# Sound Velocity and Adiabatic Compressibility of Liquid Helium Three\*

HENRY L. LAQUER, STEPHEN G. SYDORIAK, AND THOMAS R. ROBERTS Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico (Received August 22, 1958)

The velocity of first sound at 5 Mc/sec was measured in liquid He<sup>3</sup> between 0.34 and 3.14°K. The results are fitted within 0.2% by the equation  $u = 183.9 - 5.98T^2 - 0.130T^3 - 0.00176T^8$  (m/sec). Initial pressure coefficients of the sound velocities and a number of derived thermodynamic quantities are also reported.

KNOWLEDGE of the velocity of sound in any liquid as a function of temperature and pressure permits the calculation of a number of thermodynamic properties. The velocity of first sound in liquid He<sup>4</sup> has been reported by several investigators.<sup>1-6</sup> In the present study, an ultrasonic pulse-echo method operating at a frequency of 5 Mc/sec has been used to measure the velocity of sound in liquid He<sup>3</sup>.

## APPARATUS

The experimental equipment includes mechanical, electronic, and cryogenic components, all of which were modified somewhat after completion of the major part of the work reported here, but before making the check measurements.

Figure 1 is a diagram of the sound cell and its integral helium-three Dewar which was machined from a solid copper block. The whole assembly is cooled by pumping on the He<sup>3</sup> bath in the annular space through a  $\frac{1}{8}$ -in. diameter Inconel tube which also serves as mechanical support. Sound velocity measurements are made on a separate sample of He<sup>3</sup> condensed in the central cylindrical volume of 1.6 cm<sup>3</sup> marked "X-liquid."

In the original cell, A, the transmitter (bottom) crystal of X-cut quartz was mounted rigidly to its backing cone with epoxy resin and was excited at a repetition rate of about seven times per second by a highly damped (2 cycle) "wave train" of 200-volt peakto-peak amplitude from a 0.1- $\mu$ sec pulse transformer. A thin-walled perforated brass cylinder of  $\frac{1}{2}$ -in. diameter and with ends machined parallel to within 0.0001 in. determined the length of the sound path to the receiving (top) crystal, which was held only by spring pressure. When this cell was disassembled it was ascertained that, in spite of repeated soldering of the cell, the crystals had remained intact and were in the same condition as when they were first installed.

The modified cell, B, used for the check measurements has a shorter but more carefully machined brass spacer,

- A64, 826 (1951).
  <sup>4</sup> C. E. Chase, Proc. Roy. Soc. (London) A220, 116 (1953).
  <sup>5</sup> K. R. Atkins and R. A. Stasior, Can. J. Phys. 31, 1156 (1953).

a pair of crystals held in place by six "C" clamps and no backing cones. Crystal excitation is the same as with the original cell.

The received signals are amplified directly in a broadband amplifier without the use of intermediate frequency stages or detection so that the detailed structure of each signal (Fig. 2), rather than its envelope, is displayed on the timing oscilloscope. A combination of precision step and continuously variable delay circuits permits delay measurements accurate to  $\pm 0.05 \ \mu \text{sec}$  for delay times from 0 to 1000  $\mu$ sec. The continuously variable delay is controlled by a ten-turn helical potentiometer and was originally designed to cover a range of 100 µsec. For the check measurements a Kelvin-Varley circuit of fixed resistors was added, allowing an optional reduction of this range to 10  $\mu$ sec. This improves the resolution by a factor of five. The electronic equipment is, of course, similar to the more conventional radar trainer sound velocity apparatus as used by Pellam and Squire<sup>2</sup> and Atkins and Chase<sup>3</sup>; but the use of separate crystals (as did Chase<sup>4</sup>) avoids some of the problems associated with nonoverloading or gated amplifiers, and the direct display of the 5-Mc/sec oscillations instead of the pulse envelope reduces the ambiguity in locating the "foot" of a signal.

With the two crystals, one has, in principle, the choice of obtaining sound velocities either by transmission or by reflection measurements. When using both crystals, signals corresponding to 1, 3, 5, and 7 transits through cell A could readily be observed and measured in He<sup>4</sup>, but only 1 and 3 transits were dis-



FIG. 1. Sound cell and Dewars.

<sup>\*</sup> Work done under the auspices of the U. S. Atomic Energy Commission.

