

**Informal Report** 

H. 197.

UC-34 Issued: March 1978

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# Thermodynamic Properties of Fluid <u>n</u>-D<sub>2</sub> in the 75 to 300 K and 2- to 20-kbar Range

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UNITED STATES DEPARTMENT OF ENERGY CONTRACT W-7403-ENG. 36

# THERMODYNAMIC PROPERTIES OF FLUID <u>n</u>-D. IN THE 75 TO 300 K AND 2 TO 20-kbar RANGE

by

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#### ABSTRACT

The hydrogen isotope deuterium is an important material for use in various energy technologies. This rep. it is a summary of new pressure, volume, temperature, and sound velocity measurements of fluid  $\mathbf{n}$ - $\mathbf{D}_{t}$  in the 75 to 300 K and 2- to 20-kbar range. An equation of state (EOS) was fit to these data. The thermodynamic quantities, volume V, sound velocity  $\mathbf{v}_{a}$ , thermal expansivity  $\alpha_{P}$ , heat capacity at constant pressure  $C_{P}$ , isothermal compressibility  $\chi_{T}$ , and molar entropy S, are given at 25 K and 0.5-kbar increments over the range of measurements. Computer-drawn graphs of the isothermal pressure variation of these quantities are shown. Characteristics of the EOS at high temperature and pressure are determined and compared with theoretical and phenomenological equations of state.

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#### I. INTRODUCTION

The hydrogen isotopes are important materials for use in various energy technologies. To produce the high densities and temperatures in deuterium that are needed for controlled nuclear fusion, a basic understanding of the properties of deuterium is required. This knowledge is especially important in laser-fusion schemes because lasers of only marginal power are now available to drive the compression.

Deuterium is expected to become a metal at pressures in the megador region and possibly remain metallic when the pressure is reduced. Generation of metal deuterium is a challenge to our present technology, and a clear demonstration of its existence would contribute to our basic understanding of matter. The possibilities for use of the metal as a high-density fuel and perhaps as a high-temperature superconductor have kindled new interest in studies of this material.

There are fewer measurements available for deuterium than for hydrogen; in particular, the equation of state (EOS), the relationship between pressure, P, volume, V, and absolute temperature, T, is known in only a limited pressure and temperature region. Solid deuterium measurements at low temperature have been made up to 25 kbar,<sup>1,2</sup> but most studies of the gas have been restricted to <3 kbar. Measurements by Michels et al.<sup>2</sup> near room temperature extend to 2.2 kbar. An important reason for this experimental limit is the deleterious effect that deuterium has on many high-strength steels, especially at higher temperatures.

At the Los Alamos Scientific Laboratory we have developed a facility to make P-V-T studies at pressures >3 kbar on various light molecules including the hydrogen isotopes. An apparatus and technique were used for studies up to 21 kbar in the 25 to 320 K range. Earlier measurements of argon,<sup>4</sup> nitrogen,<sup>8,6</sup> and hydrogen<sup>1,8</sup> have been reported. We recently completed<sup>9</sup> P-V-T and simultaneous sound velocity, v<sub>0</sub>, measurements on fluid normal deuterium in the 75 to 300 K and 2- to 20-kbar range. In this report we will review these measurements and the EOS development based on our use of P-V-T and v<sub>0</sub> data in a double-process least-squares fitting program that used all 1340 sets of data points. From the EOS other thermodynamic properties are derived and are presented in more detail here than was possible in Ref. 9. Although comparisons with some theoretical calculations were made in Ref. 9, additional comparisons are given in this report.

#### **II. EXPERIMENTAL**

We used a thermostated piston-cylinder apparatus, described in Ref. 4, with modifications described in Ref. 9. The ends of the supported tungsten-carbide cylinder were closed with Bridgman unsupported-area seals. Volumes were determined from the piston displacement, as measured with an external dial gauge. Pressures were calculated from the ram-to-cell area ratio and from the force generated in the hydraulic press with a rotating-piston dead-weight tester. Temperatures were computed from two external thermocouples previously calibrated against an *in situ* thermocouple at lower pressures. The unsupported area of the lower fixed seal was used to mount an ultrasonic transducer in the low-pressure environment. Pulses with either 30- or 10-MHz modulation were propagated through the plug, which served as a buffer rod, into the sample in a fixed-path cell, and were reflected out again. Velocities were computed from the pulse propagation times.

Errors in these measurements are given in Ref. 9. The corrected pressures are considered accurate to a few tenths of 1% at the lowest pressures and to about 0.5% at the highest pressures. Errors in volume average about 0.4%, becoming larger at pressures and temperatures farther from the normalizing point of 2 kbar and 295 K. Temperatures at ambient and 75 K are known to within  $\pm 0.02$  K, but at intermediate temperatures the accuracy is estimated to be  $\pm 0.5$  K. Sound velocities were found to scatter from a power-law fit by  $\pm 0.5$ %, although uncertainties in the timing accuracy and cell length measurement were calculated to be  $\pm 0.4$ %.

We obtained a total of 1340 data sets along 33 isotherms in fluid  $\underline{n}$ -D<sub>2</sub> in the 75 to 300 K and 2to 20-kbar range. These data are available in Ref. 10 for use as a data base in further EOS development. Other separate sets of V(P,T) and v<sub>s</sub>(P,T) were obtained and used as checks and to determine a power-law fit with a least-squares method. To fit these data, we used

(1)

 $v_0 \simeq AP^n$ ,

where A and n are constants. A similar equation is found to represent the V vs P data, where A and n are different constants and n is negative. The magnitude of n is  $\approx 0.34$  in each case so that  $v_s \cdot V \approx \text{constant}$ , as was determined for hydrogen.<sup>6</sup> Figures 1-18 show the quality of these power-law fits for six isotherms over the full temperature range. Percentage deviations of the measured sound velocities from the fitted Eq. (1) are shown. The power-law dependence for volumes is also shown for the six isotherms. The deviations of sound velocity from a fit to Eq. (1) are plotted in Fig. 19 for nine isotherms in the 294.5 to 296.1 K range.

