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The 19 62 He3 **Scale of Temperatures II. Derivation^l**

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An Experimental Thermodynamic Equation (ETE) temperature scale valid from 0.2 to 2.0 °K has been calculated for He³. The scale is based on new comparisons, (P_3, P_4) , of He³ and He⁴ vapor pressures above 0.9 °K; on the 1958 He⁴ temperature scale; and on the best available data for several The T_{62} Full-Range Working Equation (FWE) scale,

 $\begin{array}{l} \ln\,P_3\!=\!-\!2.49174/T\!+\!4.80386\!-\!0.286001\,\,T\!+\!0.198608\,\,T^2\!-\!0.0502237\,\,T^3\!+\!0.00505486\,\,T^4\!\\ +2.24846\,\ln\,T\end{array}$

fits the ETE scale and the (P_3, T_{58}) data and is therefore valid for use from 0.2 to the critical point, 3.324 °K. The maximum deviation from the ETE scale is 0.4 mdeg and the standard deviation from the input data is 0.25 mdeg. The fit to the seven recalculated isotherms of Keller in the range of the 1962 He³ scale can be determined by converting Keller's P_4 's to equivalent P_3 's, using direct P_4 to P_3 interpolation equations. The fit of the 1962 He³ scale is as good as the fit of the 1958 He⁴ scale to the same isotherms, the average displace-
ments of the two scales both being 1.5 mdeg below the isotherms. The average standard deviations for $(T_{92}-T_{180})$ and for $(T_{98}-T_{180})$ are 1.2 and 1.0 mdeg, respectively, for these seven isotherms.

1. Introduction

At the time He^3 was first liquified $[39]^2$ a comparison of its vapor pressures, P_3 , with those of He⁴, P*⁴ ,* was given, along with a careful determination of its critical pressure, P_c , which was found to be 875 mm Hg. Subsequently, Abraham, Osborne, and Weinstock $(A\text{OW})$ presented [5] more accurate (P_3, P_4) comparisons, additional critical point data in agreement with ref. [1], and an empirical temperature scale, T_K , based on a modification [5] of the 1948 $He⁴$ scale $[41]$. They found it possible to fit all their data, ranging from 0.011 P_c (1.02 °K) up to P_c ,

to an equation for $\log (P/T^{5/2})$ having only three fitted powers of T.

In 1953, Chen and London [42] criticised the form of the T_K equation and attempted to fit the same (P_3, P_4) data of AOW to an equation having a proper theoretical form for extrapolation to 0° ^oK. Although they fitted coefficients of five powers of T , and omitted data at pressures above 0.4 P_c , the fit of their equation was not satisfactory above 0.1 *Pc.*

In 1957 Sydoriak and Roberts [9] extended the measured range of pressures and temperatures down to 0.000074 $\tilde{P_c}$ and 0.45 °K using two different paramagnetic salts calibrated against He^3 above 1° for the temperature measurement. At that time two newer He⁴ temperature scales were in use, so two He³ scales were calculated: the T_E He³ scale, based on the 1955E He^4 scale [43], and the T_L He^3 scale, based on the 1955L $He⁴$ scale [25].

¹Work performed under the auspices of the United States Atomic Energy Commission ² Figures in brackets indicate the literature references at the end of thir paper.

Only by abandoning attempts to retain an analytical expression of proper theoretical form for extrapolation to 0^oK did Sydoriak and Roberts find it possible to get a good fit to their data in combination with those of AOW. To cover the four orders of magnitude range in *P3* only four fitted powers of *T* were needed.

Following the adoption of the 1958 $He⁴$ scale [2] the present authors made a proposal [1] to the Seventh International Conference of Low Temperature Physics held at the University of Toronto in 1960 for a new $He³$ vapor pressure scale to be based on the 1958 He4 scale and on various thermodynamic properties of He³. The proposed procedure was similar to that used for the existing T_E and T_L scales except that newly available specific heat data [3, 4] could now be included instead of using a calculated "spin entropy" [44] term. In addition, a different magnetic temperature conversion was being studied for the paramagnetic salt data [9] intended to be used to extend the scale below $1 \text{ }^{\circ} \text{K}$.

The proposal was favorably received by members of the conference, with some reservations as to the feasibility of including vapor pressure data obtained with an iron alum thermometer.