 <sup>&</sup>lt;sup>1</sup> Findlay, Pitt, Grayson-Smith, and Wilhelm, Phys. Rev. 54, 506 (1938) and 56, 122 (1939).
 <sup>2</sup> J. R. Pellam and C. F. Squire, Phys. Rev. 72, 1245 (1947).
 <sup>3</sup> K. R. Atkins and C. E. Chase, Proc. Phys. Soc. (London)

<sup>&</sup>lt;sup>6</sup> A. van Itterbeek and G. Forrez, Physica 20, 133 (1954).



FIG. 2. Typical oscilloscope pattern. Sweep speed 0.4  $\mu$ sec/cm. Beginning of signal set to center line. Background (cross talk) from triggering circuits evident at left.

tinguishable in He<sup>3</sup>. When either crystal was used as an independent transducer (with the opposite crystal simply serving as reflector), echoes corresponding to 2 and 4 transits were apparent, but they were not sufficiently above noise to be used for precision velocity determinations. Also, there was no difference in gross signal strength or shape when either the tightly or the loosely bonded crystal served as the independent transducer. Finally it should be noted that the number of multiple-transit signals discernible in He<sup>4</sup> with either cell is less by almost an order of magnitude than the 40 or 50 echoes that can usually be seen in  $He^4$  with the same electronic apparatus and a sound velocity cell in which the liquid is not confined to a small capsule. From a measurement of the relative intensity of successive signals in He<sup>4</sup> near 2°K, one obtains an apparent pressure attenuation coefficient of  $\alpha = 0.4$  cm<sup>-1</sup> with cell A and  $\alpha = 0.23$  cm<sup>-1</sup> with cell B. This is, of course, much higher than has been reported for He<sup>4</sup> at that temperature and must be primarily an inherent property of each cell. It is not clear whether these high inherent cell  $\alpha$ 's can be ascribed entirely to a slight misalignment and frequency mismatch of the crystals or whether they are caused by edge or surface interference effects in the small cells. In addition to these effects there is a loss of acoustic signal strength upon transit from one medium to another. This acoustic impedance mismatch is greater between quartz and He<sup>3</sup> than with any other liquid and, as can readily be calculated from the data presented below, the increasing mismatch alone reduces the He<sup>3</sup> signal intensity at 3°K to 25% of its value at 1°K, which in turn is less than one-half of the He<sup>4</sup> intensity at about 2°K.

# THERMOMETRY

The temperature of the liquid under investigation is inferred from the vapor pressure of He<sup>3</sup> in the thermometer well shown in Fig. 1, using the  $T_E$  scale of Sydoriak and Roberts.<sup>7</sup> Depending on the temperature range, either a mercury manometer, an oil manometer, or a calibrated McLeod gauge is used to measure the vapor pressure. As discussed previously,<sup>8</sup> thermomolecular pressure ratio corrections have to be applied when the measured vapor pressures are less than about  $300 \mu$  of Hg. In our apparatus, these corrections amount to about 48% of the observed pressure or 25 millidegrees at  $0.34^{\circ}$ K. The uncertainty in the temperatures thus obtained is of the order of a millidegree. To this must be added the uncertainty in the temperature scale itself and the possibility of thermal gradients at the various interfaces, which are difficult to estimate. However, since the sound velocity is not a strong function of the temperature, thermometry errors are probably negligible in this work.

## SAMPLE ANALYSIS

Mass-spectrometric analyses of the purity of the available He<sup>3</sup> gas were performed a number of times. However, only after the check run was an analysis obtained on the liquid actually present in the sound cell. It was found to contain  $0.010\pm0.001\%$  of He<sup>4</sup>, an amount which should affect the measured velocities by less than 0.005%. Since the check measurements fell within the scatter of the earlier results, it seems safe to assume that in all measurements the liquid He<sup>3</sup> had a purity of at least 99.9\%.

# ERRORS

Table I gives our estimates of other errors which might affect the reported velocities. In cell A the distance between the crystals could be in error by  $\pm 0.0002$ in. (0.05%) whereas the length of cell B is known to  $\pm 0.00005$  in. (0.017%). The contraction of the brass spacers might well differ by 5% from calculated values<sup>9</sup> and this would make the length of the spacers uncertain to 0.02%. There is an additional delay (or path length) between the initial pulse and the first transmitted signal due to the uncertainty as to where in the crystal the electrical impulse traveling with the speed of light is converted into a mechanical pulse traveling with the speed of sound and vice versa. For cell A the magnitude of this correction has been determined empirically from a comparison of the times between higher order transits and represents an average value for measurements in He<sup>3</sup> and He<sup>4</sup>. For a transit time of  $60 \,\mu$ sec this correction,

TABLE I. Summary of errors.

