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Survey of the Properties of the Hydrogen Isotopes Below Their Critical Temperatures

National Bureau of Standards

APR 29 1974

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Issued August 1973

National Bureau of Standards Technical Note 641

Nat. Bur. Stand. (U.S.), Tech. Note 641, 122 pages (October 1973)

CODEN: NBTNAE

For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402

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List of Symbols

a	lattice parameter	T	temperature
c	lattice parameter	T*	reduced temperature
c_i	concentration, also indicated by []	T_c	critical temperature
dt	derivative with respect to time	T_t	triple point temperature
k	thermal conductivity	W	thermal resistance
n	exponent	W_i	intrinsic thermal resistance
n	index of refraction, also n_λ	V	volume
	n_∞ ... at infinite wave length	V	velocity of sound
p	specific polarization	Z	compressibility factor, PV/RT
r_λ	specific refraction at wave length λ	α	parameter in equation
	r_∞ ... at infinite wave length	α	thermal diffusivity
v	specific volume	α	thermal expansion coefficient
v_ℓ	longitudinal velocity of sound in the solid	β_i	"instantaneous" compressibility \simeq isothermal
v_t	transverse velocity of sound in the solid	β_s	adiabatic compressibility
x	quality	β_T	isothermal compressibility
A, B, C	coefficients in equations	γ	percent orthohydrogen
B	second virial coefficient	γ_P	molar fraction of p-D ₂
C	third virial coefficient	γ	specific heat ratio, C_p/C_v
C_{11} C_{12} C_{13} C_{33} C_{44}	elastic constants	γ	surface tension
C_p	heat capacity at constant pressure	ϵ	dielectric constant
	C_p^0 ... of the ideal gas	ϵ	relative elongation
C_s	heat capacity at saturation, solid or liquid	ϵ/k	molecular parameter
C_v	heat capacity at constant volume	η	viscosity
D_{ij}	diffusion coefficient	θ	Debye temperature
D_{ij}^T	thermal diffusion coefficient	λ	thermal conductivity
E	Young's modulus	λ_p	... of the para isomer
E_0^g	reference, or ideal gas internal energy	λ	wave length
G	shear modulus	Λ, Λ^*	quantum correction parameter
H	enthalpy	μ	viscosity
	H_0 reference, or ideal gas enthalpy	ρ	density
J_i	matter flux	ρ_0	reference density
L-L	Lorentz-Lorenz function	σ	molecular parameter
M	molecular weight	σ_b	tensile strength
K, K'	velocity constants	$\sigma_{0.1}$	nominal yield stress
N_{pr}	Prandtl number	χ_T	isothermal compressibility
P	molar polarizability		
P	pressure		
	P_a pressure adjustment function		
	P_t triple point pressure		
R	gas constant		
S	entropy		

SURVEY OF THE PROPERTIES OF THE HYDROGEN ISOTOPES
BELOW THEIR CRITICAL TEMPERATURES

by

H. M. Roder, G. E. Childs, R. D. McCarty, and P. E. Angerhofer

The survey covers PVT, thermodynamic, thermal, transport, electrical radiative and mechanical properties. All isotopic as well as ortho-para modifications of hydrogen have been included. Temperatures are limited to those below the respective critical points, in general below 40 K. The pressure range is not restricted, that is solid, liquid, and gas phases are covered. However, with the exception of hydrogen, very little data exists at pressures other than saturation. The literature surveyed includes all references available to the Cryogenic Data Center up to June of 1972, and for several subjects, through March of 1973. The total number of documents considered was nearly 1500 of which about 10 percent contain pertinent information and are referenced in this report. The various properties are presented in the form of tables or graphs; if extensive tables have been published elsewhere, the reader is referred to the original sources.

Key words: Compilation; density; deuterium; electrical properties; enthalpy; entropy; fixed points; hydrogen; mechanical properties; optical properties; specific heat; thermophysical properties; transport properties; tritium; vapor pressure.

1. Introduction

Hydrogen in solid, slush, and liquid states has long been used in the space industry as a fuel and a refrigerant, and in nuclear physics as targets for high energy beams and in bubble chambers. A number of new applications are now being discussed in the context of a "hydrogen economy" in light of the National energy crisis. These potential applications include the use of hydrogen as a fuel in automobiles, trains, and core-city refrigeration, the generation of hydrogen in sea water power plants, its transportation as slush, and power generation in thermonuclear fusion. A survey of properties of the solid and liquid hydrogens is therefore very timely.

Coverage

The survey covers a wide range of properties including PVT, thermodynamic, thermal, transport, electrical, radiative and mechanical. All isotopic as well as ortho-para modifications of hydrogen have been included. Temperatures are limited to those below the respective critical points, i.e., in general below 40 K. The pressure range is not restricted, that is solid, liquid, and gas phases are covered by the survey. However, with the exception of hydrogen itself, very little data exist at pressures other than saturation. Whenever the data tables in the literature are extensive, for example if the single phase gas region or the compressed liquid states are covered, then we have presented the saturation tables as an example of the kind of information that is available. The user is referred to the original sources for additional values. The literature surveyed includes all references available to the Cryogenic Data Center up to June of 1972. For several subjects, whenever one of the authors was aware of more recent pertinent papers, the survey extends through March of 1973. The total number of documents considered in this survey was nearly 1500 of which about 10 percent contain pertinent information and are referenced in this report. Referencing is done by the accession numbers of the Cryogenic Data Center because it was convenient for the authors and because we wish to remain compatible with the bibliographies furnished to the sponsor at the beginning of the contract.

Selection Criteria

In selecting sources of data, a critical evaluation was not made. However, we used the following guide.

- a) If several sets of data exist, which, in our judgement, are the "best data"?
- b) Are these values consistent with other properties presented in the report?
- c) Are tables or graphs available? Are they in a convenient form?
- d) Can we find any values at all - even if estimated?

Quite often we assemble information on all of the hydrogens into a single table to allow rapid comparison, as for example in the tables of fixed points. In general, if an author presented both experimental data and a functional representation for the data, then we present the calculated values because they will be easier to interpolate.

Inconsistencies in the Data

Many inconsistencies or discontinuities exist in the data presented because the values originate from many diverse sources. Considerable effort would be required to reconcile the discrepancies between different sources. It is best that the user be forewarned; therefore, we list a few of the discrepancies which we have found most vexing.

First, of course, is the matter of temperature scales. The NBS-39 scale, the NBS-55 scale, and more recently the IPTS-68 scale have been in common use, and their inter-relation is fairly well understood. For example, table 2-2 is on the NBS-55 scale, and table 2-3 on the NBS-39 scale. The triple point temperatures are 13.803 K and 13.813 K; they differ by 0.01 K. Next is the matter of the triple point. Vapor pressure equations for solid and liquid as well as the melting curve should intersect mathematically at the triple point, yet they rarely do. Finally, different authors choose different reference states for entropy and enthalpy. For example, in table 2-2 and 3-2 the reference enthalpy, $H_o - E_o^o$, is zero for the ideal gas at 1 atm and 0 K, in table 2-3 the reference enthalpy is zero for the ideal crystal at 0 K. The enthalpies of the solid at the triple point compare as follows: table 3-2 -740.2 J/mol, table 2-3 21.32 J/mol.

Major References

We would be remiss if we did not point out those references that have been of particular value to us in this survey. The list is a practical bibliography for those engaged in research on the hydrogens.

1. The survey paper by Woolley, et al., 1948 [6368].
2. The thermodynamic calculations by Mullins, et al., 1961 [12596].
3. The monograph on parahydrogen by Roder, et al., 1965 [29210] and its equivalent in engineering units by McCarty and Weber, 1972 [80777].
4. The papers on solid hydrogen by Dwyer, Cook, et al., ca 1965 [25302, 28604, 29550, 29881, 30327, 34352, 38751].
5. The thermodynamic properties of deuterium by Prydz, 1967 [43781].
6. The Russian survey on hydrogen, Esel'son, et al., 1969 [Esel'son, et al., 1971].
7. The survey on deuterium by Ludtke and Roder, 1971 [78262].

1.1 Recommendations

We have assembled only such data as is available in the literature; the lack of data for specific properties will be evident. We suggest that additional analysis, estimation and measurements should be undertaken to fill some of the gaps uncovered in this survey. In particular, the removal of inconsistencies in the data, touched upon above, would be highly desirable. Specific recommendations, separated into short term and longer term items, are given below:

Short Term Items

1. Maintain a literature file by monitoring the most appropriate journals for new information. For example, the January 1973 issue of Cryogenics contains an article on the mechanical properties of solid H_2 .
2. Additional analysis to resolve inconsistencies.
 - a. Re-evaluate the density of saturated solid H_2 and D_2 by including the most recent values obtained in the determination of the lattice constants. The older volumetric measurements could be off by 1.5%. Estimate solid densities for the other isotopes.
 - b. Calculate the ideal gas thermodynamic properties for HD, HT and DT for temperatures below 50 K.
 - c. Re-analyze the liquid viscosity data especially for D_2 since the estimated values for HT, DT and T_2 depend critically upon the values of D_2 .
 - d. Re-analyze the surface tension data to include the most recent values at temperatures near critical.
 - e. Analyze the difference in Debye θ 's as obtained from specific heats vs. those obtained from mechanical properties.
 - f. Try to establish which mixing rule is the most appropriate, since there is some evidence that the properties of HT cannot be taken to be equal to those of D_2 .
3. Additional estimation to fill critical gaps in the data.
 - a. Estimate property values, not presently available, at the fixed points.
 - b. Estimate densities for saturated liquid HT and DT.
 - c. Estimate thermal diffusivities for all isotopes. Note that the values should vary by about 4 orders of magnitude.
 - d. Estimate heats of sublimation for HT, DT, and T_2 and try to insure consistency at the triple point.
 - e. Estimate C_s for liquid T_2 .
 - f. Estimate densities of the liquid along the melting lines.
 - g. Summarize the information above in temperature-entropy diagrams.