We have subsequently abandoned incorporation
of any paramagnetic salt data into the scale derivation except for measurements of specific heat using a cerium magnesium nitrate thermometer [3].

An alternative procedure for establishment of the low-temperature end of the new $He³$ scale has been thoroughly discussed [10] in a report to the Fourth Symposium on Temperature, its Measurement and Control in Science and Industry, Columbus, Ohio, March 1961. In this method the thermodynamic consistency of the (P, T) data can be examined point bv point. The method showed [45] that the AOW data could not be combined with the 1958 He4 scale to yield a thermodynamically consistent scale in the range from 1 to 2 K . A detailed discussion of the inconsistency, which is equivalent to several millidegrees, is given in a companion paper $[7]$ to this one, hereafter referred to as Part III. Because of this inconsistency new (P_3, P_4) comparisons were undertaken [45] in an improved apparatus designed to mininize errors due to $He⁴$ film reflux. The results are reported in detail in another companion paper [46] to this one, hereafter referred to as Part I.

Since the measurements reported in Part I provide an explanation for the thermodynamic inconsistency, below T_{λ} , of a scale based on the AOW data, we have used only (P_3, P_4) data given in Part I in deriving the 1962 He^3 scale, reserving the AOW data above T_{λ} for the purpose of checking the final scale.

The second and third virial coefficients of He³ are needed to establish the low-temperature end of the present scale. For this purpose Keller's isotherm data [12] has been reanalyzed, using the method of multiple variable least squares [47, 48]. For a further discussion of the method and results we refer to Part III, in which the scale derived below is examined for consistency with isotherm, paramagnetic salt, and latent heat data.

2. The High-Temperature Working Equation Scale

The three steps required to arrive at a full range equation are those discussed in detail for a Method I

The first step is to derive a working equation scale by which one can interpolate between the (P_3, P_4) data points of Part I converted to a (P_3, T_{58}) table of data. The primary use of this scale is for making small corrections to experimental quantities of second order importance which enter into the thermodynamic treatment of the (P_3, T_{58}) data. It will suffice to state that we used a working equation scale which fits the *(P3,* T*58)* data with a variance of 0.25 mdeg and a maximum deviation of 0.6 mdeg. (Tn other Hes scale derivations a high-temperature working scale was considerably more important, since it was also used in the determination of paramagnetic salt calibration equations [9] and to assign temperatures to He^3 latent heat data [10].)

3. The Experimental Thermodynamic Scale

The second step is to derive an analytical expression for the thermodynamic vapor pressure equation (see eqs (2) and (4) of ref. [10]) which can be written as follows, putting on the left those terms which can be evaluated from existing thermodynamic data:

$$
\ln P_3 + f(T) + f(C_{\text{sat}}, T) - \epsilon + f(V_L, P_3, T) = -\frac{a}{RT} - \frac{b}{R}.
$$
\n(1)

The values of P_3 and T_{58} used are given in table 1, which is a portion of the data of Part I, excluding the lowest three data points, at $P_{4} \leq 40\mu$, because the calculated He4 film reflux pressure drop was excessive.

The second term is

$$
f(T) = -i - (5/2) \ln T \tag{2}
$$

where i is the chemical constant, $i = 5.31733$.

For the remaining terms on the left we write the thermodynamic function and its empirical equivalent as follows. The calculable part of the specific heat term is

$$
f(C_{\text{sat}}, T) = \frac{1}{RT} \int_{T_m}^{T} dT' \int_{T_m}^{T'} (C_{\text{sat}}/T'') dT''', \quad \text{(3a)}
$$

where C_{sat} is the specific heat of saturated liquid He³. Smoothed values of the C_{sat} data of Brewer, Sreedhar, Kramers, and Daunt [3] and data points of Weinstock, Abraham, and Osborne [4] are shown in table 2, and are used to obtain an empirical equation for $\dot{C}_{\rm sat}$

$$
C_{\text{sat, }x}/R = 0.25154 + 0.47485T - 0.54064T^2
$$

+ 0.406356T³ - 0.082729T⁴ (3b)

for $0.2 < T < 2^{\circ}$. In this and the following equations and tables the subscript, x , is used to designate either an empirical function of *T* fitted to a thermodynamic quantity or an experimental interpolation equation,

