more than sixfold. Simultaneous measurements of D and T_2 in co-existing liquid and solid He³ showed that for the same temperatures and pressures, the relaxation time T_1 and D in the solid helium are three orders of magnitudes smaller than in the liquid.

ABSENCE OF SUPERFLUIDITY

It was established in the very first experiments with liquid He³ that it is a colorless, transparent liquid which boils rapidly upon pumping and which in external form is in no way different from He-I. Repeated attempts to find a phase transition in He³ similar to the λ -transition in He⁴ have given a negative result. Among the number of such attempts were those of Abraham, Osborne and Weinstock,⁸³ who investigated the flow of liquid He³ in 1949 through a slit of width 1μ in the temperature range from 3.02 to 1.05° K. The slit was formed by a gap between a platinum wire and a pyrex glass capillary which is put on the wire in a heated state. Because of the difference of the expansion coefficients, such a union at lower temperatures leaves a gap, the size of which is determined by the velocity of flow of the gas at $T = 4.2^\circ \text{ K}$. The capillary connects two bends of a U-shaped tube, in one of which the liquid under investigation is concentrated. The fluidity of the liquid was determined from observations on the rate of increase in pressure in the evacuated chamber connected to the other end of the capillary. In contrast with He⁴, the rate of flow of which through the slit increased sharply in the transition through the λ point, a monotonic decrease in the velocity of He³ was observed with decrease in temperature, testifying to the absence of superfluidity in the range 1.05 – 3.02° K. The results of these measurements

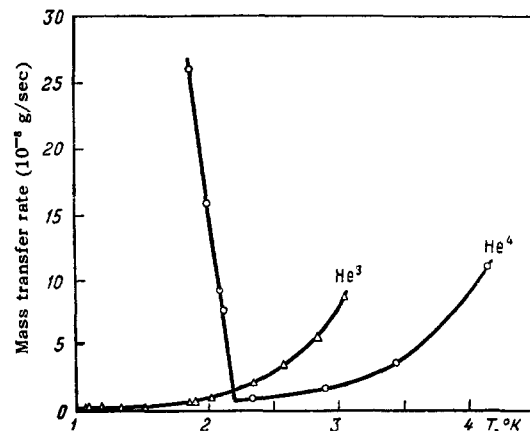


FIG. 23. Mass transfer rate of liquid He³ and liquid He⁴ through a $7 \times 10^{-3} \text{ cm}$ slit vs. temperature.

are plotted in Fig. 23. By means of subsequent investigations of the properties of He³ at much lower temperatures, it has been shown that superfluidity is absent in He³ below 1° K also. Thus, for measurements of the viscosity or the thermal conductivity of He³, to which we shall refer below, it follows that, down to temperatures of 0.2 or 0.3° K, He³ be-

haves as an ordinary liquid. Experiments on the determination of the λ curve and the diagram of separation of He^3 - He^4 lead to the same results.^{84,85,86} These experiments show that mixtures with concentrations above 82% (according to the data of reference 86) remain normal down to the lowest temperatures to which experiments have been carried (0.4°K). Extrapolating the curve of the λ -transitions of the mixture in the direction of higher concentrations, it can be established that superfluidity is in all probability always absent in pure He^3 . However, inasmuch as the possibility of superfluidity is not excluded in principle for He^3 by the general theory of quantum liquids, further experiments are necessary for a final conclusion at still lower temperatures, in particular experiments in the region of the ordered state of the spins of the nuclei.

VISCOSITY

From the qualitative measurements of Abraham, Osborne and Weinstock⁸³ on the overflow of He^3 through a small slit, it is possible to estimate the magnitude of the coefficient of viscosity and its temperature dependence. In the decrease of temperature from 2.79 to 1.04°K , the viscosity of the liquid changed from 22 to $30.4\mu\text{P}$.

Quantitative measurements of the viscosity were completed quite a bit later. In 1957, Taylor and Dash⁸⁷ measured the viscosity of He^3 by means of the method of torsional vibrations of a disk in the temperature range $1.3 - 2.15^\circ\text{K}$ and at $T = 3.34^\circ\text{K}$. The apparatus of Taylor and Dash is pictured in Fig. 24. A duraluminum disk of diameter 12.72 mm and thickness 1.34 mm was rigidly connected with a pyrex glass rod R of length 76 cm and diameter 0.8 mm , which was suspended from a platinum wire F in the glass container C , which was filled with liquid He^3 . The chamber had a height of 20 mm and diameter 25 mm . The distance between the disk and the bottom of the chamber was 5 mm . The temperature of the liquid was determined from the vapor pressure of He^3 . The damping decrement of the vibrations of the disk was measured by means of a mirror and scale. To decrease the effect of the mechanical vibrations, the cryostat was rigidly connected to a massive steel framework mounted on a cork base. The effect of electric fields created by static charges on the walls of the chamber and the dewar was decreased by covering the inside wall of the chamber and the surface of the rod R by a semi-transparent silver film, which guaranteed electrical contact between the disk and the

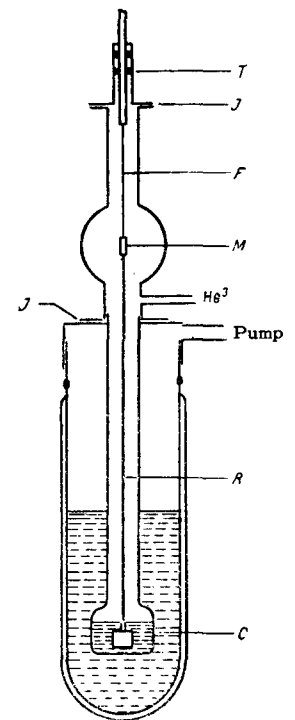


FIG. 24. Apparatus for the determination of the viscosity of He^3 .

chamber. As a consequence of the formation of bubbles and convective currents in the liquid, the reproductibility of results was not too satisfactory. In the temperature range $1.3 - 2.2^\circ\text{K}$, the mean deviation of the results amounted to $\pm 2.5\%$. Above 2.2°K , the deviation reached $\pm 10\%$, and the authors did not think it possible to include these data in the table, except for the value at $T = 3.35^\circ\text{K}$ (see Table XII). In absolute value and temperature dependence of the coefficient of viscosity, the results of Taylor and Dash⁸⁷ agree with the data of Abraham, Osborne and Weinstock.⁸³