Source	Cell A Magnitude	% in u	Cell <i>B</i> Magnitude	% in u	
Length of cell at 20°C	1.0447 ±0.0005 cm	±0.05	0.75710±0.00013 cm	±0.017	
Contraction to 0°K	$(3.8\pm0.2) imes10^{-3}$	$\pm 0.02$	$(3.8\pm0.2) imes10^{-3}$	$\pm 0.02$	
first transit	$-0.18\pm\!0.06~\mu\mathrm{sec}$	$\pm 0.10$	$-0.09\pm0.09\ \mu\mathrm{sec}$	$\pm 0.21$	
interval	$\pm 0.07 \ \mu \text{sec}$	$\pm 0.12$	$\pm 0.07 \ \mu \text{sec}$	$\pm 0.16$	
Compounded error (rms)		±0.17	••••	$\pm 0.27$	

<sup>9</sup> H. L. Johnston, Ohio State University results (unpublished); D. B. Fraser and A. C. Hollis Hallett, Proceedings of the Ninth International Congress of Refrigeration 1, 064 (1955).

<sup>&</sup>lt;sup>7</sup> S. G. Sydoriak and T. R. Roberts, Phys. Rev. **106**, 175 (1957). <sup>8</sup> T. R. Roberts and S. G. Sydoriak, Phys. Rev. **102**, 304 (1956).

amounting to  $-0.18\pm0.06 \,\mu\text{sec}$ , contributes a 0.1%uncertainty. A less extensive determination of this empirical correction for cell *B* gave a value of -0.09 $\pm0.09 \,\mu\text{sec}$  corresponding to an uncertainty of 0.21%for the shorter transit times encountered with this cell.

Under ideal conditions, the continuously variable 100- $\mu$ sec delay can be set reproducibly to  $\pm 0.02 \ \mu$ sec. However, the linearity of the helical potentiometer is only  $\pm 0.05\%$  ( $\pm 0.05 \ \mu$ sec) so that in spite of the fact that the dial is calibrated every 10  $\mu$ sec, individual time intervals have a probable error of  $\pm 0.07 \ \mu$ sec which at 1°K is 0.12% for cell A and 0.16% for cell B. Use of the expanded 10- $\mu$ sec range gives  $\pm 0.01 \ \mu$ sec reproducibility and correspondingly increased precision, but the limitations of our calibration scheme preclude an increase in the absolute accuracy. Occasionally, the arrival time of a weak pulse may be misread by as much as a

TABLE II. He<sup>4</sup> Sound velocities (m/sec).<sup>a</sup>

$T_E(^{\circ}K)$	U m	$T_E(^{\circ}K)$	$u_m$
2.557	221.45	1.779	231.4
$2.173_{5}$	$218.1_{5}$	1.465	235.9
$2.179_{5}$	$219.0_{7}$	1.235	236.8
2.239	220.5	1.066	237.3
		0.934	237.7
2.309	$221.2_{6}$	0.918	237.7
2.154	218.5		

<sup>\*</sup> Runs made on different days are separated by dashed horizontal lines.

whole cycle  $(0.2 \,\mu\text{sec})$ . It is thus evident that the major uncertainties stem from the time measurements. Combination of all the errors listed in Table I produces an over-all root-mean-square error of 0.17% for most of the data and of 0.27% for the check measurements. These numbers apply to all velocity measurements made below 2°K with the liquid at its saturation vapor pressure. Because of the increase of attenuation and impedance mismatch with increasing temperature, most measurements above 2°K were made at a number of

TABLE III. He<sup>3</sup> sound velocities (m/sec).<sup>a</sup>  $\Delta u \equiv u_m - u_c$ , where  $u_m$  is the measured sound velocity and  $u_c$  is that calculated from Eq. (2).