#### **III. EQUATION-OF-STATE DEVELOPMENT**

We followed the procedure discussed in Refs. 7 and 8 to obtain a Benedict<sup>11</sup> type of EOS of the form,

$$V = (A + BT + CT^{-1/4})P^{-1/4} + (D + ET)P^{-2/4} + (F + GT + HT^{-1/4} + JT^{-1})P^{-1},$$
(2)

where A-H and J are constants to be determined from the double-process least-squares fit.<sup>9</sup> Knowledge of the heat capacity C<sub>P</sub> over the full temperature range at our normalization pressure of 2 kbar is required to relate the sound velocity data to the EOS of Eq. (2). For deuterium there are few data available, so we have used derived values of C<sub>P</sub> from Michels et al.,<sup>8</sup> as extrapolated to 2 kbar. A polynomial equation is used to describe these values.

$$C_{P_0}(T) = 0.13637T - 5.3300T^{1/2} + 90.56 - 85.579T^{-1/2} - 956.11T^{-1}$$
. (J/mole-K). (3)

These results are used in the relation for the heat capacity,

$$C_{p} = C_{p_{0}}(T) - T \int_{P_{0}}^{P} \left(\frac{\partial^{2} V}{\partial T^{2}}\right)_{p} dP \quad , \qquad (4)$$

and, in turn, in the relation for the sound velocity,

$$\frac{1}{v_{s}} = \frac{M^{1/2}}{V} \left\{ -\left(\frac{\partial V}{\partial P}\right)_{T} - \frac{T\left[\left(\frac{\partial V}{\partial T}\right)_{p}\right]^{2}}{C_{p}} \right\}^{1/2}$$
(5)

The experimental data sets of 1340 points were augmented by 49 data sets, obtained by slight extrapolations to 2 and 20 kbar at some of the temperatures, and by 15 sets of V and  $v_{*}$  points from Michels et al.<sup>4</sup> to fill in the lower pressure region. The resulting fit of the total 1404 data sets is shown in Table I. Minor changes in the extrapolation for  $C_{P_0}(T)$  change the sign of some of the constants and although the quality of the fit is affected only slightly, the extrapolations to high pressure and temperature show significant differences.

#### **TABLE I**

# **CONSTANTS OF EQ. (2) DETERMINED BY A DOUBLE-PROCESS** LEAST-SQUARES FIT TO THE 1404 DATA SETS

A = 35.2826	F = 13.6504
B = 0.000947034	G = 0.0695631
C = 3.28433	H = -158.294
<b>D</b> = -25.0897	J = 720.003
E = 0.00639169	

The values in Table I are determined with a weighting on the points that emphasizes the higher pressures. During the fitting process, we compared the computed values of the heat capacity at constant volume from Eq. (2) with derived values of Michele et al.<sup>6</sup> and adjusted Eq. (3) for  $C_{P_0}(T)$  to give the best fit to both  $C_v$  and  $C_P$ . The resulting fit in Table I also gave the smallest average percentage volume and sound velocity deviations between the EOS and the data sets of  $\pm 0.22$  and  $\pm 0.53$ , respectively. These P-V-T and  $v_0$  measurements have provided an EOS for a previously unexplored region of the P,T plane with an accuracy comparable to that of the data.

The direct measurement of the derivative of the EOS,  $v_s = V/M^{1/2}[-(\partial V/\partial P)_s]^{-1/2}$  provides additional accuracy of the fit. This improvement was apparent from the fitting of hydrogen data to Eq. (3) without use of sound velocity data. In that case the computed values of  $v_s$  differed by a few per cent from the measured values.

The data sets and the computed values from the EOS were compared in Ref. 9; the average deviations were within the estimated accuracy of the data points.

#### **IV. DERIVED THERMODYNAMIC VALUES**

As described in Ref. 8, the volume-explicit EOS of Eq. (2) is convenient for the derivation of other thermodynamic quantities. These are summarized below.

Isobaric thermal expansion coefficient: 
$$\alpha_{\rm p} \equiv \frac{1}{V} \left(\frac{\partial V}{\partial T}\right)_{\rm p}$$
 (6)

Isothermal compressibility:  $\chi_{\rm T} \equiv -\frac{1}{\rm V} \left(\frac{\partial \rm V}{\partial \rm P}\right)_{\rm T}$  (7)

Ratio of heat capacities: 
$$\gamma \equiv \frac{C_p}{C_v} = \frac{\chi_T}{\chi_s}$$
 (8)

Adiabatic compressibility:  $\gamma \equiv -\frac{1}{V} \left(\frac{\partial V}{\partial P}\right)_{s} = \frac{1}{\rho v_{s}^{2}}$  (9)

The heat capacities are determined from Eqs. (3), (4), and (8). The molar entropy is found from the thermodynamic relation

$$S(P_o,T) - S(P,T) = \int_{P_o}^{P} \left(\frac{\partial V}{\partial T}\right)_{p} dP , \qquad (10)$$

and a relation for the entropy at the normalizing pressure of 2 kbar. Few determinations of entropy are available for deuterium, and as discussed in Ref. 9, the resulting relation was

$$S_{P-2} - S(P_0,T) = -98.830 + 31.715 \ln (T-10). (J/mole-K)$$
 (11)

for fluid D<sub>2</sub> in the 75 < T < 300 K region. The assumption was made that rotating molecules in the solid at T = 0 K have zero entropy.

A FORTRAN program, described in Ref. 8, was modified to give a listing (Table II) of deuterium properties. Table II was prepared at 25 K and 0.5-kbar increments for the quantities  $V_1 v_2, \alpha_2, C_P, \gamma, \chi_2$ , and S. These values, as a function of pressure along an isotherm, are shown in Figs. 20-27, produced by a computer program. Values for  $C_v$  are shown in Fig. 24. The special font was selected to indicate that these curves all refer to values calculated from our EOS.

#### V. COMPARISONS

A comparison of calculated molar volumes from our EOS with values measured by Michels et al.<sup>a</sup> at low pressures, as discussed in Ref. 9, shows agreement of  $\pm 0.4\%$  between 1 to 3 kbar near room temperature. A power-law fit of our measured sound velocities extrapolates smoothly to the calculated low-pressure values of Michels et al.<sup>a</sup> No other sound velocity measurements are available for comparison.

An EOS was obtained for deuterium by Ashurst<sup>12</sup> using a modified Lennard-Jones potential. Comparison with our EOS values shows that significant departures occur at higher pressures; at 275 K and 20 kbar, the variations are 7.8% in volume and 17.7% in sound velocity. Throughout the P-T range, the differences are larger than our estimated accuracy.