In 1958, measurements of the coefficient of viscosity were carried out over the wide temperature range from 0.35 to 3.2°K by Zinov'eva⁸⁸ by the method of a capillary viscometer. Figure 25 pictures one of the instruments used for the measurements in the region of low temperatures. The apparatus consisted of a Dewar i of volume of about 3 cm^3 and diameter 8 mm , from which the vapors of liquid He^3 were pumped off. The liquid flowed through a thin-walled capillary a , at the front of which the reservoir b was attached by BF cement. For protection of the capillary from contamination, the filters e , l , and d of large area (diameter 5 mm) were located at the entrance and exit. These were made of very fine cotton fiber, which held 0.1μ particles. The temperature of the liquid was determined by the thermometer R' of phosphor bronze wire. The reservoir b , together with the capillary, was suspended by the wire g and moved with the aid of

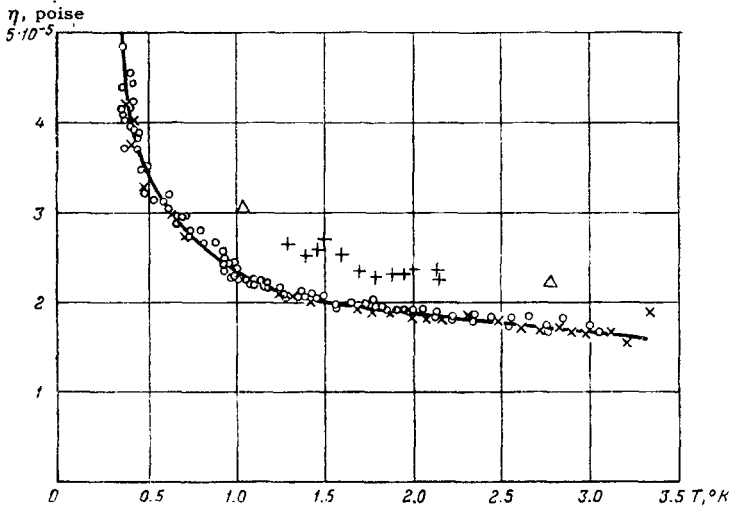


FIG. 26. Viscosity of liquid He³: o, x - Zinov'eva⁸⁸ (o - capillary diameter 102.1 μ, x - capillary diameter 74.5 μ); Δ - Abraham, Osborne and Weinstock,⁸³ + - Taylor and Dash.⁸⁷

a pressure of about 2 or 3 atmos in the temperature range 0.2 - 2.7° K. The authors used an apparatus (Fig. 28) similar to the apparatus of Fairbank and Wilks⁹⁴ for the measurement of the thermal conductivity of He⁴ below 1° K. The He³ was condensed in a thin copper-nickel capillary A (inside diameter 1.39 mm and length 2 cm) surrounded by a vacuum jacket J. On the upper soldered end of the capillary there was placed an electrical heater H; the lower end of the capillary made contact through the copper C with the salt P of chromium-potassium alum, which was cooled by adiabatic demagnetization. The salt and the capillary with the vacuum jacket were isolated from the bath of liquid helium by a hermetically sealed chamber. In the propagation of heat downward along the capillary into the liquid column, a temperature difference appeared which was measured by two carbon thermometers, T₁ and T₂, placed

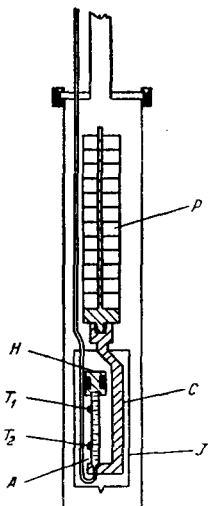


FIG. 28. Apparatus for the determination of the thermal conductivity of He³.⁹³

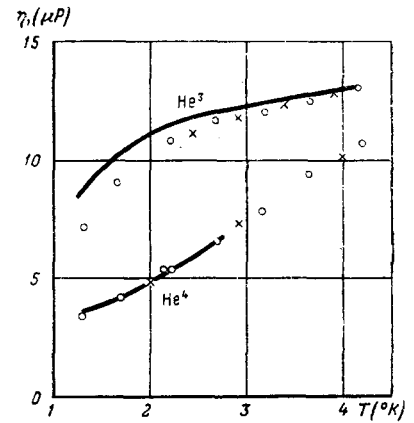


FIG. 27. The dependence of the viscosity of gaseous He³ and He⁴ on temperature. The circles and crosses are the experimental results obtained, respectively, for a lowering and raising of the temperature. The solid lines are the theoretical curves of de Boer and Cohen.

at the surface of the capillary. The correction for the thermal conductivity of the walls of the tube was made by separate measurements of the thermal conductivity of an unfilled capillary (the correction was very large, since the thermal conductivity of the tube exceeded in magnitude the thermal conductivity of a liquid column). The results of Fairbank and Lee are shown in Fig. 29 by black circles for He³ and open circles for He⁴. It is seen that in the temperature range 0.2 - 0.7° K, the thermal conductivity of the liquid He³ increases smoothly from the value 0.7×10^{-4} to 1.9×10^{-4} watt/cm-deg. The temperature dependence of the form $1/T$ expected for the coefficient of thermal conductivity of He³ from the theory of Pomeranchuk,⁴⁶ and also from the considerations of Abrikosov and Khalatnikov⁸⁹ evidently sets in below 0.2° K.

The results of Lee, Donnelly, and Fairbank were simultaneously confirmed by Challis and Wilks,⁹⁵ who measured the thermal conductivity

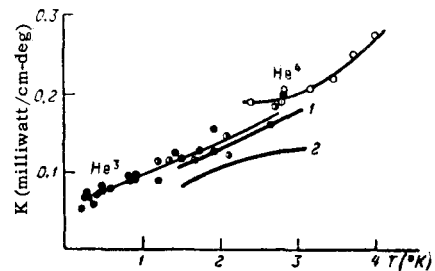


FIG. 29. Dependence of the thermal conductivity of liquid He³ and He⁴ on the Temperature. ●, ○ - Fairbank and Lee,⁹³ 1 - thermal conductivity of He³ (Challis and Wilks⁹⁵), 2 - thermal conductivity of gaseous He³ (Challis and Wilks).