				and all a second and a second and a second a se	
$T_E(^{\circ}K)$	U m	Δu	$T_E(^{\circ}\mathrm{K})$	Um	Δu
1.075	176.96	+0.14	1.776	163.84	-0.30
0.927	$178.8_{5}$	+0.19	1.467	$170.3_{1}$	-0.27
0.817	179.94	+0.10	1.227	$174.7_{1}$	+0.06
0.738	180.5	-0.03	0.964	178.67	+0.45
0.658	$180.9_{1}$	-0.36	0.818	180.34	+0.51
0.587	181.8	+0.08			
0.529	182.3	+0.09	2.244	151.16	-0.03
0.485	$182.3_{6}$	-0.12	2.374	146.81	+0.13
0.405	$182.6_{8}$	-0.23	2.568	$138.4 \pm 0.3$	-0.53
0.391	183.01	+0.03	3.136	$104.6 \pm 0.7$	-0.02
1.079	176.61	-0.16	3.001	$114.7 \pm 0.5$	-0.25
1.085	$177.2_{5}$	+0.56	2.780	$128.8 \pm 0.2$	+0.19
1.156	175.6	-0.01	1.948	159.7	-0.15
			1.601	167.7.	-0.18
2.576	$139.2 \pm 0.3$	+0.62	1.396	171.8	-0.07
2.269	$150.4 \pm 0.3$	+0.04	0.380	182.65	-0.38
2.128	$154.6 \pm 0.2$	-0.23	0.374	182.9	-0.15
				1	51-0

\* Runs made on different days are separated by dashed horizontal lines.



FIG. 3. Sound velocities in He<sup>3</sup> and He<sup>4</sup> (● this study, ● Van Itterbeek and Forrez,<sup>6</sup> □ Atkins and Stasior<sup>5</sup>).

pressures up to 200 mm of Hg above saturation pressure and then extrapolated to saturation pressure. These data are afflicted with errors increasing with the pressure coefficient of the velocity and reaching a magnitude of  $\pm 0.7\%$  at the highest temperature at which measurements were made (3.14°K).

#### SOUND VELOCITIES

The experimental results obtained with cell A on He<sup>4</sup> and He<sup>3</sup> are reported in Tables II and III. The same data are plotted in Fig. 3. It is apparent that our values for He<sup>4</sup> are in excellent agreement ( $\pm 0.2\%$ ) with the work of Van Itterbeek and Forrez<sup>6</sup> who used an interferometer technique. There is also satisfactory agreement with the data reported by Atkins and Stasior.<sup>5</sup> Similar agreement is obtained with the work of Atkins and Chase<sup>3</sup> (not shown in Fig. 3) provided velocities read from their graphs are increased by 0.8% as suggested by Chase.<sup>4</sup>

Smooth graphical extrapolation of the He<sup>3</sup> data with the derivative du/dT going to zero at 0°K yields  $u_0=183.4$  m/sec. Least-squares calculations, using 16 power series of the form

$$u = u_0 + \sum a_i T^i, \tag{1}$$

indicate that the measured velocities,  $u_m$ , can be fitted satisfactorily by the equation

$$u_c = 183.9 - 5.98T^2 - 0.130T^3 - 0.00176T^8 (m/sec).$$
 (2)

The  $\Delta u = u_m - u_c$  column of Table III shows that the rms deviation of the fit is  $\pm 0.29$  m/sec or 0.17% which matches the compounded error from Table I.

$T_E$	U m	$\Delta u$	$T_E$	U m	$\Delta u$
$\begin{array}{c} 0.337\\ 0.348\\ 0.359\\ 0.371\\ 0.394\\ 0.424\\ 0.440\\ 0.462\\ 0.488\\ 0.516\\ 0.548\\ \end{array}$	$\begin{array}{c} 183.07_{2} \\ 183.04_{5} \\ 183.02_{7} \\ 182.97_{0} \\ 182.87_{7} \\ 182.67_{7} \\ 182.65_{0} \\ 182.49_{8} \\ 182.33_{7} \\ 182.22_{5} \end{array}$	$\begin{array}{r} -0.144 \\ -0.125 \\ -0.096 \\ -0.0087 \\ -0.025 \\ -0.054 \\ +0.039 \\ +0.037 \\ +0.047 \\ +0.142 \end{array}$	$\begin{array}{c} 0.587\\ 0.617\\ 0.652\\ 0.735\\ 0.735\\ 0.780\\ 0.831\\ 0.889\\ 0.940\\ 1.018\end{array}$	$\begin{array}{c} 181.92_{4}\\ 181.81_{0}\\ 181.49_{9}\\ 181.12_{9}\\ 180.73_{4}\\ 180.37_{0}\\ 179.76_{4}\\ 179.12_{8}\\ 178.35_{5}\\ 177.42_{0}\\ \end{array}$	$\begin{array}{c} +0.111\\ +0.217\\ +0.177\\ +0.136\\ +0.116\\ +0.170\\ +0.069\\ +0.046\\ -0.152\\ -0.144\end{array}$

TABLE IV. Check run (99.990% He<sup>3</sup>, Cell B).