We compared the hydrogen and deuterium properties and found that over the P-T plane the molar volumes determined for deuterium are a few per cent lower than for hydrogen. The sound velocities scale within approximately 2% according to kinetic theory, that is,  $V_{H_2}/V_{D_2} = (M_{D_2}/M_{H_2})^{1/3}$ . At low pressures and temperatures, the departures from this scaling law are larger. This scaling is in contrast to solid behavior where the ratio of velocities is reduced. This ratio increases with pressure but does not reach the kinetic-theory value up to the maximum measured pressure of 200 bar along the melting curve.<sup>18</sup> In the liquid phase along the melting curve, the ratio of velocities is greater than the kinetic value and it decreases with increasing pressure toward the kinetic value.<sup>18</sup> No explanation for this isotopic effect has been suggested.

Kerley<sup>14</sup> has derived properties of high-density fluid deuterium from a hard sphere model combined with a cell model for the molecular solid. We compared densities and sound velocities as a function of pressure along several isotherms, as shown in Figs. 28 and 29. There are deviations of about 2% at our highest pressures between our EOS densities and those derived by Kerley. The agreement between our EOS sound velocities and those calculated by Kerley is better than 1.5% for all temperatures and pressures within our range of measurements. We consider this agreement good, although the maximum errors are outside the estimated experimental accuracy.

#### ACKNOWLEDGMENTS

We wish to thank L. C. Schmidt for building the apparatus and for assisting us with some of the measurements and G. I. Kerley, who suggested the procedure, for extrapolating heat capacities at low pressure and for providing comparison data for molar volume and sound velocities from his theoretical model.

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# TABLE II

# CALCULATED THERMODYNAMIC PROPERTIES OF n-D2

#### CALCULATED VALUES FROM E O S FOR D(2) AT 75.0 K.

PRESS.	. VOL.	¥(S)	ALPHA	C(P)	GANNA	CHI (S)	ENTROPY
Kean	CC/MOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE~K
2.00 2.50 3.00 3.50 4.00 4.50	17.952 17.044 16.341 15.770 15.291 14.879	2.090 2.277 2.434 2.571 2.693 2.803	1.921E-03 1.624E-03 1.415E-03 1.259E-03 1.138E-03 1.040E-03	32.375 30.596 29.126 27.869 26.769 25.790	1.150 1.135 1.123 1.114 1.106 1.100	1.020E-01 8.157E-02 6.843E-02 5.920E-02 5.233E-02 4.698E-02	33.56 32.02 30.76 29.69 28.76 27.94

#### CALCULATED VALUES FROM E O S FOR D(2) AT 100.0 K.

PRESS.	VOL.	Y(S)	ALPHA	C(P)	GAMMA	CH1(5)	ENTROPY
KBAR C	C/HOLE	KM/S	/K	J/WALE-K		/KBAR	J/MOLE-K
2.00 2.50 3.50 4.50 5.50 6.50 6.50 6.50	18.926 17.825 16.995 16.333 15.784 15.319 14.914 14.559 14.241 13.955 13.695	2.080 2.266 2.423 2.559 2.681 2.893 2.893 2.987 3.075 3.157 3.235	2.210E-03 1.687E-03 1.657E-03 1.657E-03 1.348E-03 1.348E-03 1.348E-03 1.72E-03 1.072E-03 1.072E-03 9.072E-04 9.072E-04	33. 154 32. 496 31. 947 31. 474 30. 483 30. 344 30. 032 29. 743 29. 743 29. 222	1.257 1.227 1.223 1.185 1.169 1.157 1.146 1.130 1.130 1.123 1.117	1.086E-01 8.618E-02 7.186E-02 6.187E-02 6.187E-02 4.878E-02 4.423E-02 4.423E-02 3.739E-02 3.739E-02 3.248E-02	43.88 42.01 40.47 39.17 38.03 35.13 35.31 34.56 33.87 35.24

#### CALCULATED VALUES FROM E O S FOR D(2) AT 125.0 K.

PRESS.	YOL.	V(S)	ALPHA	C(P)	GAPMA	CHI(S)	ENTROPY
KBAR C	C/HOLE	101/5	/K	J/MOLE-K		/KBAR	J/MOLE-K
2233544558888888888888888888888888888888	20.003 18.692 17.721 16.933 15.805 15.805 15.805 14.612 14.612 14.612 14.298 13.096 13.096 13.096 13.096 13.096 13.299 13.096 13.299 13.299 13.299 13.299	2.043 2.386 2.524 2.524 2.759 2.862 2.957 3.046 3.208 3.208 3.208 3.208 3.423	2.195E-03 1.891E-03 1.671E-03 1.573E-03 1.266E-03 1.266E-03 1.102E-03 1.102E-03 1.102E-03 1.102E-03 1.102E-04 9.328E-04 8.594E-04 8.592E-04 7.292E-04 6.829E-04 6.824E-04	33.088 32.660 32.665 32.494 32.202 32.074 31.842 31.737 31.637 31.637 31.541 31.363 31.279 31.196 31.120 31.044 30.899	1.306 1.272 1.245 1.206 1.191 1.758 1.167 1.158 1.167 1.158 1.167 1.130 1.143 1.130 1.125 1.121 1.109 1.109	1.190E-01 9.343E-02 7.724E-02 5.783E-02 4.653E-02 3.625E-02 3.625E-02 3.625E-02 3.625E-02 3.625E-02 3.625E-02 2.379E-02 2.671E-02 2.671E-02 2.314E-02 3.3142E-02 3.3142E-02 3.3142E-02 3.3142E-02 3.3142E-02 3.3142E-02	51.66 49.69 48.07 45.51 44.55 44.55 44.55 44.65 45.84 41.14 39.81 38.65 38.13 39.25 38.13 37.615 35.85