The lower limit of the fit, $0.2 \text{ }^{\circ}\text{K}$, was arbitrarily
selected within the range of measured values of C_{sat} . By this choice the lower limit of reliability of the 1962 He³ scale is chosen to be 0.2 K . By inserting eq (3b), the exact theoretical expression (3a) is converted to the experimental interpolation equation for $T_m=1.0^{\circ}$ and $0.2 < T < 2^{\circ}$.

$$
f_x(C_{\text{sat}}, T) = \frac{0.39332}{T} - 0.57013 + 0.237426T -0.090344T^2 + 0.033863T^3 - 0.0041364T^4
$$

 $+0.25154 \ln T$. (3c)

TABLE 1. He³ vapor pressure data^a used in deriving the 1962 He³ scale

Particle 11 and $\overline{H}e^y$ and $\overline{H}e^y$

In fitting the coefficients of the equation for the 1962 He³ scale, the weight given
to each data point was a function of both δP_{33} and δT_{35} . As expected, for almost all of the data points $(T_{92}-T_{58}) < (\delta T_{92}+\$

 $(T_{62}-T_{58})$.
The last entry in the table is the measured value of the critical point.⁴ In addition to the pressure measurement error listed, there is an uncertainty of ± 1.5
mm Hg in the location of the critical point.

TABLE 2. Values at selected temperatures, T, of the specific heat function^a, $f(C_{sat},T)$ appearing in the thermodynamic *equation for* $\ln P_3$

The function is evaluated by means of an explicit equation $^{\rm b}$, $C_{\rm sat}$, fitted to smoothed $C_{\rm sat}$ data of Brewer, Sreedhar, Kramers, and Daunt $^{\rm o}$ below $1^{\rm o}$ K and plotted data points of Weinstock, Abraham,

a Eq (3a)

b Eq (3a)

c Ref. [3]

c Ref. [4]. Both T and C_{sat} were adjusted to the T₅₅ scale; e.g., C_{sat} was multi-

d Hef. [4]. Both T and C_{sat} were adjusted to the T₅₅ scale; e.g., C_{sat} was multi

TABLE 3. The vapor volume term^a, ϵ , in the equation for In P₃, as evaluated by an iterative procedure, using a highspeed digital computer

For their general usefulness the table also shows solutions of the cubic equation b for the vapor volume V_G , and values of the second \circ and third \circ vircoefficients, B and C , based on reanalysis \circ of the iso

For convenience we have taken T_m to be 1.0 °K, although any other temperature in the range of eq (3b) could as well have been selected. The effect of the specific heat term on the ETE scale below 1 K is easier to identify with this choice of T_m .

^a Ref. [46] (Paper I of this series).
^b Ref. [7] (Paper III of this series).

The vapor volume term is

$$
\epsilon = \ln \left(P_3 V_c / RT \right) - \frac{2B}{V_c} - \frac{3C}{2V_c^2}.\tag{4a}
$$

For the vapor volume, V_g , we used the inverse vohune expansion form of the equation of state

$$
PV_{\sigma} = RT\left(1 + \frac{B}{V_{\sigma}} + \frac{C}{V_{\sigma}^2}\right)
$$
 (4b)

For the second and third virial coefficients of $He³$ we used the equations found in Part III in the multiple parameter least squares analysis of Keller's isotherm data [12],

$$
B=4.942 - \frac{270.986}{T} \text{ cm}^3/\text{mole}
$$
 (4c)

and

$$
C=2866/\sqrt{T}\,\mathrm{cm}^6/\mathrm{mole}^2.\tag{4d}
$$

Since a high-speed calculator was available, it was possible to use the implicit form (4a), with P*3* being taken from the (P_3, \dot{T}_{58}) data of table 1. Table 3 shows values of the term and of its component parts. The liquid volume term is

$$
f(V_L, P_3, T) = -\frac{1}{RT} \int_0^{P_3} V_L dP_3 \tag{5a}
$$

or

$$
f_x(V_L, P_3, T) = -0.005554T^3 - 0.000163T^4, \quad T < 2^{\circ}\text{K}.
$$
\n^(5b)

The coefficients of $(5b)$ were evaluated by fitting the next to last column of table 4, which is calculated from the smoothed [49] V_L data of Taylor and Kerr [50] and of Sherman and Edeskuty [51] and from the working equation (for ΔP_3).