A check run was made with cell B, using the variable time delay with improved resolution and a carbon resistance thermometer for interpolation of the temperatures. Modifications of the tubing leading to the cell allowed measurements at a slightly lower temperature (0.337°K) than had been attained in the earlier work. The check run results are given in Table IV. Graphical extrapolation again leads to a value of  $u_0=183.4$  m/sec and the new data are also closely described by Eq. (2) although they were not used in arriving at the constants entering this equation.

Table IV provides a verification of the earlier measurements which is desirable in view of the fact that when our work was first reported,<sup>10</sup> similar measurements made at another laboratory<sup>11</sup> but at higher frequencies and with a smaller sample of liquid gave sound velocities lower by about 3%. In making the check



FIG. 4. Initial variation of sound velocity with pressure.

measurements, it was also determined that at  $0.85^{\circ}$ K the velocity is unchanged when 3- and 10-db attenuators are inserted into the line from the pulse transformer to the crystal. Thus it appears that our measured velocities are independent of the magnitude of the voltage pulse, i.e., they are true sound velocities and not shock velocities.

# PRESSURE COEFFICIENTS

As already mentioned, measurements at temperatures above 2°K were made at pressures in excess of saturation pressure and extrapolated linearly to saturation. The slopes of these extrapolations, incidentally, furnish values shown in Fig. 4 for the initial pressure coefficients of the sound velocities. A more detailed study of the pressure variation of the sound velocity was made with cell *B* at  $1.525\pm0.005$ °K (Table V). These data show that the slope du/dP decreases rapidly from a value of  $0.041\pm0.001$  m sec<sup>-1</sup> (mm of Hg)<sup>-1</sup> at saturation (54 mm of Hg), shown as a solid circle in Fig. 4, to 0.030 at 200 mm and approaches a more or less constant value of 0.0245 at 500 mm.

TABLE V. Variation of velocity with pressure at 1.525°K.

Pressure	Velocity	Pressure	Velocity
(mm of Hg)	(m/sec)	(mm of Hg)	(m/sec)
817	189.02	301	176.91
810	189.64	316	177.29
700	186.59	196	174.02
697	186.40	99.1	170.82
600.5	184.80	54	169.02
506	182.12	55	169.14
396	179.44	53.7	169.06

#### DERIVED QUANTITIES AND COMPARISON WITH He<sup>4</sup>

The measurements just recorded allow the calculation of a number of other thermodynamic quantities. Table VI summarizes some of these calculations and also lists corresponding data for He<sup>4</sup>. Any bracketed number represents an extrapolation beyond the experimental range and should obviously be viewed with caution, particularly if there is the possibility of a density anomaly.<sup>12</sup> The columns of Table VI represent the following quantities:

(1) Temperatures, T.

(2) Sound velocities, u, calculated from Eq. (2), except for the 0° value which was obtained by the graphical extrapolation previously discussed. He<sup>4</sup> data were obtained from a large plot of Fig. 3 including our own measurements (Table II) and literature data.<sup>3-6</sup>

(3) Initial values of the pressure derivative du/dP from Fig. 4 for He<sup>3</sup> and average values  $\Delta u/\Delta P$  between saturation and 2.5 atmos from Atkins and Stasior<sup>5</sup> for He<sup>4</sup>.

<sup>&</sup>lt;sup>10</sup> Laquer, Sydoriak, and Roberts, Proceedings of the Fifth International Conference on Low-Temperature Physics and Chemistry, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 98. <sup>11</sup> H. Flicker and K. R. Atkins, Proceedings of the Fifth Inter-

<sup>&</sup>lt;sup>11</sup> H. Flicker and K. R. Atkins, *Proceedings of the Fifth Inter*national Conference on Low-Temperature Physics and Chemistry, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 95.

<sup>&</sup>lt;sup>12</sup> (a) L. Goldstein, Phys. Rev. **102**, 1205 (1956), (b) D. M. Lee and H. A. Fairbank, *Proceedings of the Fifth International Conference on Low Temperature Physics and Chemistry*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 90.