#### CALCULATED VALUES FROM E G S FOR D(2) AT 150.0 K.

PRESS.	VOL.	A( 2)	ALPHA	C(P)	GANNA	CHI (S)	ENTROPY
KBAR	CC/NOLE	K71/S	/K	J/HOLE-K		<b>KBAR</b>	J/MOLE-K
S°00	21-108	2.008	2.103E-03	32.751	1.329	1.2996-01	57.89
2.50	19.583	2.192	1.8268-03	32.714	1.296	1.0118-01	55.91
3.00	18.467	Z. 350	1.6246-03	32.677	1.269	8.797E-02	54.27
3.50	17.601	Z.489	1.409E-03	32.642	1.247	7.050E-02	52.65
4.00	16.899	2.614	1.3456-03	32.607	1.227	6.139E-02	51.67
4.50	16.313	2.727	1.2446-03	32.573	1.214	5.445E-02	50.60
5.00	15.813	2.831	1.160E-03	32.539	1.200	4.8986-02	49.63
5.50	15.378	2.927	1.0896-03	32.507	1.189	4.4548-02	48.76
6.00	14.995	3,017	1.0275-03	32.475	1,179	4.C88E-02	47.95
6.50	14-653	3,102	9.7395-04	32.+43	1.170	3.779E-02	47.21
7.00	14.344	3, 182	9.2596-04	32.412	1.162	3.516E-02	46.52
7.50	14.065	3,258	8.8518-04	32, 382	1.155	3.289E-02	45.88
8.00	13.809	3,330	8.477E-04	32.352	1.149	3.090E-02	45.28
<b>8</b> .50	13.573	3, 399	8.140E-04	32.323	1.143	2.915E-02	44.71
9.00	13.356	3.400	7.834E-04	32.794	1.138	2.7606-02	44.17
9.50	13.153	3.529	7.5568-04	32.265	1.133	2.6216-02	43.66
10.00	12.964	1.591	7.3025-04	2.27	1, 129	2.4045-02	43.18
10.50	12.784	3.650	7.0586-04	20.209	1.125	2. 3825-02	12.71
11.00	12.621	1 202	A ASZE-NA	12. 182	1 121	2. 2706-07	42.27
11 50	13.245	\$ 742	A 4515-14	12:155	111	2 1858-02	A1 85
12 00	12 812	1 816	A. LASE-MA	12 124	1 115	2 0005-07	11 11
12 50	12 174	3 RAR	A 2026-04	32 101	1 112	2 0195.02	11 05
11.00	12 041	2 010	4 1206-04	12 075	1 100	1 0445-02	20 47
13.60	11 014	TOAR	6 0796-04	15 46	1 107	1 8285-02	20.31
1	11 205	1 012		12 021	1 100	1 0100-02	20 04
11.00		1.041	5 7015-04	21 000		1.0140-02	37.70
12.20	11.000	4 100		31.770	1.101	1-1336-06	37.00
12.00	11.209	7.107		21.7(2	1.077	1.100-02	27.50
12.20	11.404	4.124	2.0745-04	51.940	1.007	1.049E-U2	20.78
19-00	11.362	4.198	2.547E-04	21.452	1.095	1.0000-02	58.07

CALCULATED VALUES FROM E O S FOR D(2) AT 175.0 K.

PRESS.	. VOL.	V(S)	ALPHA	C(P)	64 <b>44</b> 4	CHI (5)	ENTROPY
KSAR	CC/HOLE	K91/S	/K	37M01.E-K		/KBAR	J/MOLE-K
2.00	22.218	1.982	1.996E-03	32.359	1.341	1.404E-01	63.11
2.50	20.477	Z. 164	1.745E-03	32.413	1.310	1.085E-01	61.12
2.00	12.51	5.361	1.3008403	34.923	1. (0)	0.07%	27.2
3.20	10.24/	2. NOU	1.41/2-03	36.003	1.53	4 4 2 2 - 0	20.07
1.60	17.400	2.707	1 2000 -01	12 521	1 210	5 7118-00	55 11
2.2V	14.272	2 804	1.1296-01	3.30	1.217	5 1175-00	\$1.34
1.22	15.797	2.901	1.0526-03	32.544	1.204	A. A SAS-02	61.04
6. M	15.380	2.002	1.004F-03	\$2.553	1.195	4.2644-02	53.16
6.50	15.010	3.(78	9.5328-04	32.559	1.186	3.9336-02	52.42
7.00	14.678	3,158	9.005E-04	32.562	1.178	3.6516-02	51.73
7.50	14.377	3. 235	8.6872-04	32.564	1,172	3.40%-02	51.00
8.00	14,102	3.308	8.330E-04	32, 565	9,164	3.1986-02	50.46
8.50	13.850	3.378	8.008E-04	32.564	1.158	3.0126-02	49.91
9.00	13.618	3.445	7.716E-04	32.563	1.153	2.8475-02	49.37
9.50	13.403	3.510	7.449E-04	32.560	1.148	2.70CE-02	48.85
10.00	13-202	3,571	7.205E-04	32.557	1.143	c. 569E-02	41.37
10.50	13-014	3-631	6.9808-04	32.554	1.159	2.4495-02	47.90
11.00	12.85	3.689	6.7720-04	32.249	1.752	2.3416-02	47.40
11.50	12.013	3. (22	0.3/96-04	34.252	1.132	2.242E-UC	*/-05
12.00	12.310	3.799	0.000-04	25-232	1.120	2.1528-02	40.92
15.20	12.309	3.022		32.23	1.13	2.0091-02	-0.23
13.00	12.228	3.70		36.550	1.166	1.07922-02	47.07
13.20	11 049	3.733	5,7275-04	17 514	1 1 1 7	1.0000-02	46 11
	11.000	4 0.0	5 4415-04	32.514	1 114	1 7036-02	44 70
19.20	11 778	1.005	5.5316-04	12 500	1 112	1 7145-02	
12.53	11.621	A. 141	5.4226-04	101	1.109	1.427-02	14.16
16.00	11.514	4. 185	5. 3125-04	0.45	1.102	1.6327-02	43.83
16.50	11.412	4.228	5.2078-04	32.477	1.105	1.5846-02	43.53
17.00	11.314	4.270	5.108E-04	32.469	1.103	1.5406-02	43.24
12.50	11.220	4.312	5.0136-04	32.461	1.102	1.4986-02	42.95
18.00	11.129	4.353	4.923E-0	32.452	1,100	1.4586-02	42.68
18.50	11.012	4.392	4.837E-04	32.444	1.098	1.420E-02	42.41
19.00	10.957	4,432	4.755E-04	32.435	1.097	1.3856-02	42.14
19.50	10.875	4,470	4.676E-04	32.426	1.095	1.3516-02	41.89
20.00	10.796	4,508	4.601E-04	32,418	1.094	1.3188-02	41.63
20.50	10.720	4.545	4.529E-04	32.409	1.092	1.2886-02	41.39

CALCULATED VALUES FROM E O S FOR 5(2) AT 200.0 K.