of liquid He^3 (95% He^3 , 5% He^4) in the temperature range from 1.3 to 3.0°K. The authors possessed a small amount of He^3 and this determined the construction of their apparatus, which did not allow them to eliminate the resistance of the boundary between the solid and liquid He^3 . (As the experiments of Lee and Fairbank⁹⁶ have shown, to which we shall make reference below, such a resistance appears not only in He^4 but also in He^3 .) In the apparatus of Challis and Wilks, measurements were made on the temperature difference arising in the presence of heat flow on the edges of the plane, cylindrical layer of He^3 included between two copper blocks and bounded on the sides by a thin german-silver tube. In the opinion of Challis and Wilks, their data were lowered by 10% for the reason given above. These data are shown in Fig. 29 by the solid line 1. With consideration of the corrections for the resistance of the bounding layers between liquid and solid, they agreed quite well with the results of Lee, Donnelly and Fairbank. It must be noted that, although Challis and Wilks consider the data of Fairbank and Lee⁵⁴ very reliable, even these data are not sufficiently trustworthy, inasmuch as there is no complete assurance that convective flow was completely eliminated in the apparatus of Fairbank and Lee and, in the second place, the corrections which are introduced for the thermal conductivity of the walls of the tube were numerically larger than the thermal conductivity of the liquid itself.

In addition to measuring the thermal conductivity of liquid He^3 , Challis and Wilks measured the thermal conductivity of the vapor with the same apparatus. The results of these experiments are also shown in Fig. 29 by the solid curve 2. It is easy to see that the coefficients of thermal conductivity of the liquid and gaseous He^3 approach each other very closely at high temperatures, which gives a basis for considering the transfer phenomenon in liquid He^3 in the higher temperature region from the point of view of gas kinetic theory. Actually, in the range of temperatures above 1°K, He^3 obeys the classical formula

$$\alpha = \frac{5}{2} C_v \eta, \quad (24)$$

very well. Here κ is the thermal conductivity, C the heat capacity, and η the viscosity of liquid He^3 . The values of κ , computed on the basis of this formula from known experimental data for C_v ⁵⁸ and η ,⁸⁷ agree rather closely with the directly measured values. Equation (24) ceases to be valid in the temperature range below 1°K.

TEMPERATURE JUMPS AT THE BOUNDARY BETWEEN He^3 AND A SOLID

As was recalled above, Fairbank and Lee⁵⁴ in experiments on the thermal conductivity of He^3 discovered temperature jumps at the boundary between the solid surface, which scatters the heat, and the liquid He^3 . Investigation of the temperature jumps were completed on the apparatus for the measurement of thermal conductivity (Fig. 28) with a small modification. The liquid He^3 was condensed in a thin-walled german-silver tube (Cu-Ni) of diameter 1.5 mm, covered above and below by probes of pure copper. A heater and a thermometer were attached to the upper copper stopper, two other thermometers were located along the german-silver tube on its surface. A fourth thermometer served for the paramagnetic salt and was located in contact with the lower copper stopper. The temperature dependence of the resistance of the boundary is plotted in Fig. 30 in a logarithmic scale according to the data of Fairbank and Lee. For comparison purposes, similar data for He^4 taken from the researches of Fairbank and Wilks⁹⁴ are also plotted in the same drawing. It is easy to see that the temperature dependences of the boundary of the solid with the liquid He^3 and He^4 are identical and differ only in absolute value. The solid line drawn on the graph represents $R = 130/T^2$ for He^3 while the dotted line is $R = 45/T^2$ for He^4 . Thus the presence of the temperature discontinuity on the surface between the solid and liquid He^3 in the thermal transfer through the boundary corresponds to the fact that this property is inherent in all substances.

SURFACE TENSION

The coefficient of surface tension of He^3 was measured virtually simultaneously by Esel'son and Beresnyak,⁹⁷ Zinov'eva,⁹⁸ and Lovejoy⁹⁹ by

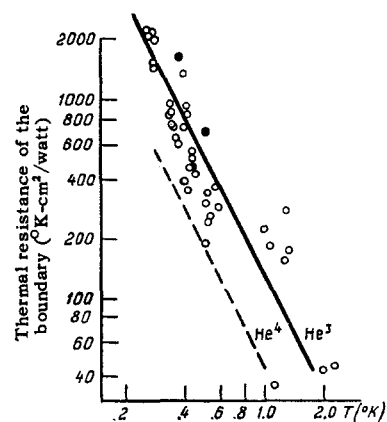


FIG. 30. Dependence of the thermal resistance of the boundary of He^3 and the solid upon the temperature.

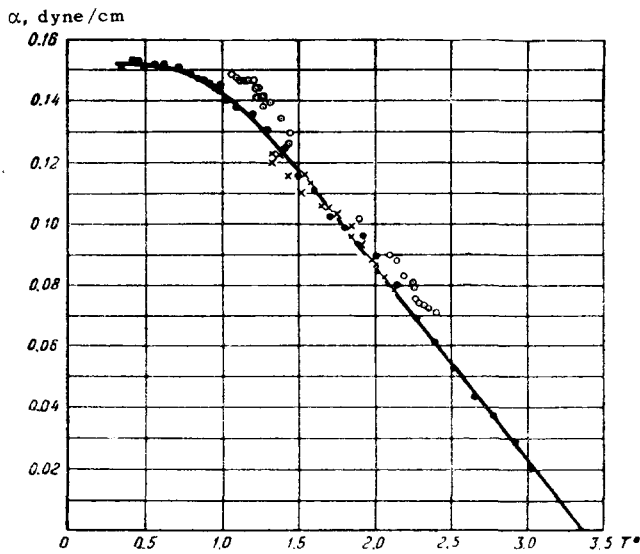


FIG. 31. Surface tension of He³. ●—Zinov'eva,⁹⁸
×—Esel'son and Beresnyak,⁹⁷ ○—Lovejoy.⁹⁹

the same method of capillary rise. The results of these experiments are shown in Fig. 31. The limiting value of the surface tension, $\alpha_0 = 0.152$ dyne/cm, was reached in He³ at $T = 0.6^\circ\text{K}$. This value is 2.3 times smaller than the limiting value of α_0 for He⁴. We note that the character of the temperature dependence of the surface tension in He³ and He⁴ is completely identical, as was shown in reference 98. In this connection, it is easy to be convinced by comparing the curves showing the dependence of α/α_0 on T/T_C for He³ and He⁴. Both curves agree within the limits of accuracy of measurement. Thus the different statistics of the two isotopes of helium and the states connected with this difference do not lead to change in the temperature variation of the surface tension in the liquid.