Т (°К)	$u \times 10^{-4}$ (cm sec <sup>-1</sup> )	$(du/dP) \times 10^{3}$ (cm <sup>3</sup> dyne <sup>-1</sup> sec <sup>-1</sup> )	V (cm³ mole <sup>-1</sup> )	α′ ×10² (°K⁻¹)	( <i>dP/dT</i> ) ×10 <sup>-5</sup> (dyne cm <sup>-2</sup> °K <sup>-1</sup> )	χs×108 (cm² c	χτ×10 <sup>8</sup> lyne <sup>−1</sup> )	C' (jou	CP le mole <sup>-1</sup>	<i>Cv</i> °K⁻¹)	γ
He <sup>3</sup>	/	1									
0 0.5 1.0 1.5 2.0 2.5 3.0	$\begin{array}{c} (1.834 \pm 0.005) \\ 1.824 \pm 0.003 \\ 1.778 \pm 0.003 \\ 1.700 \pm 0.003 \\ 1.585 \pm 0.003 \\ 1.418 \pm 0.004 \\ 1.150 \pm 0.005 \\ (0.0005 \pm 0.015) \end{array}$	$ \begin{array}{c}\\ 3.0\pm0.2\\ 3.2\pm0.3\\ 4.7\pm0.5\\ 9.8\pm0.8\\ (15) \end{array} $	$(36.63) \\ (36.65) \\ 36.873 \\ 37.459 \\ 38.708 \\ 41.202 \\ 46.775 \\ 500$	$\begin{array}{c} 0 \\ (0.33) \\ 1.95 \\ 4.56 \\ 8.94 \\ 17.0_1 \\ 38.2_5 \\ \end{array}$	0 0.0277 0.547 1.79 3.67 6.16 9.25	$\begin{array}{c} (3.610) \\ (3.652) \\ 3.868 \\ 4.300 \\ 5.110 \\ 6.795 \\ 11.73 \\ 17.45 \end{array}$	(3.610) (3.653) 3.908 4.582 6.333 11.86 	$0 \\ 3.260 \\ 4.294 \\ 5.717 \\ 7.713 \\ 10.483 \\ \cdots$	$0 \\ 3.260 \\ 4.298 \\ 5.771 \\ 8.033 \\ 12.03 \\ \cdots$	0 3.259 4.254 5.416 6.482 6.891 	$1 \\ 1.0002 \\ 1.0104 \\ 1.0656 \\ 1.239 \\ 1.745 \\ \dots$
3.2 He4	$(0.990\pm0.013)$	$(15.0\pm1.5)$	51.590	05.23	10.04	17.45	•••	•••	•••	•••	•••
0 1 2 3 4	$\begin{array}{c} (2.39 \pm 0.02) \\ 2.375 \pm 0.005 \\ 2.267 \pm 0.005 \\ 2.176 \pm 0.005 \\ 1.90 \pm 0.01 \end{array}$	(0.8) 0.8 1.0 1.5	(27.51) 27.516 27.418 28.360 31.136	$\begin{array}{r} 0 \\ + \ 0.0378 \\ - \ 1.008 \\ + \ 6.503 \\ + 12.55 \end{array}$	0 0.0153 0.929 3.63 8.29	(1.203) 1.219 1.333 1.496 2.155	(1.203) 1.219 1.335 1.929 3.921	0 0.417 20.73 9.97 15.97	0 0.417 20.73 10.19 17.60	0 0.417 20.70 7.902 9.672	1 1.00008 1.00155 1.289 1.820

TABLE VI. Derived quantities for saturated liquid. The symbols are defined in the text.

(4) Molar volumes, V, taken from the work of Kerr<sup>13,14</sup> but with a small correction for the change from the 48 to the 55E temperature scale.

(5) Thermal expansion coefficients,  $\alpha'$ , of the liquid along the saturation line taken from Kerr's<sup>13</sup> results for He<sup>3</sup> and from Atkins and Edwards<sup>15</sup> and Kerr<sup>14</sup> for He<sup>4</sup>. (6) Slopes of the vapor pressure curve of the liquid, dP/dT, from the  $T_E$  scale of Sydoriak and Roberts<sup>7</sup> for

He<sup>3</sup> and from Clement's  $T_{55E}^{16}$  for He<sup>4</sup>.