Longer Term Items

1. A more detailed survey of ortho-para conversion, of electromagnetic radiation interaction with the hydrogens, and of the mixture properties is in order. These topics were slighted in this survey through limitations in time and funding.
2. The PVT surface of parahydrogen should be improved by considering a) additional melting pressures close to the triple point, b) index of refraction measurements which yield a better critical point, c) the IPTS-68 scale, and d) recent accurate experimental measurements of the latent heat of vaporization.
3. The PVT surface of normal hydrogen should be correlated.

4. Remeasure the vapor pressures of the hydrogens. Almost all of the older experiments used the vapor pressure as the basic thermometer, and if any of the older information is to be made consistent and useful the temperature scale problem including the triple point discrepancies must be resolved.
5. Measure the thermal conductivity of solid and liquid D_2 , the present uncertainty is at least 20%.
6. Measure the viscosity of liquid D_2 . Values are basic to the estimation of the viscosities of HT, DT and T_2 but have an uncertainty of at least 10%.
7. Measure the heats of fusion for all isotopes except H_2 and D_2 .
8. Examine the possibility of an additional phase transition in solid hydrogen.

2. PVT PROPERTIES

2.1 Crystal Structure

Lattice Parameter

Hydrogen is an extremely interesting solid because for certain isotopic compositions it may undergo a transition at low temperatures. This transition has been studied by x-ray diffraction, electron diffraction, neutron diffraction, nmr [29574], and infrared absorption [30699]. In general all hydrogen isotopes solidify in the hexagonal close packed (hcp) structure, particularly if cooled from the liquid state. Parahydrogen and orthodeuterium appear to be stable in hcp down to the lowest temperatures investigated. In contrast, H_2 rich in the ortho component (> 60%) and deuterium rich in para undergo a transition to the face centered cubic (fcc) structure in the temperature range 1.2 to 4.2 K. In addition, there is considerable evidence that unconverted hydrogen or deuterium if condensed from the vapor in thin films is in fcc [35812]. Other experiments show that the substrate can affect the lattice spacing introducing the fcc structure, and that plastic deformation initiates the fcc structure [38726].

The transition temperature is a function of ortho-para concentration [47572] as well as density or pressure [47821]. The volume change of the transition is small, 1.3%, but has been measured [43353]. The transition corresponds to the λ anomaly in the specific heat but exhibits considerable hysteresis. It is obvious that the difference in energy of the two close packed structures of hydrogen is exceptionally small. It is also well known that solid phases frequently undercool and remain in a metastable form; thus hysteresis is often encountered in such a martensitic transformation [38726]. A different explanation is that the changes in thermodynamic properties are due mainly to the order-disorder transition of the rotational motions, and not to the crystalline phase change per se [65524].

The ranges of the lattice parameters have been assembled from various sources into table 2-1. It should be emphasized, though, that the references given above and in table 2-1 are not exhaustive, only representative.

Table 2-1. Lattice Parameters

species	structure	a	c	c/a	References
p-H_2	hcp	3.75 - 3.761	6.105 - 6.14	1.623 - 1.633	[15709] [38726] [54572]
n-H_2	hcp	3.76 - 3.78	6.105 - 6.16	1.623 - 1.633	[24558] [32872] [43557] [54572]
	fcc	5.312 - 5.338	—	—	[32872] [43557] [54572]
D_2	hcp	3.54 - 3.65	5.83 - 5.91	1.60 - 1.634	[24558] [38726] [43557]
	fcc	5.081 - 5.092	—	—	[37741] [43557] [32807] [53755]
HD	hcp	3.642	5.951	1.634	[43557]
	fcc	5.182	—	—	[43557]
T_2	hcp*	3.53	5.76	1.633	[29055]

* Structure is listed as bct; recalculated assuming ideal hcp structure and molar volumes to be the same.

Thermal Expansion (solid)

Explicit values for any of the hydrogens were not found in the literature. We can, however, estimate the thermal expansion from the volume change. In the absence of preferred orientation the linear expansion is to a very good approximation one-third of the volume expansion. As an example consider deuterium between 1.175 K and its triple point, 18.723 K. The linear thermal expansion over this range is

$$\alpha\Delta T = \frac{\text{length}_{T.P.} - \text{length}_{1.175}}{\text{length}_{T.P.}} = \frac{1}{3} \left(\frac{20.48 - 19.75}{20.48} \right) = 0.012$$

where the molar volumes of deuterium at the appropriate temperatures were taken from table 2-8. Solid deuterium in this temperature range contracts by about 1% in contrast to a typical metal which contracts very little if at all for the same temperature change. The calculated values for parahydrogen shown below were taken from Ahlers [21136].

Temperature K	Thermal expansion coefficient $\alpha \times 10^4$, 1/K	Temperature K	Thermal expansion coefficient $\alpha \times 10^4$, 1/K
1	0.022	8	21.7
2	0.22	9	26.7
3	1.0	10	30.4
4	3.0	11	34.6
5	6.5	12	39.4
6	11.3	13	44.7
7	16.5	13.80	49.4

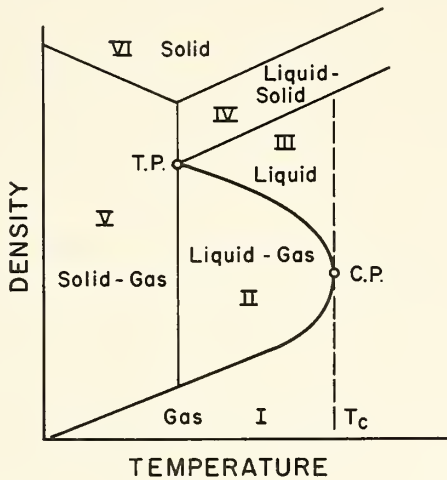
Metallic Hydrogen (solid)

Solid hydrogen under extreme compression - about 2.8 megabars - undergoes a phase transition to the metallic state [82898], and may be superconducting. While this subject is outside the scope of our report, we include a short bibliography. The bibliography is intended to provide a starting point for those interested in pursuing this fascinating subject. A survey of the various equations of state of the solid which have been extrapolated to extreme pressures is given in [41169]. A basic reference is E. Wigner and H. B. Huntington, J. Chem. Phys. 3, 764 (1935). Others are:

[16708]	[55479]	[74178]	[80692]
[31008]	[62125]	[76172]	
[40176]	[65872]	[78271]	
[47933]	[69744]	[80394]	

2.2 PVT Data

A large number of papers deal with PVT data for the hydrogens. In order to organize the material it is helpful to recall the density-temperature phase diagram shown schematically below.



The presentation of data for each isotope is done according to the six regions on the schematic phase diagram. Ordinarily PVT data are not measured in the coexisting regions liquid-gas, liquid-solid, or solid-gas, rather they are established from the phase boundaries in terms of a variable called quality. The phase boundaries are included in the description of the adjacent single phase region. The chart below is intended to serve as a rapid index showing what is available for a given isotope.

Note: Within the limits of this survey it has not been possible to remove discrepancies between the sources shown. For example, the triple point of H_2 is 13.803 and 13.813 depending on the temperature scales used and in addition the enthalpy and entropy bases are different.

Phase modification	Region I Gas	Region II two-phase Vapor/ Liquid	Region III compressed Liquid	Region IV two-phase Liquid/ Solid	Region V two-phase Vapor/ Solid	Region VI compressed Solid
H_2	✓	✓	✓	✓	✓	✓
D_2	✓	✓	✓		✓	✓
T_2		✓				
HD		✓				
HT						
DT						

Hydrogen

The primary sources are Roder, et al. [29210], McCarty and Weber [80777], Malyshenko [81975], Mullins, et al. [12596], Cook, et al. [29881], Dwyer, et al. [29550], Stewart [11238], Megaw [487], and Woolley, et al. [6368].

For parahydrogen Roder, et al. [29210] present extensive tables. The basic measurements cover the entire liquid and gas region up to 350 atm of pressure. The uncertainty is 0.1% in volume except near the critical point where it may be as much as 6%. The temperatures are on the NBS-1955 scale. Values for the saturation boundary table 2-2 are taken from this source to illustrate the type of data presented.