SPEED OF SOUND

In 1957, Flicker and Atkins¹⁰⁰ and simultaneously Lacquer, Sydoriak, and Roberts¹⁰¹ measured the speed of sound of liquid He³.

The measurements of Flicker and Atkins were carried out in a temperature range from 1.05 to 2.2°K by means of 14- μsec pulses. The authors used an electronic circuit similar to that described by Atkins and Chase.¹⁰² The sound was excited by means of a quartz crystal located at the bottom of a chamber of volume 0.12 cm³ at a distance of 1.013 cm from a reflector. In the temperature region studied, the speed of sound increased from 145 m/sec at 2.2°K to 1.70 m/sec at 1.0°K. The accuracy of the measurements was within $\pm 5\%$.

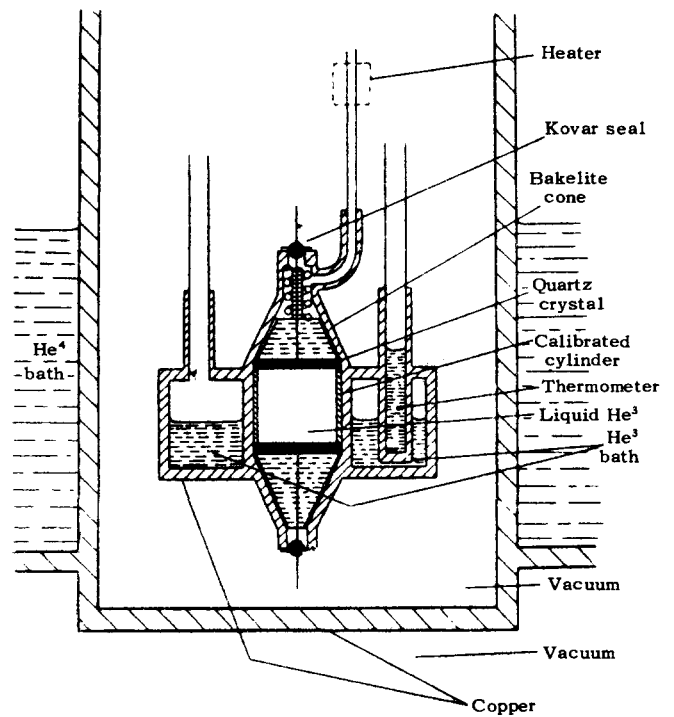


FIG. 32. Apparatus for measuring the speed of sound in liquid He³.¹⁰¹

Lacquer, Sydoriak, and Roberts¹⁰¹ measured the speed of sound in a much wider temperature range, from 0.37 to 3.14°K, using the apparatus shown in Fig. 32. The measurements were carried out by a pulse method at a frequency of 5 Mcs. The sound was excited by means of a quartz crystal located at the bottom of a thin brass cylinder of length 1.0447 cm. The vibrations were excited with a repetition frequency of seven per second, and had a duration of 0.4 μsec . A second crystal mounted above, served as a detector of the sound. The received signal, after amplification by a broad-band amplifier, was applied to the screen of an oscilloscope. By means of a special electronic circuit, the time delay of the received signal was determined with great accuracy relative to the incidence signal. The temperature of the liquid was determined according to the vapor pressure of He³. The values of the speed of sound above 2°K were determined by extrapolation of the results of measurement at much higher pressures to the pressure of the saturated vapor. The temperature dependence of the speed of sound according to the data of Lacquer, Sydoriak, and Roberts, are shown graphically in Fig. 33. The mean value of the sound velocity, with an estimate of the accuracy of measurement given by these authors,¹⁰¹ is given in Table XIV. The values of du/dp and the adiabatic compressibility χ are also included in this table.

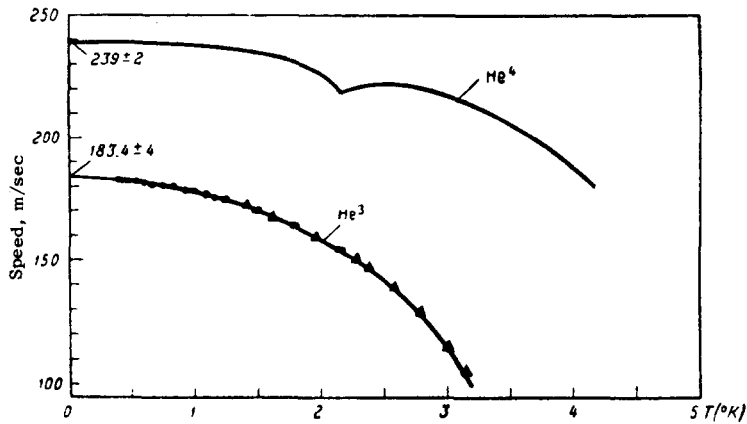


FIG. 33. Temperature dependence of the speed of sound in liquid He^3 .¹⁰¹

TABLE XIV

T , K	u , cm/sec	du/dP , cm ² /dyne-sec	ρ , gm/cm ³	χ , cm ² /dyne
He³				
0	$1.834 \pm 0.005 \cdot 10^4$	—	$8.235 \cdot 10^{-2}$	$3.61 \cdot 10^{-3}$
0.5	1.824 ± 0.003	—	8.22	3.66
1.0	1.780 ± 0.003	—	8.19	3.85
1.5	1.697 ± 0.003	$(1.9 \pm 0.4) \cdot 10^{-3}$	8.061	4.31
2.0	1.584 ± 0.003	(2.5 ± 0.3)	7.805	5.11
2.5	1.421 ± 0.004	(4.5 ± 0.5)	7.335	6.75
3.0	1.148 ± 0.005	(9.8 ± 0.8)	6.462	11.7
He⁴				
0	2.39 ± 0.02	—	14.6	1.20

COMPRESSIBILITY OF He^3

Direct measurements of the coefficient of isothermal compressibility $\chi = (1/\rho)(\partial\rho/\partial p)_T$ in He^3 were completed by Peshkov³⁸ by an optical method. In the determination of the isothermal compressibility, the temperature of the bath was kept constant and the displacement of interference bands was observed under the increase of pressure inside the optical chamber. The results of the measurements of Peshkov for liquid He^3 are shown graphically in Fig. 34.