(7) Adiabatic compressibilities

$$\chi_S = V/Mu^2, \tag{3}$$

where  $M_3 = 3.0162$  g/mole and  $M_4 = 4.0028$  g/mole. (8) Isothermal compressibilities

$$\chi_T = \gamma \chi_S, \tag{4}$$

where  $\gamma = C_P / C_V$  (see below).

(9) Heat capacities, C', of the liquid along the saturation line from the equation of Roberts and Sydoriak<sup>17</sup> for He<sup>3</sup> and from Hill and Lounasmaa<sup>18</sup> and Kramers, Wasscher and Gorter<sup>19</sup> for He<sup>4</sup>.

(10) Heat capacities at constant pressure<sup>20</sup>

$$C_P = C' / [1 - A(1 + B)],$$
 (5)

where  $A = \alpha' V T (dP/dT)/C'$ , and  $B = V (dP/dT) M/u^2 \alpha'$ , (11) Heat capacities at constant volume<sup>20</sup>

$$C_V = C' / [1 + A(1 + 1/B)].$$
(6)

(12) The ratio of the heat capacities

$$\gamma = C_P / C_V = [1 + A(1 + 1/B)] / [1 - A(1 + B)]. \quad (7)$$

The extrapolated value of the adiabatic compressibility of He<sup>3</sup> is 3.66% per atmosphere at 0°K, which

- <sup>13</sup> E. C. Kerr, Phys. Rev. 96, 551 (1954).
   <sup>14</sup> E. C. Kerr, J. Chem. Phys. 26, 511 (1957).
   <sup>15</sup> K. R. Atkins and M. H. Edwards, Phys. Rev. 97, 1429 (1955).
- <sup>16</sup> W. E. Keller, Nature 178, 883 (1956).

<sup>17</sup> T. R. Roberts and S. G. Sydoriak, Phys. Rev. 98, 1672 (1955).

- <sup>18</sup> R. W. Hill and O. V. Lounasmaa, Phil. Mag. 2, 143 (1957).
   <sup>19</sup> Kramers, Wasscher, and Gorter, Physica 18, 329 (1952).
- <sup>20</sup> L. Goldstein (to be published).

should be compared with Brueckner and Gammel's<sup>21</sup> calculated value of 5.3% per atmosphere. We can also compare our value of 4.05% per atmosphere at  $1.2^{\circ}$ K with the isothermal compressibility of 3% per atmosphere from susceptibility measurements reported by Walters and Fairbank<sup>22</sup> as an average over a pressure range of several atmospheres. Within their implicit accuracy, their estimate appears quite reasonable and their calculated sound velocity of 195 m/sec may well apply for He<sup>3</sup> at a pressure of about 3 atmos. A similar comparison can be made with Peshkov's<sup>23</sup> optical compressibility determination. Initial isothermal compressibilities, estimated from the initial slopes of his  $\Delta \rho$  vs  $\Delta P$  plots, fall within  $\pm 10\%$  of a smooth curve drawn through our calculated  $\chi_T$  values from Table VI.



FIG. 5. Comparison of the isotopes.

<sup>21</sup> K. A. Brueckner and J. L. Gammel, Phys. Rev. 109, 1040

- (1958). <sup>22</sup> G. K. Walters and W. M. Fairbank, Phys. Rev. **103**, 263
- (1956). <sup>23</sup> V. P. Peshkov, J. Exptl. Theoret. Phys. (U.S.S.R.) **33**, 833 (1957) [translation: Soviet Physics JETP **6**, 645 (1958)].

The decrease of the compressibility with increasing pressure is evident from the curvature of his data at all temperatures except the lowest. His calculated sound velocity of 170 m/sec at 1.6°K compares closely with our measured value of 168 m/sec. However, his calculated velocity of 80 m/sec at 3.0°K is much too low, possibly from an underestimation of the magnitude of  $\gamma$ .

Since most properties of He<sup>3</sup> vary smoothly and monotonically with temperature, they provide a convenient frame of reference for the properties of He<sup>4</sup>. Figure 5 is a plot of the ratio of some properties in He<sup>4</sup> to those in He<sup>3</sup> taken at equal values of the reduced temperature  $\theta = T/T_{crit}$ . Although the  $\lambda$ -phenomenon is clearly apparent, it is interesting to note that both the ratios of the sound velocities and of the particle densities of the isotopes are fairly constant and lie within 5% of the ratio of their atomic weights over the entire range of available data.