Table 2-2. Saturation Properties, Parahydrogen, Liquid-Vapor

TEMPERATURE DEG. KELVIN	PRESSURE ATM	VOLUME CM ³ /GMOL ^F	$(\partial P/\partial \rho)_T$ ISOTHERM DERIVATIVE CM ³ /ATM/GMOLE	$(\partial P/\partial T)_\rho$ ISOCHORE DERIVATIVE ATM/K	INTERNAL ENERGY J/GMOLE	ENTHALPY J/GMOLE	ENTROPY J/GMOLE-K	C _p , HEAT CAPACITY J/GMOLE-K	C _v , HEAT CAPACITY J/GMOLE-K	VELOCITY OF SOUND METER/SEC
13.803	0.069 ₅	26.17	23328.	9.403	-622.9	-622.7	10.00	9.50	13.13	1273
13.803	0.069 ₅	16056.81	1098.	0.005	169.8	282.8	75.63	12.52	21.20	305
14.	0.077 ₈	26.23	22864.	9.370	-620.3	-620.1	10.19	9.56	13.31	1264
14.	0.077 ₈	14519.41	1110.	0.006	172.0	286.5	74.96	12.52	21.24	307
15.	0.133	26.53	21360.	9.132	-606.5	-606.2	11.14	9.90	14.08	1235
15.	0.133	9049.48	1170.	0.009	182.9	304.6	71.86	12.58	21.51	317
16.	0.213	26.84	19950.	8.919	-592.0	-591.4	12.08	10.26	14.92	1207
16.	0.213	5955.49	1223.	0.014	193.2	321.7	69.15	12.65	21.87	325
17.	0.325	27.18	18327.	8.746	-576.6	-575.7	13.01	10.61	15.92	1175
17.	0.325	4096.06	1267.	0.020	202.9	337.8	66.75	12.74	22.31	333
18.	0.476	27.54	16910.	8.589	-560.2	-558.9	13.95	10.94	16.98	1148
18.	0.476	2920.83	1302.	0.029	211.9	352.7	64.60	12.84	22.86	341
19.	0.673	27.93	15663.	8.423	-542.9	-541.0	14.88	11.24	18.05	1124
19.	0.673	2145.87	1326.	0.040	220.0	366.2	62.64	12.96	23.51	347
20.	0.923	28.36	14525.	8.252	-524.6	-521.9	15.83	11.51	19.15	1102
20.	0.923	1616.07	1340.	0.053	227.2	378.3	60.85	13.08	24.28	353
20.268	1.000	28.48	14091.	8.204	-519.5	-516.6	16.08	11.57	19.53	1093
20.268	1.000	1506.86	1343.	0.057	229.0	381.7	60.41	13.11	24.50	355
21.	1.233	28.82	13123.	8.057	-505.1	-501.5	16.78	11.74	20.49	1072
21.	1.233	1245.28	1344.	0.070	233.5	389.1	59.21	13.20	25.18	358
22.	1.613	29.34	11914.	7.842	-484.5	-479.7	17.74	11.94	21.84	1046
22.	1.613	973.39	1333.	0.090	238.5	397.6	57.64	13.33	26.27	363
23.	2.069	29.90	10507.	7.583	-462.6	-456.3	18.72	12.11	23.51	1012
23.	2.069	771.82	1310.	0.115	242.3	404.2	56.15	13.46	27.59	367
24.	2.611	30.54	9273.	7.291	-439.3	-431.2	19.72	12.25	25.25	980
24.	2.611	619.37	1272.	0.146	244.7	408.5	54.73	13.59	29.22	370
25.	3.245	31.26	8021.	6.965	-414.5	-404.2	20.74	12.37	27.35	943
25.	3.245	501.85	1219.	0.183	245.4	410.4	53.34	13.74	31.28	373
26.	3.982	32.08	6810.	6.591	-387.9	-374.9	21.79	12.49	29.79	903
26.	3.982	409.62	1149.	0.228	244.1	409.4	51.98	13.90	33.98	375
27.	4.829	33.04	5613.	6.182	-359.3	-343.1	22.89	12.61	32.94	858
27.	4.829	336.00	1061.	0.284	240.5	404.9	50.61	14.09	37.62	377
28.	5.794	34.19	4425.	5.736	-328.3	-308.2	24.04	12.74	37.39	807
28.	5.794	276.24	954.	0.354	234.0	396.1	49.21	14.34	42.82	378
29.	6.887	35.59	3339.	5.253	-294.2	-269.3	25.27	12.89	43.64	753
29.	6.887	226.85	823.	0.443	223.7	382.0	47.74	14.67	50.79	378
30.	8.118	37.38	2300.	4.720	-255.9	-225.1	26.61	13.07	54.20	692
30.	8.118	185.17	665.	0.561	208.1	360.4	46.14	15.12	64.50	377
31.	9.501	39.85	1376.	4.118	-211.3	-172.9	28.15	13.33	74.81	623
31.	9.501	148.88	471.	0.727	184.4	327.7	44.30	15.77	93.87	375
32.	11.051	43.83	593.	3.393	-154.4	-105.3	30.08	13.79	134.66	539
32.	11.051	115.21	256.	0.977	145.3	274.3	41.95	16.47	176.84	371
32.976	12.759	64.14		1.874	-5.3	77.6	35.42	19.88		349
32.976	12.759	64.14		1.874	-6.2	76.7	35.40	19.66		351

* THE FIRST ENTRY FOR EACH TEMPERATURE PERTAINS TO THE SATURATED LIQUID

The data of Roder, et al. [29610] were used by McCarty and Weber [80777] to produce similar set of PVT data in engineering units, and to extrapolate the liquid surface to pressures of 10,000 psia (~ 680 atm). An alternative extrapolation is found in a paper by Alyshenko [81975]. This paper is mentioned because the extrapolation goes to 1000 atmospheres and is the only source of PVT for liquid hydrogen at pressures to 1000 atmospheres.

The calculations of Mullins, et al. [12596] presents PVT data for parahydrogen below the triple point temperature down to 1 K. These data are based on available experimental values and a thermodynamically consistent calculation.

A composite table of values from this source is presented in table 2-3.

The PVT surface of solid hydrogen is best summarized in figure 2-1 taken from Cook, et al. [28604]. The authors include their own measurements on the compressed solid (table 4), values on the saturated solid by Dwyer, et al. [29550, table 2-5] as well as the changes in volume measured by Stewart [11238, shown separately in figure 2-2 and in table 2-6] which are in agreement with the earlier measurements by Megaw [487]. Figure 2-1 shows rather unusual behavior near the melting line indicating perhaps an as yet undisclosed phase transition [see also Roder, 1973].

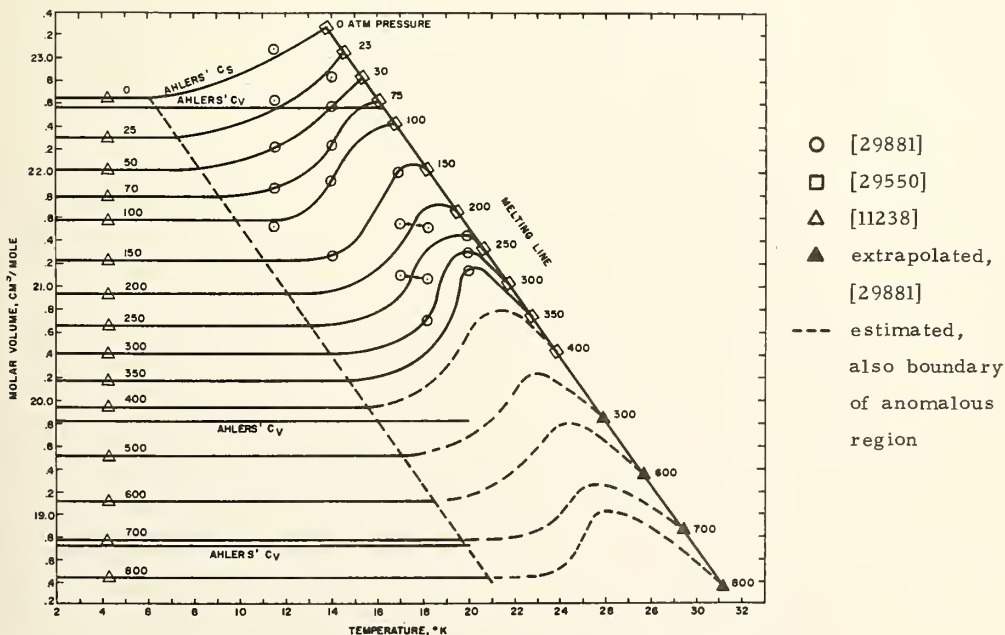


Figure 2-1. PVT Diagram for Solid Hydrogen

Table 2-3. Saturation Properties, Parahydrogen, Solid-Vapor

Temperature K	Pressure mm HG	Enthalpy of the Solid Cal/mol	Enthalpy of Saturated Vapor Cal/mol	Heat of Sublimation Cal/mol	Compress- ibility of the Vapor Z	Solid Heat Capacity Cal/mol K	Entropy of the Solid Cal/mol K	Entropy of the Vapor Cal/mol K	Molar Volume of the Solid*
13.813	52.89	5.096	250.00	244.90	0.98436	1.360	0.4940	18.224	23.211
13.000	30.13	4.054	246.35	242.30	0.98974	1.164	0.4175	19.056	23.125
12.000	13.78	2.990	241.70	238.71	0.99434	0.9438	0.3334	20.226	23.031
11.000	5.567	2.141	236.90	234.76	0.99719	0.7471	0.2602	21.602	22.947
10.000	1.917	1.479	232.03	230.55	0.99878	0.5751	0.1975	23.252	22.873
9.000	5.343×10^{-1}	0.9788	227.10	226.12	0.99956	0.4280	0.1450	25.269	22.809
8.000	1.118×10^{-1}	0.6139	222.15	221.53		0.3054	0.1022	27.794	22.754
7.000	1.561×10^{-2}	0.3598	217.18	216.82		0.2065	0.06842	31.043	22.711
6.000	1.198×10^{-3}	0.1933	212.21	212.02		0.1301	0.04291	35.379	22.678
5.000	3.570×10^{-5}	0.09275	207.24	207.15		0.07420	0.02473	41.455	22.660
4.000	2.080×10^{-7}	0.03799	202.28	202.24		0.03799	0.01266	50.572	22.650
3.000	4.832×10^{-11}	0.01202	197.31	197.30		0.01603	0.005342	65.771	22.646
2.000	3.985×10^{-18}	0.002374	192.34	192.34		0.004749	0.001583	96.170	22.645
1.000	8.255×10^{-39}	0.0001484	187.37	187.37		0.0005936	0.0001979	187.372	22.645

* from Ahlers [21136], Mullins, et al. set this value constant

Table 2-4. Parahydrogen, Molar

Volume of the Solid

Pressure (atm)	Temperature (°K)				
	11.49°	13.98°	16.89°	18.20°	19.97°
0	23.08				
25	22.63	22.83			
50	22.22	22.58			
75	21.85	22.24			
100	21.53	21.93			
150		21.26	22.00		
200			21.55	21.52	
250			21.10	21.07	21.45
300				20.70	21.30
350					21.15

* All values of the molar volume are in cubic centimeters per mole.

with permission from G.A. Cook, et al., J. Chem. Phys. 43, 1313 (Aug. 1965).