Along with the direct measurements of the isothermal compressibility, indirect calculations of the coefficient of adiabatic compressibility of the liquid were carried out by Flicker and Atkins¹⁰⁰ and independently by Lacquer, Sydoriak, and Roberts¹⁰¹ on the basis of the measurement of the velocity of sound in liquid He^3 . The data of Lacquer, Sydoriak, and Roberts¹⁰¹ on the adiabatic compressibility of the liquid are given in Table XIV. These were computed from a knowledge of the velocity of sound u and the density ρ (according to the measurements of Kerr⁵³).

The coefficient of adiabatic compressibility of liquid He^3 computed by Flicker and Atkins¹⁰⁰ is shown graphically in Fig. 35.

ADSORPTION

The adsorption of He^3 in activated carbon at the temperatures $T_1 = 2.45^\circ\text{K}$ and $T_2 = 3.0^\circ\text{K}$ was determined experimentally by Hoffman, Edeskuty, and Hammel.¹⁰³ The results of the measurement are shown graphically in Fig. 36, where data for He^4 and N_2 are also plotted for comparison. The measurements showed that the volume of adsorbed gas that is necessary for the

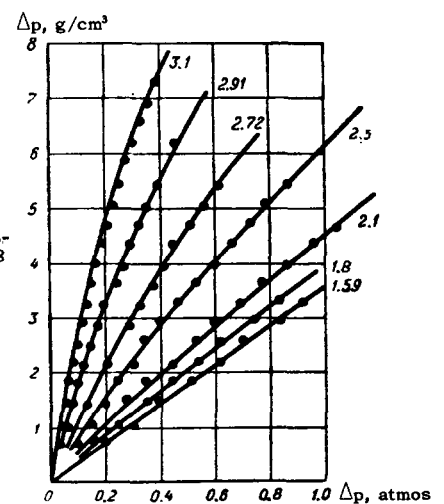
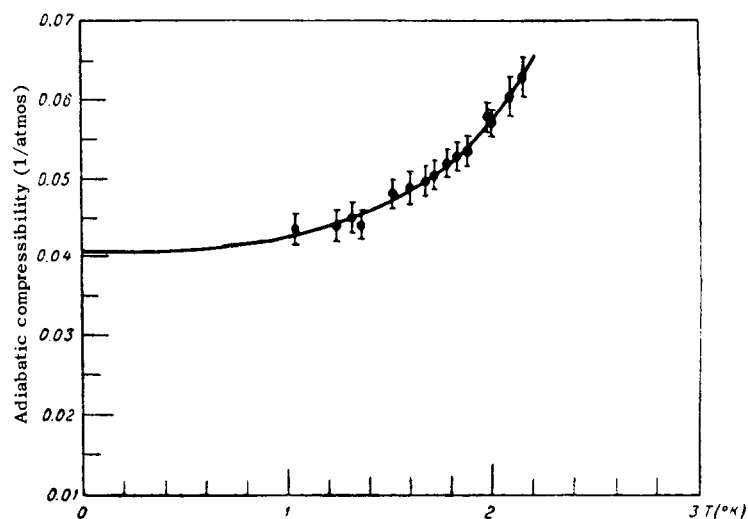


FIG. 34. Curves of the isothermal compressibility of liquid He^3 .³⁸ The numbers on the curves indicate T in deg K.

FIG. 35. Adiabatic compressibility of liquid He³.¹⁰⁰

formation of a monomolecular layer is almost the same for the two isotopes of helium and amounts to $0.43 \text{ cm}^3 \text{ (NTP)/m}^2$ for He³ and $0.45 \text{ cm}^3 \text{ (NTP)/m}^2$ for He⁴. In reference 103, the absence of capillary condensation was shown for pressures less than the boiling pressure. The heat of adsorption E_1 and the heat of vaporization E_{vap} of the isotopes of helium and nitrogen are given in Table XV.

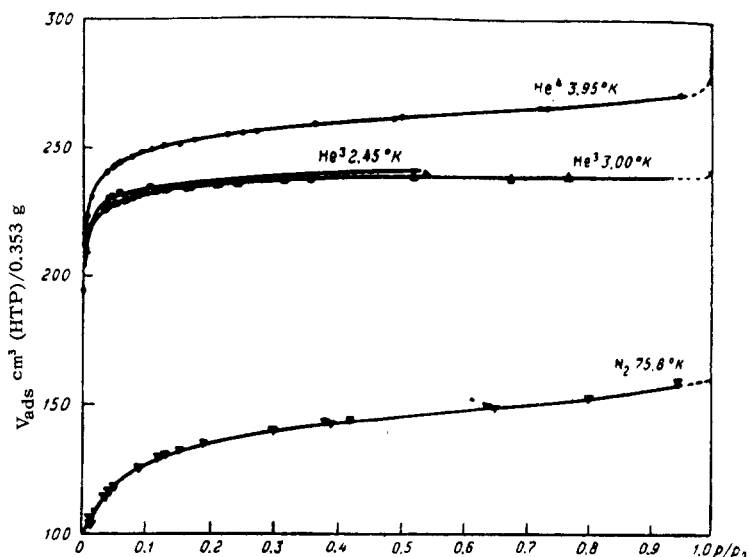
TABLE XV

Gas	$T^\circ \text{K}$	E_{vap} cal/mole	E_1 cal/mole
N ₂	75.6	1330	2100
He ⁴	3.95	20.95	71.2
He ³	3.00	7.88	45.3
He ³	2.45	10.59	42.4

CRYOSTATS WITH He³

The use of He³ instead of He⁴ for obtaining low temperatures by pumping of the helium vapor has a whole series of advantages. The vapor pressure of the He³ at 1°K is 35 times, and at 0.5°K 10,000 times, larger than for He⁴. Moreover, the presence in He⁴ of the superconducting film, which creeps along the walls of the Dewar upward and evaporates there, makes very difficult the obtaining of pumping at extremely low temperatures. The lower temperature obtained by pumping off He⁴ vapor that has been recorded is 0.71°K. Keesom¹⁰⁴ obtained it by using a pump of capacity 675 l/sec, while Lazarev and Esel'son¹⁰⁵ achieved it by means of a pump of capacity 15 l/sec and a diaphragm of diameter 0.05 mm directly under the level of the liquid helium.