PRESS.	VOL.	V(S)	ALPHA	C(P)	GANNA	CHI (S)	ENTROPY
KBAR	CC/MOLE	KM/5	/K	J/MOLE-K		/KEAR	J/MOLE-K
2.00	23.323	1.964	1.8905-03	32.001	1.347	1.5018-0	1 67.58
3.00	19.965	2,209	1.4955-03	32. 107 10. 178	1.296	9.3776-0	2 43.90
3.50	18.893	2.437	1.3638-03	32.241	1.276	7.893E-0	2 62.59
4.00	18.035	2.562	1.256E-03	32.293	1.258	6.818E-0	2 61.39
4.50	17.528	2.6/0	1.100E-03	8.3%	1.244	6.004E-0	S Christ
2.00	10.731	2 879	1.0742405	****	1.231	J. JOYE-U	5 77.33
6.00	15.766	2.971	9.7656-04	0.00	1.209	4.439-0	5 57.74
6.50	15.367	3.057	9.285E-04	32.457	1.200	4.081E-0	2 56.94
7.00	15.01	3.139	8.862E-04	32.478	1.192	3.7826-0	2 54.25
7.50	14.659	3.210	8.484E-04	32.497	1.185	3-222-0	\$ \$5.60
5.W	14. 370	3.00	2 8286-04	52.214	1.1/8	3.3012-0	
0.JU	13.441	3.428	7.5596-04	32.547	1.144	2.037-0	5 51.80
9.50	13.652	3.493	7.30AE-04	32.553	1, 161	2.7778-6	2 53.36
10.00	13.440	3.555	7.070E-04	32.563	1.156	2.639E-0	2 52.89
10.50	13-241	3.615	6-854E-04	32.572	1.152	2.514E-0	2 52.43
11.00	13.000	3.0/4	6.655E-04	32.200	1.14	2.4016-0	<u> </u>
12 00	12.001	3.730	A. 2075-04	32.30/	1 140	2 2036-0	2 31.20
12.50	12.561	3.838	6.1365-04	\$ 599	1.137	2.1165-0	50.76
13.00	12.414	3.890	5.9868-04	32.604	1.134	2.0368-0	2 50.34
13.50	12.274	3.940	5.8446-04	32.608	1.131	1.962E-0	2 50.02
14-00	12.142	3.989	5.7116-04	32.612	1.128	1.8946-0	2 49.66
16.50			2.200E-UA	52.312	1.120	1.8308-0	2 49-32
15.50	11 778	A. 129	5.3556-04	32.619	1 121	1.7155-0	5 40.57
16.00	11.667	6.173	5.2486-04	0.621	1.119	1.6625-0	46.36
16.50	11.561	4-217	5.147E-04	32.622	1.116	1.6138-0	2 48.06
17.00	11.459	4.260	5.051E-04	32.623	1.114	1.567E-0	2 47.77
17.50	11.361	4.302	4.960E-04	32.623	1.112	1.5248-0	2 47.48
	11.200	4.242	4.873E-UA	32.023	1.111	1.4858-04	
19.00	11.087	4.422	4.7095-04	32.623	1 107	1 4075-00	2 40.43
19.50	11.003	4.461	4.633E-04	32.672	1.106	1.3726-0	2 46.41
20.00	10.921	4.499	4.560E-04	32.621	1.104	1.339E-0	2 46.16
20.50	10.841	4.536	4.490E-04	32.620	1.102	1.307E-0	2 45.91
21-00	10.764	4.573	4.4226-04	32.618	1.101	1.277E-Q	2 45.67
51-20	10.690	4.007	4.325E-04	52.61/	1.100	1.2496-0	2 42.43
22.50	10.547	4.680	L 2355-04	32 413	1.007	1 1056-0	2 3 20
CC+ JU	10.341			JE-013	1.071	1.1736-0	L 49070

CALCULATED VALUES FROM E O S FOR D(2) AT 225.0 K.