Table 2-5. Parahydrogen, Molar Volumes

of the Solid Along the Melting Line

Temp. (°K)	Pressure (atm)	Molar volume (cm ³ /mole)		Difference Exptl. --Calc.
		Exptl.	Calc.	
13.803	0.0695	23.31	23.272	+0.038
13.97	...	23.16*	23.225	-0.065
13.99	...	23.22*	23.219	+0.001
14.00	5.887	23.26	23.216	+0.044
15.00	36.88	22.97	22.933	+0.037
16.00	70.19	22.67	22.650	+0.020
16.89	...	22.42*	22.398	+0.022
17.00	105.68	22.36	22.367	-0.007
18.00	143.22	22.06	22.084	-0.024
18.20	...	22.00*	22.027	-0.027
19.00	182.72	21.76	21.801	-0.041
19.97	...	21.52	21.526	-0.006
20.00	224.10	21.48	21.518	-0.038
21.00	267.29	21.20	21.235	-0.035
22.00	312.23	20.94	20.952	-0.012
23.00	358.88	20.69	20.669	+0.021
23.82	...	21.51*	20.437	+0.073
24.00	407.20	20.45	20.386	+0.064

Mean 0.032

* Values marked with an asterisk were measured directly in the piezometer. All other molar volumes were computed from our smoothed values for heat of fusion. All pressures were calculated from the equation derived by Goodwin (Ref. 7). Calculated values of the molar volume were obtained by use of Eq. (3).

with permission from R.F. Dwyer, et al., J. Chem. Phys. 43, 801 (Aug. 1965).

Table 2-6. Compressions at 4 K

Pressure kg/ cm ²	HELIUM				HYDROGEN				DEUTERIUM				NEON			
	$\frac{\Delta V'}{V_0}$	$\frac{V}{V_0}$	M.V. cm ³	$10^5 \beta_i$ cm ² / kg	$\frac{\Delta V'}{V_0}$	$\frac{V}{V_0}$	M.V. cm ³	$10^5 \beta_i$ cm ² / kg	$\frac{\Delta V'}{V_0}$	$\frac{V}{V_0}$	M.V. cm ³	$10^5 \beta_i$ cm ² / kg	$\frac{\Delta V'}{V_0}$	$\frac{V}{V_0}$	M.V. cm ³	$10^5 \beta_i$ cm ² / kg
P_0 or 0	—	1.000	17.1	115	—	1.000	22.65	49	—	1.000	19.56	30	—	1.000	14.0	10
200	—	.924	15.8	77	—	.928	21.0	31	—	.948	18.5	22	—	.982	13.7	—
400	—	.846	14.5	41	—	.883	20.0	23	—	.911	17.8	18	—	.966	13.5	—
600	—	.792	13.5	29	—	.847	19.2	19	—	.882	17.2	15	—	.952	13.3	—
1000	—	.722	12.4	19	—	.794	18.0	14	—	.840	16.4	12	—	.927	13.0	5.3
2043	0	.625	10.7	9.8	0	.711	16.1	8.1	0	.764	14.9	7.3	0	.882	12.3	3.8
3000	.079	.579	9.9	7.0	.065	.667	15.1	6.1	.061	.715	14.0	5.5	.032	.858	12.0	—
4000	.134	.544	9.3	5.5	.110	.632	14.3	4.8	.103	.680	13.3	4.5	.055	.832	11.6	2.6
6000	.205	.497	8.5	3.8	.181	.583	13.2	3.5	.173	.632	12.4	3.3	.100	.797	11.1	2.0
8000	.256	.466	8.0	2.9	.227	.549	12.4	2.7	.215	.597	11.7	2.6	.130	.770	10.8	1.7
10,000	.295	.440	7.5	2.4	.266	.523	11.8	2.3	.253	.571	11.2	2.2	.156	.747	10.4	1.41
12,000	.325	.421	7.2	2.0	.297	.500	11.3	1.9	.281	.549	10.7	1.9	.178	.728	10.2	1.23
16,000	.370	.392	6.7	1.5	.343	.467	10.6	1.4	.325	.514	10.0	1.4	.213	.696	9.7	.98
20,000	.403	.373	6.4	1.3	.376	.445	10.1	1.2	.358	.485	9.5	1.2	.240	.669	9.4	.81

with permission from J.W. Stewart, Phys. Chem. Solids 1, 146 (1956).

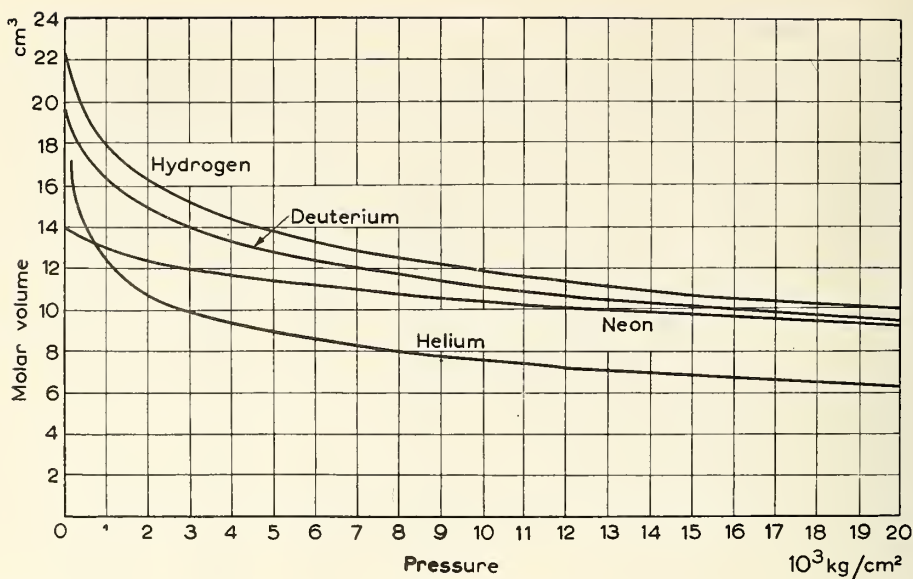


Figure 2-2. The Molar Volumes of Helium, Hydrogen, Deuterium, and Neon at 4 K with permission from J. W. Stewart, Phys. Chem. Solids 1, 146 (1956).

The difference in PVT between the ortho-para modifications is very slight and for most practical purposes may be neglected. There is, however, a difference and if a distinction is necessary, the work of Woolley, et al. [6368] is recommended (see also the section on ortho-para conversion). The vapor pressure of normal hydrogen is slightly different than parahydrogen and therefore the saturated densities will be slightly different. Once off of the saturated boundary, however, the differences are probably less. The differences are illustrated for the saturation boundary in table 2-2a which is taken from [23790].

Deuterium

The recommended source of PVT for liquid and gaseous normal deuterium is the correlation by Prydz [43781] accomplished with a modified BWR equation of state. The correlation covers the temperature range from the triple point to the critical point for pressures to 400 atm. Extensive thermodynamic tables of values are presented in metric units. This source is recommended because it is the only source which covers both the gaseous and liquid phases in a consistent manner. The accuracy of the PVT data in the gaseous phase is probably not too good. Very little experimental data were available and the correlated values deviated from the experimental data by $\pm 0.3\%$ in density at 20.4 K to $+4\%$ in pressure at 41 K. The maximum pressure of the liquid data was slightly over 100 atmospheres. The deviations between Prydz's correlation and experimental values for compressed liquid states range up to $\pm 0.5\%$ in density. Table 2-7 gives saturation data for normal deuterium.

Table 2-2a. Saturation Properties, Normal Hydrogen, Liquid-Vapor

Temp. °K	Pressure atm [31]	Vapor Pressure Slope, dP/dT (atm/°K)	Density, g mole/cm ³	
			Sat. Liquid [12]	Sat. Vapor
13.947	0.071 ₁	0.041 ₁	0.03830	0.000063 ₁
14	0.073 ₂	0.042 ₁	0.03828	0.000064 ₂
15	0.125 ₇	0.063 ₇	0.03786	0.000104 ₅
16	0.202 ₇	0.091 ₄	0.03742	0.000159 ₆
17	0.310 ₈	0.125 ₉	0.03695	0.000232 ₉
18	0.456 ₈	0.167 ₄	0.03647	0.000327 ₈
19	0.648 ₈	0.216	0.03595	0.000447 ₅
20	0.891 ₆	0.273	0.03540	0.000595 ₈
20.380	1.000	0.296	0.03519	0.000660 ₆
21	1.196	0.337	0.03483	0.000776 ₉
22	1.569	0.409	0.03421	0.000995 ₄
23	2.018	0.490	0.03355	0.001257
24	2.551	0.579	0.03285	0.001569
25	3.178	0.676	0.03209	0.001938
26	3.906	0.783	0.03127	0.002377
27	4.746	0.898	0.03036	0.002900
28	5.705	1.023	0.02935	0.003527
29	6.794	1.157	0.02821	0.004290
30	8.023	1.302	0.02689	0.005241
31	9.401	1.457	0.02528	0.006482
32	10.94	1.622	0.02312	
33	12.65	1.800	0.01903	
33.18	12.98	1.833	0.01494	0.01494