TABLE 4. The liquid volume term, $f(V_L, P_3, T)$, in the equation
for ln P_3 is calculated by numerical integration at selected
temperatures, T, using smoothed values α , V_L , of liquid volume
data of Kerr and Taylor $\$

For P_3 the high-temperature working equation is used. Column 3 shows values d calculated from a power series in T^n fitted to column 2. The last two columns show the mismatch and its equivalent in millidegrees.

• Ref. (49)

 $\frac{b}{c}$ Ref. (50)
 $\frac{c}{d}$ Ref. (51)
 $\frac{d}{d}$ Eq. (5b)

On the right side of eq (1) we have those terms of the thermodynamic equation which cannot be adequately calculated from existing data on He³:

$$
a = L_o - \int_0^{T_m} C_{\rm sat} dT \tag{6}
$$

and

$$
b = S_L(T_m) \tag{7}
$$

where L_0 is the value of the latent heat of vaporization at absolute zero, and $S_L(T_m)$ is the liquid entropy at *Tm-*

Using the ordinary method of least squares analysis and weighting each data point equally in this step of the derivation of the 1962 He^3 scale, we find

$$
a/R = 2.09842 \pm 0.00070 \text{ for } T_m = 1.0^{\circ} \tag{8a}
$$

and

$$
b/R = 1.08360 \pm 0.00046
$$
 for $T_m = 1.0^\circ$. (8b)

By combining all the above functions we obtain an experimental thermodynamic equation scale (ETE) which is valid from $0.\overline{2}$ to 2.0 °K.

Because of the complexity of the ETE equation, and the fact that it is implicit in the pressure, iterative solutions were obtained with the aid of an electronic digital computer. A table in steps of 1 mdeg was prepared for comparison with the working equation scale.

4. The 1962 He^3 Full-Range Working Equation Scale

To obtain an expression valid over the full range from $0.2 \text{ }^{\circ}\text{K}$ to the critical point we now fit selected portions of eq (1) to a power series in $Tⁿ$, using as input pressures all of the (P_3, T_{58}) data of table 1. Using the method of multiple variable least squares analysis [47, 48] we fit

$$
\ln P_3 + f(T) + f_x(C_{\text{sat}}, T) + \frac{a}{RT} + \frac{b}{R} = \sum_{n=1}^{4} d_n T^n. \tag{9a}
$$

Note that in this fitting the vapor and liquid volume terms are expected to be fitted by the power series. To be acceptable it will therefore be necessary to demonstrate not only that the scale fits the input data but also that the scale of (9a) agrees with the ETE scale below the range of the input data, i.e., below $P_3=5.254$ mm.
The solution of the analysis, combining coefficients

of identical powers of *T,* is the full range working equation scale, FWE,

$$
\begin{aligned}\n\ln P_3 &= -2.49174/T + 4.80386 - 0.286001T \\
&\quad + 0.198608T^2 - 0.0502237T^3 + 0.00505486T^4 \\
&\quad + 2.24846 \ln T \qquad 0.2 &< T < 3.324 \,^{\circ}\text{K.} \quad \text{(9b)}\n\end{aligned}
$$

The upper limit is the critical point temperature consistent with the redetermination of the critical pressure found in Part I to be at 873 .0 mm Hg.

As shown in figure 1, a comparison of the two tables we generate from the ETE scale and the FWE scale shows excellent agreement: nowhere below $2 \degree K$ do the scales differ by more than 0.4 mdeg. $Equation (9b)$ is therefore in effect an experimental thermodynamic scale from 0.2 to 2.0 K and an empirical scale above $2 \text{ }^{\circ} \text{K}$.

A comparison of the fit of the 1962 He3 scale to the input (P_3, T_{38}) data is given in table 1 and figure 1. The standard deviation of the data from the scale is 0.25 mdeg.

The He⁴ lambda point occurs at $P_{4,\lambda} = 37.80 \text{ mm}$ ^o ^o ¹ ² Hg (corrected to 0° C and standard gravity). On the 1958 He⁴ scale this corresponds to 2.1720 °K. In Part I a direct interpolation procedure is described by which the value of *P3* which corresponds to $P_{4,\lambda}$ is 203.25 mm Hg. Hence $T_{62,\lambda}=2.1721$ °K, for *P 3=203.25* in good agreement with the value of this fixed point on the 1958 He⁴ scale of temperatures.