	PRESS.	VOL.	¥( 5)	ALPHA	C(P)	GANNA	QU(S)	ENTROPY
	KBAR	CC/MOLE	101/S	/K	J/HOLE-K		/KBAR	J/MOLE-K
	2.00	24.422	1.952	1.793E-03	31.715	1.350	1.5918-0	1 71.50
	<b>2.50</b>	22.254	2.128	1.5876-03	31.637	1.325	1.2206-0	1 69.54
	3.00	20.70	2.282	7.4326-03	31.934	1.303	9.8698-0	2 67.92
	3.50	19.534	2.420	1.310E-03	32.013	1.284	8.281E-0	2 66.55
	4.00	18.600	2.544	1.2116-03	32.079	1.266	7.1326-0	2 65.35
	4.50	17.833	2.658	1.1296-03	32.136	1.254	6.264E-0	2 64.23
	5.00	17, 187	2.763	1.0608-03	32.185	1.242	5.585E-0	2 63.32
	5.50	16.633	2.862	1.000E-03	32.225	1.230	5.0416-0	2 62.45
	6.00	16.150	2.953	9.487E-04	32.266	1.221	4-595E-O	2 61.66
1	6.50	15.723	3.040	9.0345-04	32.301	1.212	4-2228-0	2 60.92
	7.00	15.342	3,122	8.632E-04	32.331	1,204	3.9076-0	2 60.23
	7.50	16.999	3,200	8.273E-04	32.359	1, 196	3.63/JE-D	2 59.59
	6.00	14.688	3.274	7.950E-04	32.384	1.190	3.4016-0	2 58.99
	8.50	14.404	3.345	7.558E-06	32.406	1.184	3.195E-0	2 58.42
	9.00	14.142	3.413	7.3916-06	32.427	1, 178	3.013E-0	2 57.89
	9.50	13.901	3.478	7.1685-04	32.446	1.173	2.8528-0	2 57.38
1	0.00	13.677	3.541	6.924E-04	32.464	1.168	2.707E-0	2 56.89
1	0.50	13.448	3.602	6.718F-04	32.480	1.163	2.5766-0	2 56.43
1	1.00	13.273	3.441	A. 5245-04	12.495	1,159	2.458E-0	2 55.09
j.	1 50	11 000	\$ 717	A LOC-M	10. SOL	1, 155	2.3516-0	3 55.54
	່າທີ່	12 917	1 771	A 1816-04	0.01	1.152	2.2526-0	2 55 14
	2 50	12 754	1 826	A 0205-04	0.00	2 148	2. 1426-0	5 51 74
	້ະກິ	12 400	3 979	S BETE-OL	5.41	1 145		2 54 30
	1. KV	12 .54	1 0 20	5 7175-01	3.44	1 175	2 0028-0	
1	2.70	12 316	1 070	5 4102-04	12 642	1 110	1 0116-0	
		12.313	4 024		12 671	1 14	1.7510-0	
1	2.20	12.105	4.020	5 1816-04	12 670	1 176	1.0050-0	2 53.33
	2.00	12.030	4.0/3	7-3036-04	26.3/7		1.0040-0	
	2.30	11.730		5 1736-04	32.300	1 1 20	1.4076-0	2 32.00
		11-920	9.109		32-373	1.127	1.0722-0	
19	<u>9.20</u>	11./10	•• <u>2</u> 00	3.0/36-04	×	1-14	1.0416-0	2 <u>2</u> 2. W
1	<u>.</u>	11.00	1.01	4.98(2-04	32.00		1.394240	\$ 21.78
1	<u>'-20</u>	11.702	<b>*</b> 33	4.8Y3E-UA	22-011	1-122	1.747240	2 21.49
10	o.w	11.408	- 22	4.0U/E-U4	22-010	1.121	1.307E-0	2 21-21
1	8. <u>50</u>	11.309	4.3/4	4.728E-04	52.020	1.119	1.40/6-0	2 20-94
T T	9.00	11.218	4.414	4.651E-04	32.024	1.11/	1.429E-0	2 50-68
I. I.	9.50	11.130	4.453	4-577E-04	25.050	1.115	1.393E-0	2 50.42
2	0.00	11.045	4.497	4.506E-04	52.632	1.114	1.359E-0	2 50.17
<u>Š</u> t	0.50	10.963	4.529	4.439E-04	32.635	1.112	1.326E-0	2 49.92
S	1.00	10.883	4.566	4.372E-04	32.638	1.311	1.296E-0	2 49.68
2	1.50	10.806	4.602	4.310E-04	32.641	1.109	1.2666-0	2 49.45
2	2.00	10.731	4.638	4.249E-04	32.643	1.108	1.2386-0	2 49.22
2	5.20	10.659	4.673	4_191E-04	32.645	1.107	1.211E-0	2 48,99

## CALCULATED VALUES FROM E O S FOR DC2) AT 250.0 K.

PRESS.	. YOL.	¥(S)	ALPHA	C(P)	GANNA	041(5)	ENTROPY
KBAR	CC/MOLE	101/5	/K	J/MOLE-K		/XBAR	J/MOLE-K
2.00	25.513	1.945	1.7048-03	31.517	1.351	1.6748-0	74.99
2.50	23.135	2.118	1.517E-03	31.649	1.328	1.280E-0	73.06
3.00	21.448	2.270	1.3/48-03	51.754	1.500	1.035E-0	1 11.42
2.20	20.1/2	2.406	1.2016-03	51.841	1.31	5.040E-U	
4.00	19.102	2.550	1.1086-03	21.212	1.2/0	7.429E-U	69.99
4.20	18.332	2.644	1.0928-03	21.9/8	1.202	6.510E-0	07.82
3-00	17-041	2.749	1.02/E-05	<u>x</u> .m	1.00	2./94E-U	60.8/
3.20	17.048	2.84/	9.7000-04	32.122	1.200	3.22E-U	
0-00	10.23	2.959	9.218E-UN	×.10	1.20	4.730E-0	<u>(</u> ), (]
<u>ð</u> -20	10.07	2.020		32.104	1.221	4-558E-0	04-47
1.00	13-613	3.105	5.4102-04	7.125	1.214	4.UC/E-U	03.14
1.20	12.204	5.180	8.000E-04	3.22	1.200	3.743E-U	<u>[</u> ]
9.00	14.979	3.260	7.759E-04	52.201	1.200	3.497E-U	62.35
8-20	14-0/9	3.335	7.480E-04	52.250	1.194	3. 28ZE-0	61.97
<b>A</b> -00	14-402	3.400	(. <u>(</u> .)	56.213	1.100	3.0925-0	01.49
<u>x-30</u>	14.147	3.400	0.77.6-18	56.250	1.185	2.923E-0	00-32
າແໜ	13.915	2.20	0.//8E-UA	ર્સ-ઝુ(	1.1(8	2.773E-0	00-47
10.20	13.094	3.590	0.56(E-UA	2.50	1.1/4	2.637E-0	60.00
11.00	13.467	3.649	6. 59/E-UA	36.22	1.109	2.514E-0	59.50
11.20	13.00	3.705	0.220E-UA	52.412	1.102	2.4025-0	59.14
12.00	13.110	3.762	6.06/E-04	32.428	1.102	2.300E-0	58.74
12.50	12.945	3-515	5.918E-06	32.444	1.155	2.207E-0	58.35
12.00	12.(83	3.000	2.//8E-UA	24-928	1.122	2.121E-U	: <u>3/.y/</u>
12-20	12-0-22	2.919	2.04/E-UA	X•*/1	1, 152	2.041E-0	27.01
14.00	12.48/	5.965	2.225-04	52.483	1.169	1.968E-0	57.26
14-20	12.350	4.017	2.4005-04	32.493	1.140	1.900E-0	50.92
15.00	12.210	<b>6-064</b>	5.296E-04	32.500	1.144	1.83AE-U	50.59
15.20	12.033	4.110	5.191E-04	52.510	1.151	1.777E-0	56.27
10.00	11.9/3	6.155	2.0922-04	22.20	1.159	1./21E-0	>>.%
10.20	11.838	4 TYY	4.990E-04	36.335	1.150	1.009E-U2	>>.00
17.00	11.758	4.243	4.905E-04	¥.24	1-125	1.620E-02	22-22
11.20	11-042	6.(3)	4.8222-04	2.22	1.152	1.5/4E-U	55.08
18.00	11.261	4.320	4.74UE-UA	22.200	1.150	1.550E-02	24-51
15-20	11.443	4.507	4.00(t-U)	32-39	1.128	1.489E-04	24-24
12.00	11.355	4.407	4.38/E-04	32.214	1.129	1.450E-0	24.27
19-20	11.27	4.440	4-212E-04	32-251	1.13	1.413E-02	24-01
20.00	11.109	4.452	4.44/2-04	32.387	1.12	1.5/85-02	23.70
20.20	11.004	4.266	5.301E-04	35.343	1.12	1.345E-02	22.22
<1.W	11.002	4-201	4.31/2-04	26.388	1.120	1.3156-02	23.28
21-20	10.922	4.595	4.230E-04	32.004	1.115	1.2336-02	55.06
çç.00	10.845	4-632	4.19/E-04	22.000	1.11/	1.2548-02	52.91
22 <b>-</b> 50	19.770	4.005	4.147E-04	32.615	1.115	1.2276-02	52.59