Table 2-7. Saturation Data for Normal Deuterium

TEMP. K	PRESSURE ATM	DENSITY MOL/LITER		ENTHALPY J/MOL		ENTROPY J/MOL-K	
		VAPOR	LIQUID	VAPOR	LIQUID	VAPOR	LIQUID
18.710	0.16872	0.111221	43.1598	638.7	-648.9	87.09	18.27
19.000	0.19181	0.124876	43.0160	640.8	-644.7	86.52	18.86
20.000	0.29057	0.181841	42.5081	660.4	-615.6	83.81	20.02
21.000	0.42405	0.256456	41.9781	675.1	-587.7	81.48	21.35
22.000	0.59925	0.352051	41.4228	687.2	-558.9	79.30	22.66
23.000	0.82353	0.472322	40.8403	696.7	-529.2	77.25	23.95
24.000	1.10456	0.621418	40.2287	703.3	-498.8	75.31	25.22
25.000	1.45018	0.804045	39.5858	707.0	-467.7	73.44	26.46
26.000	1.86843	1.02561	38.9085	707.7	-436.0	71.65	27.66
27.000	2.36751	1.29242	38.1926	705.4	-403.6	69.91	28.84
28.000	2.95576	1.61194	37.4326	700.1	-370.5	68.22	29.98
29.000	3.64163	1.99316	36.6212	691.5	-336.6	66.56	31.10
30.000	4.43377	2.44712	35.7488	679.6	-301.9	64.93	32.21
31.000	5.34100	2.98769	34.8025	664.5	-265.6	63.31	33.31
32.000	6.37235	3.63281	33.7643	645.8	-227.6	61.71	34.42
33.000	7.53713	4.40669	32.6085	623.2	-187.1	60.11	35.56
34.000	8.84496	5.34403	31.2963	596.2	-142.9	58.49	36.75
35.000	10.30579	6.49920	29.7650	563.9	-93.3	56.83	38.05
36.000	11.93005	7.97036	27.9000	524.2	-34.7	55.07	39.54
37.000	13.72861	9.98197	25.4469	471.7	40.4	53.06	41.41
38.000	15.71290	13.4367	21.4462	382.7	160.0	50.23	44.37
38.340	16.43200	17.3280	17.3280	278.6	278.6	47.38	47.38

Woolley, et al. [6368] present molar volumes for the solid at the triple point and at elevated pressures along the melting line. The latter values are based on volumetric measurements in compressed liquid states by Bartholome [5118] extrapolated to the appropriate phase boundary and combined with the calculated volume changes on fusion published by Clusius and Bartholome [5595]. The volume changes on fusion were established from measured heats of fusion, the slope of the melting curve, and the Clausius-Clapeyron equation.

The densities of saturated solid deuterium were approximated by Ludtke and Roder [78262] with the power series

$$\rho = \rho_0 - AT^2 = 0.20459 - 0.000026337T^2 \text{ with } \rho \text{ in g/cm}^3 \text{ and } T \text{ in kelvins.}$$

The polynomial is based on the data by Schuch and Mills [37741], Megaw [487], Mucker, et al. [32807] and Woolley, et al. [6368]. The comparison of calculated and experimental values is shown in table 2-8.

Table 2-8. Densities of Saturated Solid Deuterium

T (K)	V (cm ³ /mol)	V-CALC (cm ³ /mol)	ρ g/cm ³	ρ -CALC g/cm ³	DIFF (%)	AUTHOR	MODIFI- CATION
1.175	19.75	19.69	0.2040	0.2045	-0.3	Schuch and Mills	various
1.450	19.79	19.70	0.2036	0.2045	-0.5	Schuch and Mills	para
4.200	19.56	19.74	0.2059	0.2041	0.9	Megaw	normal
13.000	20.10	20.13	0.2004	0.2001	0.1	Muckers, et al.	normal = ortho
17.000	20.65	20.45	0.1951	0.1970	-1.0	Schuch and Mills	normal
18.723	20.48	20.62	0.1967	0.1954	0.7	Woolley, et al.	normal

For the compressed solid states of deuterium there are two sources of data, Megaw [487] and Stewart [11238]. The two sources agree very closely, and Stewart's values are shown in figure 2-2 and table 2-6.

The differences in PVT between ortho-para modifications of deuterium are small and for most practical purposes may be neglected. We note that for hydrogen the para and normal modifications differ by up to 0.26% in liquid density, see for example Woolley, et al. [6368] or Goodwin, et al. [11625]. We can apply a simple ratio of 0.333/0.75 to get a change of 0.115% for the difference between normal and ortho deuterium. This change is smaller than the precision inherent in the data treated by Prydz.

Tritium

The only experimental PVT data for T_2 revealed in the literature search are densities for the saturated liquid and gas for temperatures from 20.61 to 29.13 K by Grilly [445]. The values are given in table 2-9 below. The densities of the liquid should be in error by no more than 0.23%. Additional values for the density of the liquid given in table 2-10 were obtained by Rogers and Brickwedde [28408] from corresponding states.

Table 2-9. Molar Densities of Liquid

Tritium				
T, °K.	$\rho_{liq} - \rho_{gas}^a$ mole/liter	ρ_{gas}	ρ_{liq}^b	Dev. from curve
20.61	45.17	0.13	45.39	0.04
22.50	43.97	.16	44.21	-.01
22.99	43.51	.29	43.88	-.04
23.59	43.11	.35	43.55	.00
24.41	42.49	.44	43.01	-.01
24.72	42.24	.48	42.81	-.02
25.66	41.53	.62	42.25	.03
26.36	40.91	.73	41.74	.00
27.09	40.17	.87	41.14	-.05
28.32	39.06	1.14	40.31	.07
29.13	38.02	1.37	39.49	-.06

^a With 2.0% HT present. ^b Corrected to 0.0% HT.

with permission from E.R. Grilly, J. Am. Chem. Soc. 73, 5307 (1951).

Table 2-10. Density of Liquid Tritium

T, K	ρ , g/cm ³
30.0	0.234
31.0	0.228
32.0	0.222
33.0	0.216
34.0	0.209
35.0	0.202
36.0	0.193
37.0	0.183
38.0	0.171
38.5	0.164
39.0	0.155
39.5	0.144
40.0 ^{+ 1.2} - 2.2	Critical Point 0.109 ^{+ 0.003} - 0.003

with permission from J.D. Rogers, et al., J. Chem. Phys. 42, 2822 (Apr. 1965).

The only reference to the density of solid tritium is that of Kogan, et al. [29055]. They report a density of 0.324 g/cm³ at 4.2 K.

Hydrogen Deuteride - HD

A paper by Varekamp and Beenakker [12725] gives second virial coefficients for HD, see page 20. These values may be used with the virial equation to calculate pressures for densities below saturation.

The only PVT data for liquid HD revealed by the literature search are densities for the saturated liquid published by Rudenko and Slyusor [59955]. Table 2-11 is taken from this reference. Densities given in the earlier paper by Woolley, et al. [6368] are about 0.4% higher than those of Rudenko and Slyusor and should be ignored.

Table 2-11. Densities of Liquid HD

T, K	ρ , g/cm ³	T, K	ρ , g/cm ³
16.60	0.1228	28.00	0.1023
17.00	0.1223	29.00	0.0997
18.00	0.1209	30.00	0.0968
19.00	0.1195	31.00	0.0936
20.00	0.1180	32.00	0.0900
20.39	0.1173	32.50	0.0880
21.00	0.1164	33.00	0.0858
22.00	0.1147	33.50	0.0834
23.00	0.1130	34.00	0.0805
24.00	0.1111	34.50	0.0772
25.00	0.1091	35.00	0.0730
26.00	0.1070	35.50	0.0671
27.00	0.1048		

Woolley, et al. [6368] report the volume of solid HD at the triple point to be 21.84 cm³/mol at $p = 0.126$ kg/cm² and $T = 16.60$ K.

Note that HD does not exhibit ortho-para modifications.

Hydrogen Tritide - HT

No references were found for the PVT of HT, however, the assumption is usually made that the PVT for HT is approximately equal to that of D₂.

Deuterium Tritide - DT

No references for the PVT of DT were found for the temperature range of this survey, however, Oppenheim and Friedman [23817] give some theoretically calculated values of PVT for DT at temperatures slightly above the critical temperature.

Discussion

The existing PVT data for parahydrogen are adequate, but additional experimental data for all the other isotopes would be desirable.

2.3 Equations of State

The meaning of the phrase "equation of state" is taken here to be a mathematical model of the PVT surface. Usually the model consists of a single equation which is explicit in pressure. This is most convenient because the various derivatives, which are required to calculate thermodynamic functions, can be obtained in closed form. At times the models are not explicit in any variable, and occasionally a single equation does not exist at all, instead a computer program is substituted for the equation of state. In this last case the PVT surface is often broken into regions with a different formulation in each region.

Hydrogen

The PVT surface presented by Roder, et al. [29210] covers both gaseous and liquid regions from the triple point to the critical point. The surface is described by a collection of equations each representing an isotherm or an isochore with interpolation for intermediate points. The formulation is not easy to use but is recommended as the best surface available.

A virial representation of the gaseous PVT surface of hydrogen by Mullins, et al. [12596] extends the equation of state to lower temperatures. Mullins, et al. [12596] are reasonably consistent with Roder, et al. [29210] in the region of overlap. Discontinuities do, however, occur and have been mentioned in section 2.2.

Two other equations of state are mentioned here because they may be preferred for particular applications. One is Goodwin's equation of state [63091] which gives qualitatively correct behavior in the critical region, i. e., "scaling-law" behavior. The other is an early adaptation of the Benedict-Webb-Rubin equation of state to hydrogen by Roder and Goodwin [12540] using 17 coefficients. Analytical equations of state such as the BWR cannot have "correct" behavior in the critical region. They are, however, much easier to use. Roder and Goodwin [12540] present two sets of coefficients, one to be used for the gas, and one for the liquid. They estimate a maximum error in density of 0.35% except in the critical region. Both of these equations are valid for the PVT surface between the triple point temperature and the critical point temperature.

An extension of the liquid parahydrogen data to higher pressures was performed by McCarty and Weber [80777]. The authors do not give an explicit equation, however, the PVT surface is represented through a computer program in various regions. The extrapolation to higher pressures by Malysenko [81975] is based on the Tait equation for which the author gives values of the coefficients.