The most fundamental test which can be given to the 1962 He³ scale is its fit to He³ and He⁴ isotherm data, of which those of Keller [11, 12] are the most complete and accurate. Although Keller used a He⁴ thermometer in most of his isotherms, his observed P_4 's can be related to P_3 's by the direct interpolation equations described in Part 1. In Part III Keller's isotherms have been reanalyzed by the method of multiple variable least squares. Results

FIGURE 1. The solid line is the deviation of the 1962 He³ scale (i.e., the Full-range Working Equation, eq 9b) from the *Experimental Thermodynamic Equation (ETE)* scale in the range of validity of the ETE scale.

Plotted points are deviations of the 1962 He³ scale from the (P_2, P_i) input data: $T_{22}(P_3) - T_{35}(P_i)$. The + represents data at the critical point.

of the comparison with the 1962 He^3 and with the 1958 He4 scales are shown in figure 2. We note that the weighted average of $T_{N. 62}$ is 1.52 mdeg below T_{1so} , whereas for the same isotherms $T_{N, 58}$ averages 1.50 mdeg below T_{iso} . Since for these isotherms $T_{N,62}-T_{1\rm so}$ and $T_{N,58}-T_{1\rm so}$ have average standard deviations of 1.2 mdeg and 1.0 mdeg, respectively, we conclude that not much would be gained by basing a He³ scale more directly on the isotherm data. In other words, the 1958 He⁴ scale, which is

itself fundamentally based on these isotherm data, has evidently been an adequate interpolation parameter for fitting the He³ data to these isotherms.

Table 5 shows vapor pressures calculated from eq (9b) in steps of 10 mdeg. In table 6 we show dP/dT and $T_{82}-T_x$ in 0.1 deg steps, where T_{82} corresponds
to eq (9b) and T_x to the various He³ scales in use
in the past. We also have included on the far right the two He⁴ scale differences $(T_{L55}-T_{58})$ and $(T_{55E}-T_{58})$, as given in the NBS Monograph 10

"The 1958 He⁴ Scale of Temperatures" [2]. In a few recent publications an attempt has been made
to "correct" the T_L and T_F He³ scales by adding
these He⁴ scale "correction" terms. To convert these temperatures to the 1962 He^3 scale it is necessary to apply the sum of columns 3 and 6 (or 4 and 7) to the "corrected T_L " or "corrected T_E " scales.
Detailed (P,T) and (T,P) tables have been

published [8] by the Los Alamos Scientific Laboratory and in Part IV of this series.

In order to make the advantages of He³ as a vapor pressure thermometer more widely accessible, specially purified $He³$ is being made available for purchase³ for thermometry through the United States Atomic Energy Commission isotopes program.

 3 Information may be obtained from Gaseous I
sotopes Sales, Monsanto Research Corporation, Mound Laboratory, Miamisburg, Ohio.

Deviations of temperature scales from Keller's FIGURE 2. $(ref. [11, 12])$ isotherm temperatures.

 T_{iso} , as reanalyzed in Part III (ref. [7]). \bullet ($T_{\text{iso}}-T_{\text{iso}}$) for He' isotherms; \blacksquare ($T_{\text{iso}}-T_{\text{iso}}$) for He' isotherms; \bigcirc , \Box ($T_{\text{ss}}-T_{\text{iso}}$) for He' and He' isotherms, respectively.
To get T triangles respectively.

If it were possible the authors would acknowledge in detail the aid given by numerous cryogenists in the course of the development and evaluation of the 1962 He^3 scale of temperatures and the discussions concerning the scale by the Advisory Committee on Thermometry of the International Committee on Weights and Measures. We are especially indebted to F. G. Brickwedde, B. M. Abraham, H. van Dijk, **TABLE 6.** The temperature derivative dP_3/dT_{62} and deviations of various He^3 temperature scales from the present 1962 He³ scale

Columns 3, 4, and 5 give differences of the T_E , T_L , and T_K He³ scales respectively from the 1962 He⁴ scale in the form $(T_{82}-T_x)$ expressed in millidegrees. In Columns 6 and 7 are reproduced the deviations of th

 $\footnotesize\begin{array}{l} \text{a Ref. [9]}\\ \text{b Refs. [5, 41]}\\ \text{c Ref. [2]} \end{array}$

M. Durieux, R. I. Joseph, W. E. Keller, D. W. Osborne, and J. H. van Vleck. Since our correspondence ran to several dozen letters it would be impractical to specify all of the participants and to elaborate on their various contributions.

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