The greatest difficulty in the use of He³ is the

FIG. 36. Adsorption of He³, He⁴ and N₂ in activated carbon.

necessity of becoming accustomed to small quantities of it, and to taking steps for its conservation. However, the undoubted advantages of He^3 for obtaining temperatures below 1°K has led to the development in a number of laboratories of cryostats with its use as a working substance.^{35,63,106,107,108} The cryostat which operates in the Institute for Physical Problems in Moscow¹⁰⁷ permits a visual observation at temperatures down to 0.3°K . The apparatus is pictured in Fig. 37 at the left while the circuit arrangements are shown at the right. The cryostat consists of two Dewars, 1 with He^4 , inside of which is placed a transparent glass Dewar 2 with liquid He^3 with volume of about 3 cm^3 . The He^3 vapor is pumped out by means of a thin-walled tube of stainless steel 3 of diameter 15 mm which is connected with a small Dewar by means of the copper connection 4. For protection from radiation above, the screen 5 was placed inside the copper connector and was in contact with the gas of He^4 .

The vapor was pumped out with a mercury diffusion pump DRN-50 (Fig. 37b) at a pumping rate of 30 l/sec, operating against a back-pressure of 25 to 30 mm mercury. A liquid nitrogen trap T was placed at the entrance to DRN-50. The exit to the pump is connected to the coil 6 which is immersed in the bath of liquid He^4 and which is connected by the valve 7 with the small Dewar. The coil goes successively through the outer and inner Dewars with liquid He^4 , in which the temperatures are maintained at 1.5 and 1°K , respectively, by pumping with separate pumps. Because of the low temperature of the bath of He^4 , the mercury pump DPN-50 can operate without a forevacuum, having at the exit a pressure approximately equal to the vapor pressure of He^3 at 1°K — (8 to 10 mm of mercury). The He^3 , condensed and cooled in the coil down to 1°K is by necessity poured into the small Dewar 2 by means

of the valve 7. In the case of a high temperature bath of He^4 , and also for the enrichment of He^3 in the apparatus, the Toepler mechanical mercury pump (T.P.) constructed by Danilov¹⁰⁹ is connected in series with the diffusion pump.

By means of this apparatus, temperatures of $T = 0.3^\circ\text{K}$ ($P = 0.002$ mm mercury) were obtained. The temperature of the liquid He^3 was determined by a resistance thermometer made of phosphor bronze wire of diameter 30μ , calibrated by means of the vapor pressure of He^3 and by the magnetic susceptibility of potassium chrome alum. The possibility of periodically pouring off the liquid He^3 permitted research to be carried on continuously in this apparatus at low temperature for a period of 8 — 10 hours with an unchanged supply of gaseous He^3 (about 3 liters). A temperature of 0.5°K was obtained within 5 to 10 minutes after pouring off. Upon cessation of pumping, warming up of the liquid from 0.3 to 1°K took not less than 3 hours.

A second type of instrument which is used in the Institute for Physical Problems in obtaining low temperatures in large volumes is shown in Fig. 38. The circuit arrangement is similar to that shown in Fig. 37. The Toepler pump (T.P.) is replaced by a hermetically sealed oil pump NVG-2 developed at the Vacuum Research Institute (NIVI).¹¹⁰ The gaseous He^3 is condensed in the coil 1 (diameter 2.0×1.4 mm, length 5 m), which is located in a bath of He^4 , and which is connected with the coil 3 and the He^3 storage tank 4 by means of the regulating valve 2. The coil 3 was prepared from a capillary of internal diameter 0.2 mm, length 200 mm. The He^3 flowing through the coil is additionally cooled by the vapors which are evaporated from the liquid below. The valve 2, which is located in contact with the outer tank, serves simultaneously as a screen from the radiation going upward. The storage tank

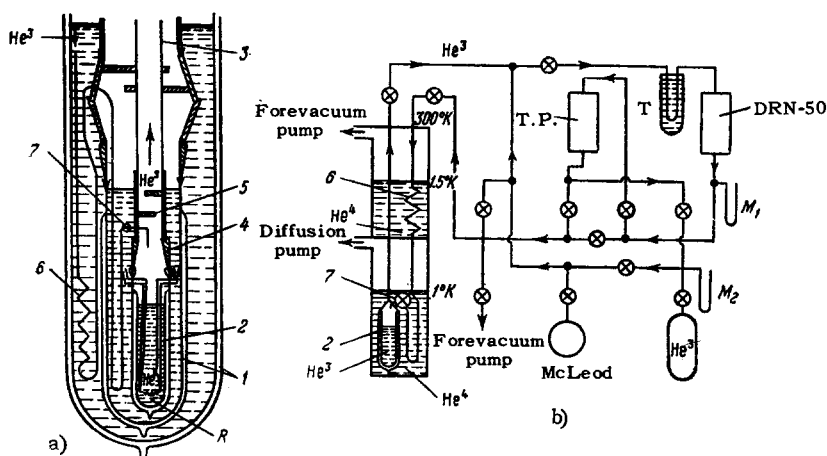


FIG. 37. Glass cryostat with He^3 and the circuit arrangement.

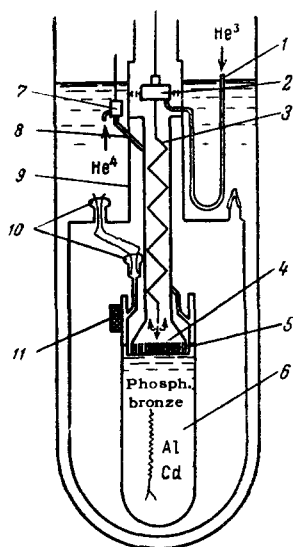


FIG. 38. Metallic cryostat with He^3 for continuous operation.

of He^3 (4) is a copper container which is solid soldered to a thin walled tube of stainless steel of length 100 mm, diameter 12×11.6 mm. The upper end of the tube is soldered to the body of the apparatus. A spiral copper band 5 of length 5 mm and thickness 0.3 mm, with separation between turns of 0.2 mm was soldered to the bottom of the container of He^3 . The increase in the surface of the container which is contact with the boiling He^3 is necessary for better transmission of cold from the surface (coldest) layer of He^3 to the walls of the container.

A copper vessel 6 of about 200 cm^3 capacity was soldered to the He^3 container. The liquid He^4 from the bath was poured into the container 6 by means of the valve 7 the german-silver tube 8 (diameter 2×1.4 mm, length 160 mm). The He^3 container and the vessel 6 are isolated from the helium supply by a vacuum jacket 9. Twelve electrical leads are passed through the vacuum jacket.