A review of the properties of solid hydrogen, including the so-called "metallic hydrogen" is given by Douglas and Beckett [41169]. Numerous equations of state are presented, most of which are based on the intermolecular potential. Another good review of the properties of solid hydrogen is given by Stewart [31446], while Ahlers [21136] calculates

some of the derived properties such as compressibility from the Grüneisen equation. The equation proposed by Burshtein and Rabinovich [79779] while "anchored" with experimental densities along the solid melting line is not satisfactory because it does not yield the proper densities of the solid under its own vapor pressure. Therefore, a unique equation of state from which one could calculate the PVT properties at an arbitrary point in the solid does not exist.

Deuterium

The recommended source for PVT data of normal deuterium is the paper by Prydz [43781]. The PVT surface is represented by Prydz with a BWR type equation of state, in this instance using 28 coefficients. As with most BWR equations, a program is available and is fairly easy to use. The PVT surface presented by Prydz covers both the gaseous and liquid regions from the triple point to the critical point with pressures to about 100 atmospheres in the liquid region. Deviation plots show a maximum deviation of $\pm 0.4\%$ in density between calculated and experimental values.

Other Isotopes

No practical equations of state for the other isotopes were found in the literature. Some second virial coefficients have been published, and some theoretical work has been done. The theoretical results are of a qualitative nature, see for example Oppenheim and Friedman [23817] and Henderson [13537].

Discussion

From the lack of references for the equation of state of isotopes other than H_2 and D_2 , it is evident that a lot of work needs yet to be done. However, until experimental PVT data become available, empirical equations of state such as we have recommended for H_2 and D_2 will not be possible. Theoretical predictive methods are limited in scope. Application of the theory of corresponding states to the existing equations of state for hydrogen, while possible, have not been satisfactory for the combination hydrogen-deuterium.

2.4 Ideal Gas Properties

The thermodynamic functions for the ideal gas are the starting point in the calculation of the thermodynamic properties of the hydrogens. The most complete tabulations are given by Haar, et al. [11271]. Note, however, that these tables do not include nuclear spin. In other words numerical differences exist in the values of entropy and free energy between these tables and earlier sources such as Woolley, et al. [6368] who include nuclear spin. Use of these tables implies that the reference state is the ideal gas at 0 K, with the energy E_0^0 taken to be zero, while the entropies are tabulated for an assumed pressure of 1 atm. To

give a feel for the differences between isotopes, the dimensionless ideal gas enthalpies $(H_0 - E_0^\circ)/RT$ are compared in table 2-12 at 10 and 30 K.

Table 2-12. Comparison of Dimensionless Ideal Gas Enthalpies, $(H_0 - E_0^\circ)/RT$

Species	Temperature, K	
	10	30
n-H ₂	15.28	6.76
e-H ₂	2.500	2.669
p-H ₂	2.500	2.500
n-D ₂	5.365	3.460
e-D ₂	2.502	2.732
o-D ₂	2.500	2.508
n-T ₂	6.819	3.963
e-T ₂	2.658	3.624
p-T ₂	2.500	2.589
HD		
HT	not tabulated below 50 K	
DT		

An older source for the ideal gas thermodynamic functions of T₂ and HT needs to be mentioned here because it contains values below 50 K, and includes graphs on ortho-para distribution as well as graphs of the rotational heat capacity at constant volume. Be it noted, however, that the calculations of Jones [18037] deviate by about 3% in C_p° at 50 K for T₂ from those of Haar, et al. [11271], and by about 1% in C_p° at 50 K for HT.

2.5 Compressibility

Tables of compressibility have been published only for hydrogen. These include the measurements by Udovidchenko and Manzhelii [68779] on solid parahydrogen which are reproduced in table 2-13. The measurements on normal hydrogen by Stewart [11238] and Megaw [487] have already been mentioned with values given in table 2-6. For liquid or gaseous parahydrogen the isothermal compressibility can be formed from the proper combination of isotherm derivative and volume, see for example table 2-2. The isothermal bulk modulus is given directly for parahydrogen in the tables prepared by McCarty and Weber [80777].

Table 2-13. Isothermal Compressibility of Solid

		Parahydrogen $\chi_T \times 10^5$, cm ² /kg			
P , kg cm ⁻²	T , °K	14.5	13.5	12.0	10.0; 8.0; 6.0
	0			59.8 ^a	57.1 ^a
20			54.4 ± 1.4	52.3 ± 1.3	49.4 ± 1.6
40	53.1		49.9	48.2	45.8
60	48.8		46.2	44.7	42.8
80	45.0		42.9	41.8	40.2
100	41.8		40.2	39.3	38.0
120	39.1		37.9	37.2	36.2
140	36.7		36.0	35.4	34.8
180	33.0 ± 0.7	32.9 ± 0.7	32.5 ± 0.8	32.3 ± 1.2	
200					31.2 ^a

^aObtained by extrapolation.

with permission from B.G. Udovichenko, et al. J. Low Temp. Phys. 3, 429 (Oct. 1970).

2.6 Virial Coefficients

The virial coefficients at low temperatures have been measured by Varekamp and Beenakker [12725] for normal hydrogen, HD and normal deuterium. The authors experimental results are reproduced in figure 2-3. At temperatures higher than 20 K the virial coefficients for parahydrogen, normal hydrogen, and normal deuterium are collected from various sources in table 2-14.

3. Derived Thermodynamic Properties

3.1 Enthalpy, Entropy, and Internal Energy

Extended tables of derived thermodynamic properties for the hydrogens are rare and appear to be limited to liquid or gaseous states. Consistent tables for at least a part of the phase diagram exist only for parahydrogen, normal hydrogen, and normal deuterium. A number of theoretical papers touch upon entropy, internal energy, or free energy. However, actual values are seldom presented, and if presented only a limited set of conditions is considered. Henderson [13537], for example, calculates several values of entropy for liquid states of HD and ortho deuterium which are the only values for these isomers found during this survey.

Parahydrogen

The coverage of parahydrogen is the most complete of all isomers. It is the only isomer for which values in the solid state can be obtained primarily because the specific heats were measured by Ahlers [21136]. Enthalpy estimates for the solid are presented in tables 3-1, 3-2, and 3-3. The tables were assembled from the reports of Dwyer and

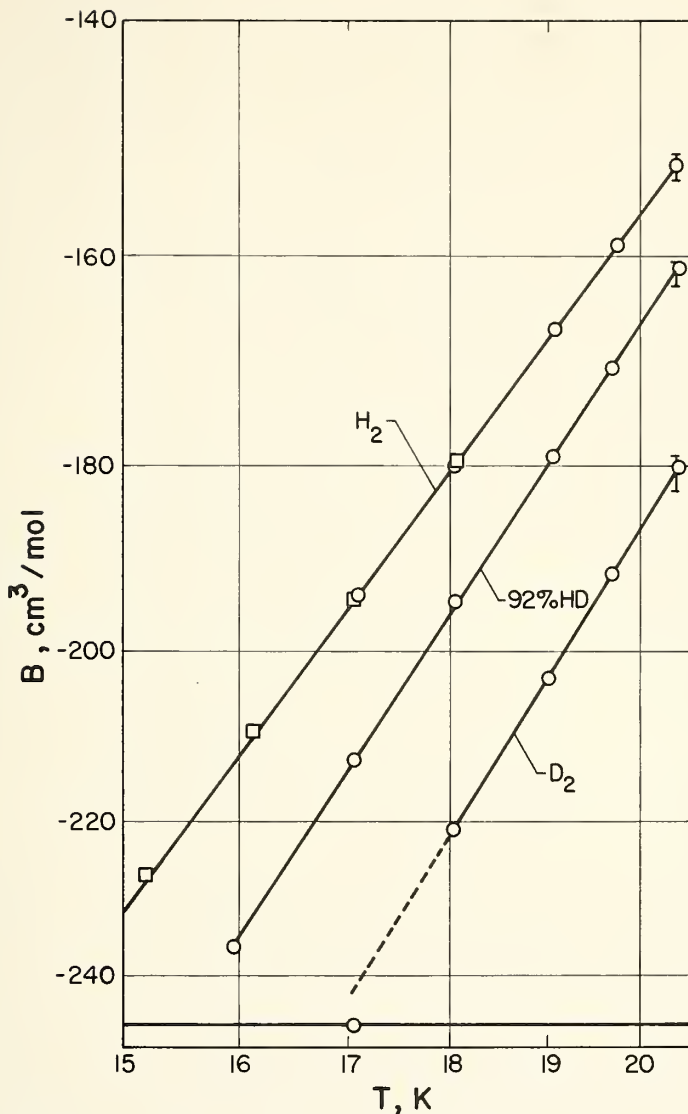


Figure 2-3. Experimental Virial Coefficients for the Pure Isotopes

Cook [28604, 34352, 25302]. Molar volumes and specific heats are included to round out the tables. Values for enthalpy and entropy of the solid in equilibrium with the vapor have already been introduced in table 2-3. Note, however, that the enthalpy bases differ. Tables for liquid and gas are even more complete, they include enthalpy, entropy, internal energy, and several PVT derivatives as shown in sample table 2-2. The equivalent tables (except for units) by McCarty and Weber [80777] include several additional derived properties such as specific heat input, the energy derivative, the isothermal bulk modulus, and volume expansivity.

Table 2-14. Virial Coefficients for the Hydrogens

Para Hydrogen [20547]			Normal Hydrogen [34613]			Normal Deuterium [43781]		
T, K	B cm ³ /mol	C (cm ³ /mol) ²	T, K	B cm ³ /mol	C (cm ³ /mol) ²	T, K	B cm ³ /mol	C (cm ³ /mol) ²
16	-204.2							
18	-172.9							
20	-148.8							
22	-129.7		20.59	-140.980				
24	-112.8	1207.	22.58	-122.630				
25	-106.2	1402.	24.65	-108.020		25	-150.125	-31.
26	-100.3	1580.	26.75	-95.995		30	-118.396	4490.
27	-94.8	1627.	28.83	-90.275	2200.0	35	-90.059	3694.
28	-89.66	1612.	30.86	-80.734	2078.0	40	-69.029	2555.
29	-85.03	1615.	33.00	-73.439	2050.0	45	-53.717	1740.
30	-80.73	1600.	35.10	-64.300	1700.0			
31	-76.75	1585.						
32	-72.99	1550.						
33	-69.53	1516.						