## CALCULATED VALUES FROM E O S FOR D(2) AT 275.0 K.

PRESS.	VOL.	¥(5)	ALPHA	C(P)	ganna	CHI(S)	ENTROPY
KBAR	CC/HOLE	109/5	/K	1/MOLE-K		/KBAR	J/MOLE-K
Ş.00	25.596	1.941	1.6245-03	31.412	1.351	1.7528-01	78.13
5.00	22.182	2. 261	1.37/06-03	31.656	1.392	1.0776-01	74.60
3. Sõ	20.806	2.396	1.2156-03	31.745	1.296	8.9946-02	73.24
4.00	19.720	2.519	1.1296-03	31.821	1.281	7.7128-02	72.06
4.50	18.834	2.632	1.0576-03	31.887	1.269	6.746E-02	71.00
5.00	18.093	2.737	9.9548-04	31.945	1.27	5.994E-02	70.05
5.50	17.460	2.835	9.4258-04	51.976	1.247	5.3918-02	67.20
P. (0)	10.912	2.761	5.703E-04	32.042	1.230	4.4995-02	47 48
2.00	16.001	3.096	8.1936-06	32,120	1.222	4. 143-02	AZ.00
2.50	15.617	3. 174	7.8686-04	32.135	1.215	3.847E-02	66.37
8.00	15.269	3.249	7.575E-04	32.186	1.209	3.590E-02	65.77
8.50	14.953	3.320	7.3088-04	32.215	1.203	3.3666-02	65.21
9.00	14.663	3.389	7.065E-04	32.242	1.197	3.1686-02	64.68
<u>9.50</u>	14-396	3.455	6.542E-04	32.207	1-192	2.9938-02	64.17
10.00	14, 144	2.218	0.0300-08	¥.80	1.10/	2 4045-02	43.27
11.00	13 204	000.0	4 37/16-04	5:16	1.178	2 5446-02	22.20
11.50	13,503	1.696	6.106E-06	9.35	1.174	2.4536-02	62.37
12.00	13.315	3.752	5.9536-04	12.349	3.171	2.3476-02	67.97
12.50	13, 137	3.806	5.8096-04	32, 386	1.167	2.251E-02	61.58
13.00	12,969	3.859	5.6758-04	32.402	1.164	2.1026-02	61.20
13.50	12.810	3.910	5.5486-04	32.417	1.161	2.080E-02	60.84
14.00	12.660	3.960	5.4298-04	32.431	1-158	2.004E-02	60.49
14-20	16-210	4.008	2.2106-04	¥.#	1.122	1.9336-02	00.12
15.66	15.350	•.000	5.0000-04	X. 57	1.122	1.0002-02	59.63
13.30	12.20	4 147	5 01 25-04	12 480	1 147	1 7406-02	59 20
16.50	12.005	A. 192	4.9205-04	0.791	1.145	1. 6946-02	58.90
17.00	11.892	4.235	4.8336-04	2.502	1.143	1.6458-02	58.61
17.50	11.782	4,278	4.750E-04	32.511	1,141	1.598E-02	58.33
18.00	11.677	4.319	4.671E-04	32.521	1.139	1.5538-02	58.05
18.50	11.576	4.360	4-595E-04	32.530	1.137	1.511E-02	57.78
19.00	11.478	4.400	4.5236-04	32.536	1.135	1.471E-02	57.52
19.20	11.384	4.640	4.453E-UA	32.340	1.135	1.4556-02	21.20
20.00	11 204	4.517	4 1225-04	12 541	1 130	1.37/2-02	54 77
21.00	11,121	1.554	4.261F-04	12.44	1,128	1.3316-02	54 52
21.50	11.039	4.521	4.20%E-04	32.575	1.127	1.3006-02	56.29
22.00	10,959	4.627	4.1446-04	32.581	1.125	1.270E-02	56.06
55-20	10.582	4.663	4.089E-04	32.587	1,124	1.2426-02	55.84