A screen 11 with activated carbon (1.5 cm) was fastened on the outside wall of container 6 in the vacuum jacket. The jacket was filled with He^4 under pressure $p = 0.5$ mm mercury at room temperature and sealed off. The presence of He^4 in the jacket decreased the cooling time of the vessel and the container of He^3 during the starting up of the cryostat. In establishing the low temperature, the He^4 in the jacket is absorbed by the carbon. The time necessary for cooling the vessel of the cryostat from $T = 4.2^\circ \text{K}$ to $T = 0.5^\circ \text{K}$ did not exceed one hour. The time of cooling the container of He^3 and the vessel with liquid He^4 from $T = 1.15^\circ \text{K}$ to $T = 0.5^\circ \text{K}$ amounted to 5 or 10 minutes. Regulating the velocity of pumping off of He^3 , and also the speed of its delivery into the container through the valve 2, it is possible to es-

tablish a temperature of 0.5°K and higher in the vessel. In a case in which there is no He^4 in the sphere, the temperature can be lowered to 0.35°K . The principal head lead to the vessel goes from the tank through the helium inside the tube 8. The temperature inside the vessel 6 is measured by a thermometer of phosphor bronze. As supporting points, on which each time the calibration of temperature is controlled, the points of transition to the superconducting state of wires of pure aluminum of diameter 50μ and cadmium of diameter 67μ connected in series with the thermometer.

Sydoriak and Roberts^{35,63} used the cryostat shown in Fig. 6 for measurement of the thermal properties and vapor pressure of He^3 . A Dewar with He^3 placed in a tank of He^4 was made completely of metal.

The He^3 was condensed in a seamless copper sphere of diameter 12.7 mm. Crystals of paramagnetic salt were placed inside the sphere for measurement of the temperature. The primary and secondary coils were wound directly on the brass vacuum jacket. For thermal isolation, the copper sphere was fastened to the brass block of the instrument by means of a tube of an alloy containing 70% Cu and 30% Ni of length 200 mm, diameter 3 mm with wall thickness 0.25 mm. Insert pieces could be screwed into the brass block for the measurement of the vapor pressure, the specific heat and other properties as desired. The insert for the measurement of the vapor pressure is shown in Fig. 6. Temperatures below 1°K were obtained by pumping off the He^3 vapor. The lowest temperature reached by the authors with this apparatus was 0.37°K .

Seidel and Keesom¹⁰⁶ developed a special type of cryostat pictured in Fig. 39 for the measurement of the specific heat in the temperature region around 0.3°K . It consists of a Dewar a with liquid He^4 , inside of which, in a vacuum jacket g, is placed a copper vessel consisting of three separate sections d, h, l. The liquid He^3 , which is condensed in the container d, is pumped off through the german-silver tube of diameter 4 mm and wall thickness 0.1 mm. The specimen i with a carbon thermometer and heater on it is suspended inside section h on a nylon thread, attached to the steel wire f of diameter 0.12 mm. The wire extended upward through the system of blocks l and capillaries b of diameter 1 mm. By means of the wires, the specimen could hang and be found under adiabatic conditions or could be lowered and in this way brought into thermal contact with the copper plate k. The plate was connected by a copper rod and by a copper jacket to the He^3 bath.

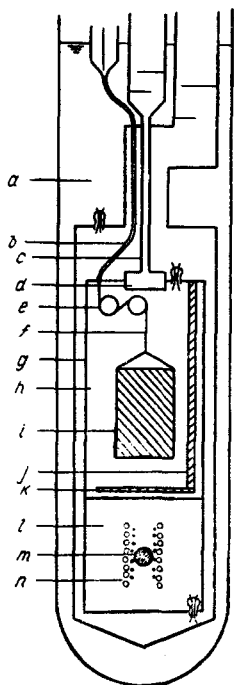


FIG. 39. Cryostat for the measurement of specific heat.

To speed up the cooling of the specimen, traces of air were left inside section h; these brought about rapid cooling down to 77° K. At lower temperatures, the air was frozen out and good thermal insulation was assured. For measurement of temperature, a powdered paramagnetic salt-iron ammonium alum — was place inside the section l in an atmosphere of He⁴ and soldered to the glass sphere m (at p = 1 atmos and T = 77° K). The measurements were carried out on an ac bridge. To avoid heating by eddy currents, the primary coil n was made of niobium wire (superconducting below 8° K) and was placed inside the section l, the walls of which were covered with a superconductor (50% lead, 50% tin). For thermal contact, the section l was soldered at room temperature with atmospheric pressure He⁴ inside it.

The pumping system for He³ is similar to that shown in Fig. 37, only in place of the mercury there an oil diffusion pump was used with a pumping speed of 25 l/sec, with a forevacuum oil rotational pump of speed 1.3 l/sec. The condensation of the gas was carried out not by means of a special capillary, but in the pumping tract. The temperature was regulated not by a change in the speed of pumping, as was the case in the previous sets of apparatus, but by absorption at the entrance of the diffusion pump of a regulated quantity of He³ from the supply.

The work on the apparatus proceeded in the following order. After cooling the equipment to 77° K, liquid helium is poured into the Dewar and is maintained at atmospheric pressure from the

moment the temperature of 4.2° K is reached by the specimen (from 8 to 24 hours). Thereafter, the remaining He⁴ is pumped out of the space inside the vacuum jacket g, the temperature of the bath is lowered to 1° K, and the He³ is condensed in the container d. After this, pumping is begun with the rotary pump, and the pressure in d decreases to 0.5 mm of mercury; the diffusion pump is then turned on. The specimen, which is located on the copper plate k, is cooled to 0.35° K approximately 6 hours after the temperature in the container d reaches 0.3° K. The low temperature is not decreased inasmuch as the thermal contact between the specimen and the copper at these temperatures is very weak and a heat lead of 0.1 erg/sec in the carbon thermometer produces a temperature difference of 0.05° K. This circumstance also permits a measurement of a specific heat of the specimen without lifting it, because in the lifting, heat is released which warms the specimen by several tenths of a degree, and temperature fluctuations appear that are brought about by mechanical vibrations. Above 1° K, the thermal insulation was insufficient and the specimen had to be lifted. Temperatures are plotted in Fig. 40