Table 3-1. Parahydrogen Properties - Saturated Solid

Temperature K	Pressure mm Hg	Volume* cm ³ /mol	C _s mJ/mol-K	est. C _p mJ/mol-K	Enthalpy [†] J/mol
0.00		22.645	0.0	0.00	-758.0
1.00	8.3 × 10 ⁻⁸⁸	22.645	1.03	1.03	-758.0
2.00	4.0 × 10 ⁻¹⁸	22.645	8.57	8.57	-758.0
3.00	4.8 × 10 ⁻¹¹	22.646	30.7	30.7	-758.0
4.00	2.1 × 10 ⁻⁷	22.650	78.7	78.7	-757.9
5.00	3.6 × 10 ⁻⁶	22.660	168.5	168.5	-758.8
6.00	1.2 × 10 ⁻³	22.678	318.8	318.8	-757.6
7.00	1.6 × 10 ⁻²	22.711	551	551	-757.2
8.00	1.1 × 10 ⁻¹	22.754	882	882	-756.4
9.00	5.3 × 10 ⁻¹	22.809	1333	1333	-755.3
10.00	1.97	22.873	1917	1917	-753.7
11.00	5.68	22.947	2651	2652	-751.5
12.00	14.0	23.031	3548	3550	-748.4
13.00	30.4	23.125	4618	4622	-744.3
13.803	52.8	23.211	5640	5646	-740.2

* This column from Ahlers [21136].

† This column changed by -0.9 J/mol to bring it into agreement with Roder, et al. [29210].

Normal Hydrogen

The very convenient tables prepared by Dean [6560] include enthalpy, entropy, and internal energy. The tables are based on the correlation of Woolley, et al. [6368] and are limited to conditions of the vapor below saturation pressures.

Normal Deuterium

The tables presented by Prydz [43781] include enthalpy, entropy, and internal energy for both liquid and vapor. The tables are based on an equation of state, the coefficients of which are derived by considering all available PVT data as input, see table 2-7.

3.2 Thermodynamic Diagrams

Thermodynamic diagrams or Mollier charts are used by the engineer in many applications because these charts summarize in one place the phase diagram, the PVT surface, and some of the derived thermodynamic properties. A temperature-entropy chart, for example, contains the phase boundaries, lines of constant pressure, volume, enthalpy, and quality. Table 3-4 shows the charts available, most of them for parahydrogen, while the most appropriate one of the triple point region is reproduced in figure 3-1.

Table 3-2. Parahydrogen Properties Along the Melting Line

Temperature K	Pressure, ** atm	Molar volume ** of the liquid cm ³ /mol	Molar volume ** of the solid cm ³ /mol	Enthalpy of the liquid, J/mol	Heat of fusion, J/mol	Enthalpy of the solid, J/mol
13.803	0.0695	26.176	23.31	-622.9 [†]	-117.3	-740.2 [†]
14.0	5.89	26.062	23.26	-606.9	-118.4	-725.3
15.0	36.88	25.513	22.97	-523.9	-124.1	-648.0
16.0	70.19	25.006	22.67	-436.2	-130.3	-566.5
17.0	105.7	24.540	22.36	-344.7	-136.9	-481.6
18.0	143.2	24.103	22.06	-249.6	-143.8	-393.4
19.0	182.7	23.702	21.76	-151.3	-151.1	-302.4
20.0	224.1	23.328	21.48	-50.1	-158.7	-208.8
21.0	267.3	22.978	21.20	+54.3	-166.7	-112.4
22.0	312.2	22.654	20.94	+161.1	-175.0	-13.9
23.0	358.9	22.351	20.69	+272*	-183.6	+88.4*
24.0	407.2	22.065	20.45	+387*	-192.5	+194.5*

* Extrapolated.

** From Roder, et al. [29210].

† These entries changed by -0.9 J/mol to bring them into agreement with Roder, et al. [29210].

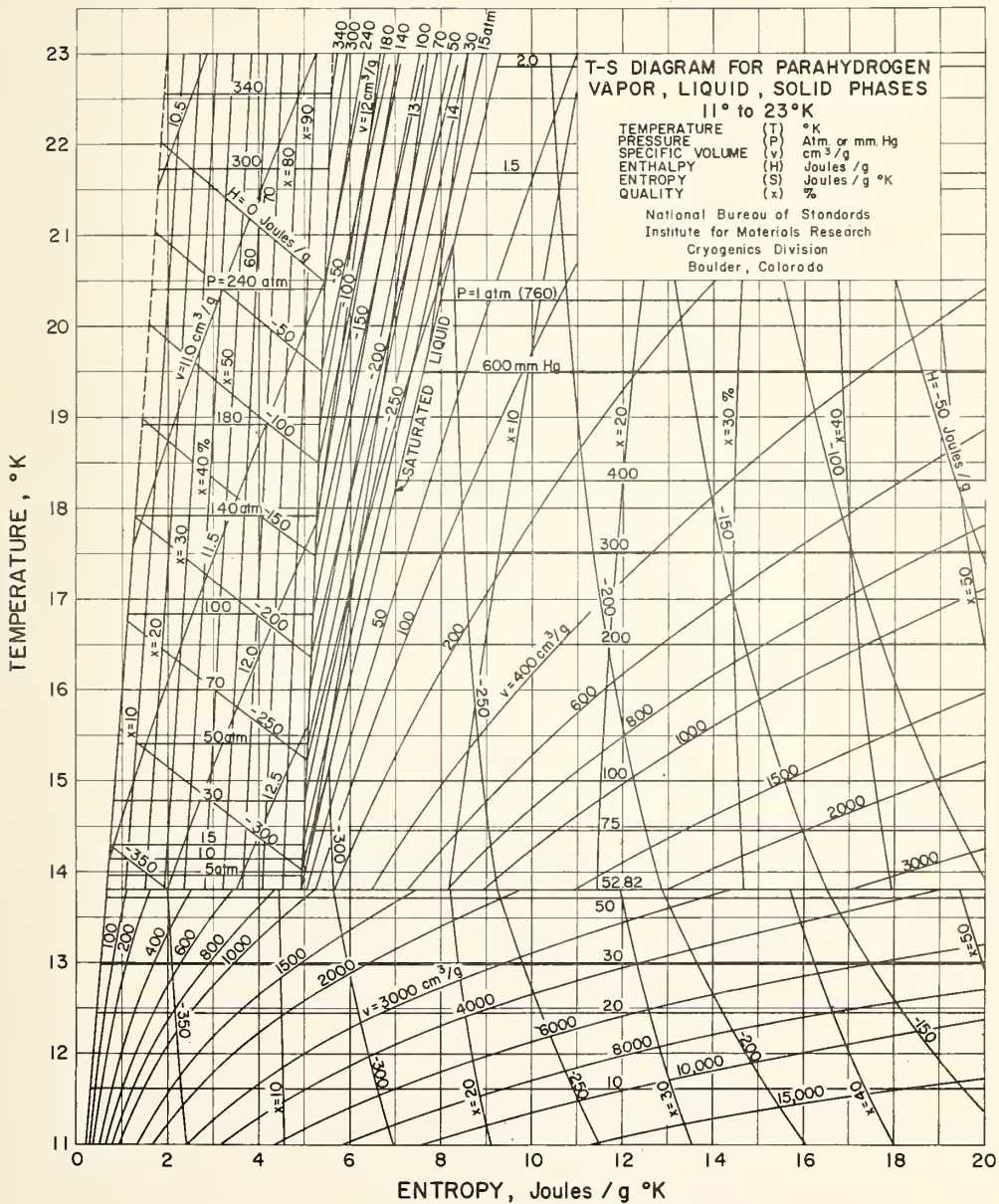


Figure 3-1. Parahydrogen, Temperature-Entropy Diagram

Table 3.3. Parahydrogen Properties - Compressed Solid States

Temperature K	Ahlers' 22.56 cm ³ /mol isochore			Ahlers' 19.83 cm ³ /mol isochore			Ahlers' 18.73 cm ³ /mol isochore			
	est. pressure atm	C _v mJ/mol-K	est. C _p mJ/mol-K	est. enthalpy J/mol	est. pressure atm	C _v mJ/mol-K	est. enthalpy J/mol	est. pressure atm	C _v mJ/mol-K	est. enthalpy J/mol
0	5.3	0	0	-758.0	427	0	-636.0	710	0	-476.0
1	5.3	0.93	0.93	-758.0	427	0.40	-636.0	710	0.29	-476.0
2	5.3	7.58	7.58	-757.9	427	3.26	-636.0	710	2.31	-476.0
3	5.3	26.4	26.4	-757.9	427	11.18	-636.0	710	7.88	-476.0
4	5.3	65.3	65.3	-757.9	427	27.1	-636.0	710	18.94	-476.0
5	5.3	134.6	134.6	-757.8	427	54.2	-635.9	710	37.5	-476.0
6	5.3	247	247	-757.6	427	96.8	-635.9	710	66.3	-475.9
7	6.5	418	455	-757.3	427	159.1	-635.7	710	107.7	-475.8
8	8.5	662	743	-756.7	427	247	-635.5	710	164.9	-475.7
9	12	990	1209	-755.8	427	367	-635.2	710	242	-475.5
10	16	1413	1818	-754.3	427	523	-634.8	710	343	-475.2
11	22	1919	2589	-752.1	427	723	-634.2	710	471	-474.8
12	28	2499	3504	-748.9	427	968	-633.3	710	632	-474.2
13	37	3142	5112	-744.5	427	1259	-632.2	710	830	-473.5
14	50	3866	6816	-738.5	427	1597	-630.8	710	1067	-472.9
15	63	4678	8505	-731.0	427	1978	-629.0	710	1340	-471.4