#### CALCULATED VALUES FROM E O S FOR D(2) AT 300.0 K.

PRESS.	VOL.	(2)¥	ALPHA	C(P)	GNMA	01(5)	ENTROPY
KDAR	CC/HOLE	KM/S	/K	J/MOLE-K		/KBAR	J/MOLE-K
2.00	27.672	1.940	1.5518-03	31.401	1.3.9	1.8246-0	80.99
÷20	23.013	2.107	1.3736-03	51.333	1.330	1.3712-03	77.07
1.00	21.14	5.03	1.1726-03	1.711	1.7.9	9.1755-00	76.13
<b>L</b> .00	20.776	2.510	1.0928-03	31.809	1.285	7.9648-0	74.95
4.50	19.330	2.623	1.0248-03	31.875	1.24	6.973E-G	73.91
5.00	18.542	2.727	9.6618-04	31.933	1.263	6.187E-0	72.96
5.50	17.871	2.825	9.161E-04	31.985	1.23	5.558E-02	72.11
6.00	17.290	2.917	8.7236-04	ន៍.លីរ	1.264	5.0H4E-02	. <u>7</u> . <u>2</u>
\$.X	16.750	3.005	7 0015-04	×	1.250	4.01/2-04	11.00
2.00	16.023	3-080	7 4408-04	30.144	1.222	1 0175-72	44.76
<b>Å</b> . m	15.554	1,200	7.4005-04	5.17	1.21	1416-0	4.70
8.50	15.225	£310	7.145E-04	32.208	1.210	14486-02	68.14
9.00	14.921	3.379	6.9128-04	32.236	1.205	3.2438-07	67.61
9.50	14.641	3.445	6.698E-04	32.262	1.200	3.065E-0	67.11
10.00	14.382	3.509	6.501E-04	¥.26	1.195	2.899E-U	60.03
10.50	14, 142	3-20	6. 518E-UA	×	1.190	2.735	00.17
11.00	13.918	2.000	5 0005-04	¥.%	1 182	2 5026-02	45 11
12.00	11.512	3.743	5 84 3 - 14	3.44	1.120	2. 1915-00	AL.01
12.50	13.327	3,798	5.704E-04	32.386	1.175	2.2938-0	64.52
13.00	13.153	3.850	5.574E-04	32.402	1.172	2.202E-02	64.15
13.50	12.958	3.902	5.452E-04	32.418	1.169	2.1172-02	63.79
14.00	12.831	3-952	5.337E-04	52.433	1.166	2.0396-02	63.44
14.50	12.682	4. <u>00</u>	5.225E-04	2.47	1.163	1.967E-02	63.10
12.00	12.241	4.045	5.1246-04	¥.%	1.100	1.0772-02	02.78
15.30	12 2/7	A 140	A 0775-04	5.44	1,155	1.7776-02	62.40
16.50	12.154	4 185	4.845E-04	32.498	1.153	1.7228-02	61.86
17.00	12.035	4-228	4.761E-04	32.509	1.151	1.6718-02	61.57
17.50	11.922	4.271	4.680E-04	32.570	1.149	1.6226-02	61.28
18.00	11.813	4.313	4.603E-04	52.550	1.146	1.576E-02	61.01
18.50	11.709	4.354	4.530E-04	2.50	1.144	1.533E-02	60.74
19.00	11.005	<b>6.376</b>	4.4372-04	32.349	1-1-3	1.472-00	00.40
20 00	11.417	4.473	4. 3275-04	32.564	1.130	1 4165-02	500.62
20.50	11.324	4511	4.2648-04	32.575	1.137	1.3816-02	59.73
21.00	11.239	4.549	4.204E-04	32.583	1.136	1.3486-0	59.49
21.50	11, 154	4.586	4.147E-04	32.590	1.134	1.3166-02	59.25
22.00	11.072	4.622	4.091E-04	32.597	1.133	1.286E-02	2 59.03
22.50	10,993	4.658	4.038E-04	32.604	1. 131	1.258E~02	2 58.80



Log  $v_* v_* \log P$  in fluid <u>n</u>-D<sub>1</sub> at T = 300 K. The solid line is from a least-squares fit of Eq. (1) to the data.



Log V vs log P in fluid  $\underline{n}$ -D<sub>1</sub> at T = 300 K.



Fig. 2.

Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 1.



Fig. 4.

Log  $v_s$  vs log P in fluid <u>n</u>-D<sub>2</sub> at T = 245 K. The solid line is from a least-squares fit of Eq. (1) to the data.



Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 4



Log  $v_{\bullet}$  vs log P in fluid <u>n</u>-D<sub>2</sub> at T = 187 K. The solid line is from a least-squares fit of Eq. (1) to the data.



Log V vs log P in fluid  $\underline{n}$ -D<sub>2</sub> at T = 245 K.



Fig. 8. Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 7.



Fig. 9. Log V vs log P in fluid  $\underline{n}$ -D<sub>1</sub> at T = 187 K.



Fig. 11.

Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 10.



Fig. 10. Log  $v_{\bullet}$  vs log P in fluid <u>n</u>-D<sub>2</sub> at T = 145 K. The solid line is from a least-squares fit of Eq. (1) to the data.



Log V vs log P in fluid  $\underline{n}$ -D<sub>1</sub> at T = 145 K.



Log  $v_{\bullet}$  vs log P in fluid <u>n</u>-D<sub>1</sub> at T = 102 K. The solid line is from a least-squares fit of Eq. (1) to the data.



Log V vs log P in fluid  $n-D_2$  at T = 102 K.



Fig. 14.

Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 13.



Fig. 16.

Log v. vs log P in fluid <u>n</u>-D<sub>2</sub> at T = 75 K for three runs: 0, T = 75.15 K on January 25, 1977;  $\Delta$ , T = 75.20 K on February 7, 1977; and +, T = 75.00 K on March 2, 1977.



Fig. 17.

Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for data of Fig. 16.

Fig. 18. Log V vs log P in fluid  $\underline{n}$ - $D_1$  at T = 75 K for three runs with symbols as in Fig. 16.





Fig. 19.

Percentage deviations of the sound velocity values from the least-squares fit to Eq. (1) for the nine different runs in the temperature region  $295.3 \pm 0.8 \text{ K}$ .





Fig. 23. Heat capacity at constant pressure of fluid  $\underline{n}-D_2$  vs pressure along five isotherms computed from Eqs. (4), (7), and (8). Symbols are the same as in Fig. 20.



Fig. 24. Heat capacity at constant volume of fluid <u>n</u>-D, vs pressure along five isotherms computed from Eqs. (4), (7), and (8). Symbols are the same as in Fig. 20.







Log pressure of fluid  $n-D_2$  vs density along three isotherms, truncated at the freezing pressure. The points are calculated from molar volumes in Table II, where  $\bullet$ ,  $T = 300 \ K$ ;  $\blacksquare$ , T= 200 K; and  $\bullet$ ,  $T = 100 \ K$ . The smooth curves are derived densities by Kerley, Ref. 14. In addition, the freezing pressure is indicated by the horizontal lines.

Fig. 29.

Sound velocity of fluid  $n-D_2$  vs pressure along two isotherms. The points are calculated from molar volumes in Table II, where  $\bullet$ , T = 300 K and  $\bullet$ , T = 100 K. The smooth curves are by Kerley, Ref. 14. The lower temperature isotherm is truncated at the freezing pressure.