We also note that the values of  $\gamma$  for the two isotopes are essentially the same for corresponding values of  $\theta$ . From these statements and Eq. (3) it follows that both adiabatic and isothermal compressibilities should be in the inverse ratio of the fourth power of the atomic weights or that He<sup>3</sup> is about 3.1 times as compressible as He<sup>4</sup>. No such simple relationships seem to hold for the various heat capacities.

# ACKNOWLEDGMENTS

The electronic apparatus was developed for one of us (H.L.L.) about 1951 by C. W. Johnstone, formerly of this laboratory, and was further improved through suggestions from Dr. W. L. Briscoe. We are indebted to Dr. T. L. Jordan for help and advice with the leastsquares fits, and we are grateful to Dr. Louis Goldstein for many enlightening discussions and the compact form of the thermodynamic relations used in calculating the heat capacities.

PHVSICAL REVIEW

#### VOLUME 113. NUMBER 2

**JANUARY 15, 1959** 

# Stored Energy Release in Copper Following Electron Irradiation below 20°K\*

C. J. MEECHAN AND A. SOSIN

Atomics International, Division of North American Aviation, Incorporated, Canoga Park, California (Received September 25, 1958)

The stored energy release in copper has been measured in the temperature range 20°-60°K following irradiation with 1.2-Mev electrons. A differential temperature measurement was made between an irradiated specimen and an unirradiated standard. The specimens were immersed in liquid helium during irradiation; subsequent heating of the specimen was carried out in vacuum. A value of the total energy release of  $2.5 \times 10^{-2}$ cal/g was observed for an integrated flux of  $9 \times 10^{17}$  e/cm<sup>2</sup>. The stored energy-resistivity ratio obtained is  $(5.4\pm0.8)$  cal/g per micro-ohm-cm. The energy associated with a Frenkel pair is calculated to be  $(5.4\pm0.8)$ ev for a value of 3.6 micro-ohm-cm per atomic percent Frenkel defects.

#### I. INTRODUCTION

FEW years ago, Cooper, Koehler, and Marx<sup>1</sup> A measured the electrical resistivity recovery occurring in the noble metals near 30°K following deuteron bombardment at 12°K. This experiment marked the first successful attempt to study recovery of radiation damage in metals in this temperature range, now called Stage I. In a relatively short time following this important beginning, many experiments were reported involving several different physical property changes which occur in this temperature region following irradiation with deuterons,<sup>2,3</sup> neutrons,<sup>4-6</sup> and electrons,<sup>7,8</sup> and also damage by cold work.<sup>9</sup>

In order to accurately describe the Stage I recovery phenomena, it seemed necessary to measure the energy associated with this recovery following various types of irradiation. This problem was first attacked by Blewitt, Holmes, Coltman, and Noggle<sup>5</sup> on neutronirradiated copper. The first results reported on the energy release following neutron irradiation were somewhat smaller than expected if one assumed that all of the Stage I recovery was due to interstitial-vacancy recombination. Since the fraction of the damage produced by neutron irradiation which is attributable to interstitials and vacancies is not well determined, it was deemed necessary to perform the stored energy measurement on an electron-irradiated specimen. It is generally agreed that electrons with energies near 1 Mev are energetically capable of producing only point

<sup>\*</sup> This work was performed under contract to the U.S. Atomic Energy Commission.

<sup>&</sup>lt;sup>a</sup> Cooper, Koehler, and Marx, Phys. Rev. 97, 599 (1955). <sup>a</sup> R. O. Simmons and R. W. Balluffi, Phys. Rev. 109, 1142 (1958).

<sup>&</sup>lt;sup>3</sup> R. Vook and C. Wert, Phys. Rev. 109, 1529 (1958).

<sup>&</sup>lt;sup>4</sup> Blewitt, Coltman, Klabunde, and Noggle, J. Appl. Phys. 28, 639 (1957)

<sup>&</sup>lt;sup>5</sup> Blewitt, Holmes, Coltman, and Noggle, Oak Ridge National

Laboratory Progress Report ORNL-2188, August, 1956 (unpublished).



FIG. 2. Typical oscilloscope pattern. Sweep speed 0.4  $\mu$ sec/cm. Beginning of signal set to center line. Background (cross talk) from triggering circuits evident at left.