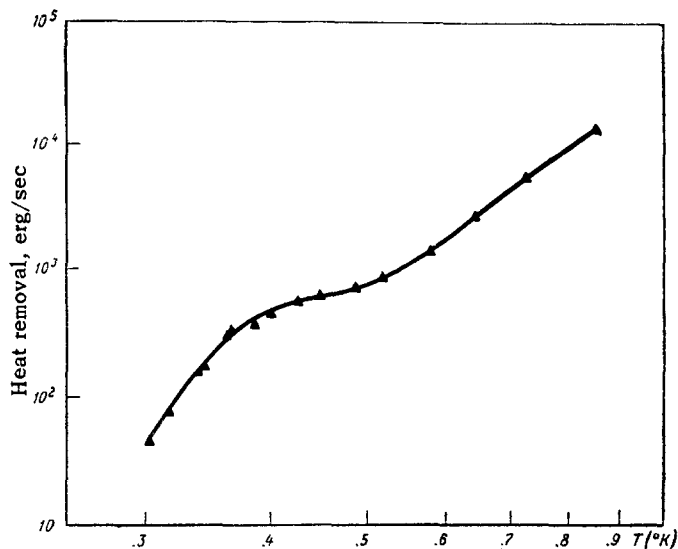


FIG. 40. Heating curve for liquid He³ (q vs. T).

which are established in the apparatus as a function of the heat generated. For minimum heat generators of the order of 100 ergs/sec, and for a supply of 3 l₀ of He³ in the container (at T = 0° C and p = 760 mm mercury), temperatures around 0.3° K could be maintained continuously for 80 hours.

As is seen from a description of the apparatus, the presence of 1 to 3 l₀ of He³ is quite sufficient for successful operation of the cryostat with He³

at temperatures below 1°K. For not very large heat removal, by using the first or second variant of the apparatus and a He⁴ bath at 1°K, one can get along with a single mercury diffusion booster pump and reduce the necessary amount of He³ to 0.05 – 0.1 l_0 . Reich and Garwin¹⁰⁸ reported on such a cryostat requiring merely 0.05 l_0 , at the Delft Conference. The advantages of such cryostats with He³ for research of temperatures from 0.4 to 1°K in comparison with methods using paramagnetic salts for He⁴ are quite evident.

OBTAINING LOW TEMPERATURES BY CRYSTALLIZATION OF He³

As was mentioned above, Pomeranchuk⁴⁶ proposed that while in liquid He³ the ordering of spins begins at temperatures below 1°K, the ordering in the solid begins only at $T = 10^{-7}$ °K. Primakoff⁶⁷ assumed that ordering in the anti-parallel state in solid He³ begins as early as $T \sim 0.5$ °K; however, even in this case, the entropy of the solid helium must be greater than that of the liquid He³. It is impossible, on the basis of current experimental data, to say definitely which of these assumptions is correct. The experiments of Walters and Fairbank⁴⁷ have shown that the melting of solid He³ at $T = 1.2$ °K is accompanied by cooling, while at 0.2°, by heating of the solid He³, that is, in the vicinity of 0.2°, the entropy of the solid helium is greater than the entropy of the liquid He³; therefore, in this region it is possible in principle to lower the temperature by adiabatic compression of helium. If the assumptions of Pomeranchuk are valid, it is possible to achieve theoretically temperatures of the order of $10^{-7} - 10^{-6}$ °K by adiabatically transforming liquid He³ into the solid state under pressure; however, the amount of cold stored in this way is very small. Actually, as soon as sufficient heat is conveyed to the solid He³ to increase the entropy to $R \ln 2$, the temperature rapidly increases to some tenths of a degree. Inasmuch as the heat is supplied at the level of $10^{-7} - 10^{-6}$ °K, then not more than $R \ln 2 \times 10^{-6} \sim 70$ ergs/mole is required for heating a gram atom of He³, or about 3 ergs/cm³. It is difficult to conceive of a compression process in which such an amount of heat would not be generated by internal friction.

However, one must point out the possibility of producing a thermostat for very low temperatures, making use of the increase in pressure in the melting of He³ with decrease in temperature, and consequently the absorption of heat in the crystallization under pressure. If we begin to raise adiabatically

the pressure of the He³ cooled to a temperature of 0.2 or 0.3°K, then, after attaining the melting pressure, crystallization begins, accompanied by a notable absorption of heat. The mixture of solid and liquid He³ begins to cool, and a temperature is established corresponding to the equilibrium at the given pressure. Continuing with a corresponding compression, to maintain a constant pressure, one can maintain a constant temperature, compensating the heat supply by the heat of crystallization. In this case the cold reserve will be greater the higher the regulating temperature, and its order of magnitude will be ΔST_p , i.e., about 10^{-3} cal/mole at $T_p \sim 10^{-3}$ °K.

Such a process also has the convenience that because of the very high interpenetration of the two phases, the temperature will be constant and homogeneous over wide limits, which is a not inconsiderable advantage, since the thermal conductivity of He³ is very small.

CONCLUSION

A review of experimental data shows that the chief properties of liquid and gaseous He³ in the range of temperatures from 0.2°K and higher are known, and the question reduces only to the exact determination of one parameter or another. In particular, more accurate experiments on the determination of the thermal conductivity of gaseous, liquid and solid He³ are desirable. The properties of solid He³ have been incompletely studied. It is interesting to obtain data relative to the variation of the curve of transition from α to β phase of He³ over all regions of temperatures corresponding to the existence of the two phases, and also data relative to the change in volume and heat content in the transition, and also the magnetic properties of the phases.

A great interest attaches to the properties of liquid and solid He³ at temperatures below 0.2°K. The temperature dependence of the entropy of solid He³ in this region is unknown; by its variation one could determine the character of the interaction between the spins of the He³ nuclei, which is very important for making clear the possibilities of the use of crystallization of He³ by pressure for obtaining and maintaining very low temperatures.

Below 0.05°K, the properties of He³ can be described theoretically on the basis of the concept introduced by Landau⁶⁷ of the spectrum of excitations of a Fermi liquid. This problem is discussed in more detail in the review of the Abrikosov and Khalatnikov.⁹⁰

There is considerable interest as to whether

He³ is a superfluid or not. Inasmuch as the atoms of the He³, similarly to the electrons in superconductors, are inclined to considerable pair interaction because of the presence of the nuclear spins, one can therefore expect the appearance of the superfluidity of He³ at very low temperatures. The solution of this problem has great basic importance.

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