Table 3-4. Sources of Thermodynamic Diagrams

Authors	Citation	Modification	Type
Woolley, et al.	[6368]	normal hydrogen	T-S
Roder and Goodwin	[12540]	parahydrogen	T-S, and H-S
Roder, et al.	[29210]	parahydrogen	T-S, and H-S
McCarty and Roder	[49619][40029]	parahydrogen	T-S, large, for saturated liquid
Mullins, et al.	[12596]	parahydrogen	T-S, low temperatures
Sindt and Mann	[38237]	parahydrogen	T-S, triple point region
Prydz	[43781]	normal deuterium	T-S, PV/RT vs P

3.3 Joule-Thomson Data

Experiments on the Joule-Thomson effect are seldom conducted at low temperatures in spite of the tremendous importance in refrigeration processes. Since we limit the temperature to values below critical, only a small portion of the Joule-Thomson inversion curve is of interest here. This is the part of the curve lying entirely in the liquid state and intersecting the vapor pressure curve a few degrees below critical on the vapor side. Tables or graphs of calculated values are given by Woolley, et al. [6368] for normal hydrogen, by Roder, et al. [29210] for parahydrogen, and by Prydz [43781] for normal deuterium.

3.4 Velocity of Sound

Solid

The velocity of sound has been measured by a number of authors for both hydrogen and deuterium. Bezuglyi and Minyafaev measured the longitudinal and transverse velocity of sound in polycrystalline normal hydrogen [47659] and in polycrystalline normal deuterium [51024]. Bezuglyi, et al. [78548] made similar measurements on parahydrogen. Their results are reproduced in table 3-5. The transverse velocities agree very well with later measurements on single crystal hydrogen and deuterium made by Wanner and Meyer [81564]. The longitudinal velocities, however, differ by about 10%.

Liquid

The most comprehensive measurements on liquid hydrogen have been made by Younglove [32387]. Figure 3-2 is taken from this reference to illustrate the slight difference in velocity of sound between saturated liquid normal and parahydrogen. The agreement between velocities of sound calculated from the PVT surface by Roder, et al. [29210] or McCarty and Weber [80777] and illustrated in table 2-2 is about 2% along the saturation boundaries and approximately 0.3% in the single phase liquid or vapor regions.

Table 3-5. Sound Velocity in Solid Hydrogen and n-Deuterium

T, K	V cm ³ /mol	v _l m/s	v _t m/s	β _s × 10 ¹⁰ cm ² /dyn	"adiabatic" Poisson's ratio	β _T · 10 ¹⁰ cm ² /dyn	isothermal Poisson's ratio
n-hydrogen							
4.2	22.64	2190	1160	3.8	0.30	4.4	
11.49	23.08	2110	1080	3.9	0.32	4.6	
13.96	23.24	2060	1030	4.1	0.33	5.1	
p-hydrogen							
	g/cm ³						
0	0.0892	2110*	1140*	4.12	0.29	4.1	0.29
1	0.0892	2110*	1140*	4.12	0.29	4.6	0.29
2	0.0892	2110	1140	4.12	0.29	4.7	0.29
3	0.0891	2105	1140	4.15	0.29	4.8	0.26
4	0.0890	2100	1140	4.19	0.29	4.9	0.26
5	0.0889	2095	1140	4.22	0.29	5.2	0.25
6	0.0888	2090	1135	4.24	0.29	5.3	0.25
7	0.0887	2085	1130	4.26	0.29	5.3	0.25
8	0.0885	2075	1125	4.31	0.29	5.4	0.25
9	0.0883	2065	1120	4.36	0.29	5.4	0.25
10	0.0880	2050	1115	4.46	0.29	5.4	0.25
11	0.0877	2035	1105	4.53	0.29	5.4	0.25
12	0.0874	2010	1095	4.64	0.29	5.6	0.25
n-deuterium							
	cm ³ /mol						
4.2	19.56	1920	1005	2.1	0.31	2.1	0.31
6	19.64	1920	1005	2.1	0.31	2.1	0.31
8	19.72	1920	1005	2.1	0.31	2.1	0.31
10	19.83	1920	1000	2.1	0.31	2.2	0.30
12	19.98	1910	990	2.1	0.31	2.3	0.30
14	20.12	1890	960	2.2	0.32	2.5	0.30
16	20.26	1860	930	2.2	0.33	2.8	0.30

* extrapolated

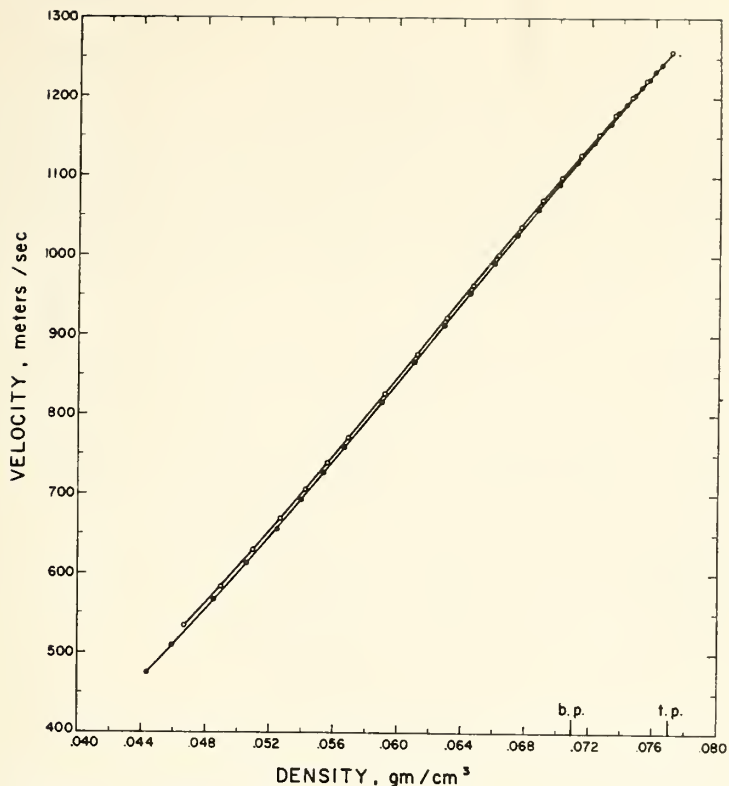


Figure 3-2. Velocity of Sound in Saturated Liquid n- and Parahydrogen

Gas

Measurements on liquid and gaseous deuterium have been made by Bezuglyi and Minyafaev [51024], by Güsewell, et al. [64932], by van Itterbeek and van Paemel [1860], and by van Itterbeek and Vandoninck [5709]. The results are collected together in table 3-6.

Table 3-6. Velocity of Sound of Normal Deuterium, Liquid and Gas

liquid at saturation		gas [5709]		
T, K	velocity m/s	T, K	P, atm	velocity m/s
19	1070 [51024]			
20	1060			
25	934.4 [64932]	18.90		255.3
26	907.2	-19.55		259.6
27	877.4	20.36	0.295	262.35
28	846.0	20.36	0.125	263.52
29	813.2	20.38		265.2
30	777.9	20.98		268.9
31	740.1	29.80		261.4

} [1860]

4. Specific Heat

Some fairly extensive experimental work has been done on the heat capacity of hydrogen and deuterium. For the other hydrogens the information available is more limited. For the isotopes having ortho-para isomers a large dependence of specific heat on composition is found for the solid, but not for the liquid. We distinguish between heat capacity at saturation (C_s), heat capacity at constant pressure (C_p) and heat capacity at constant volume (C_v).

The general features of the property for hydrogen are as follows: Below ≈ 14 K the heat capacity (C_s) of the solid shows a broad anomaly for compositions between para and normal; with increasing ortho concentration in this temperature range the heat capacity increases; for ortho concentrations larger than 60%, a λ transition exists at low temperatures ~ 1.3 to 4.2 K. The values of C_s are commonly taken to be the same as C_p ; the values of C_v are lower than C_s at all temperatures but the difference is negligible below 5 K; a significant difference exists between C_s of the solid and the liquid; similarly a large difference exists between the C_s and the C_v of the liquid.

4.1 Hydrogen

Solid

A composite picture for the heat capacity of the solid (C_s) is shown in figure 4-1. Values for parahydrogen are given in table 3-1; they were taken from Ahlers [21136]. Values for other ortho-para compositions are from the paper by Hill and Ricketson [6354]. Not shown in any detail is the λ -transition near 2 K, the best description of which is given by Ahlers and Orttung [21806]. Ahlers [21136] also measured the specific heat C_v for three molar volumes of parahydrogen in compressed solid states. The values have been shown in table 3-3; they were converted to estimated values of C_p by Dwyer and Cook [28604], and are presented in figure 4-2. More recent measurements by Roberts and Daunt [75211] are in very good agreement with those of Ahlers [21136].

Liquid

Values for the C_p , C_v , and C_s of liquid parahydrogen are shown in figure 4-3. Values for the graph have been taken from the thermodynamic tables of Roder, et al. [29210, see table 2-2] which in turn are based on the measurements of C_v and C_s by Younglove and Diller [14129, 13696]. Equivalent tables of values for saturated liquid and compressed liquid states are given by McCarty and Weber [80777] in a different set of units. Additional experimental measurements at pressures greater than 340 atm have been published by Orlova, et al. [50155]. As mentioned above, the differences in specific heats for different ortho-para compositions in the liquid state are very small.

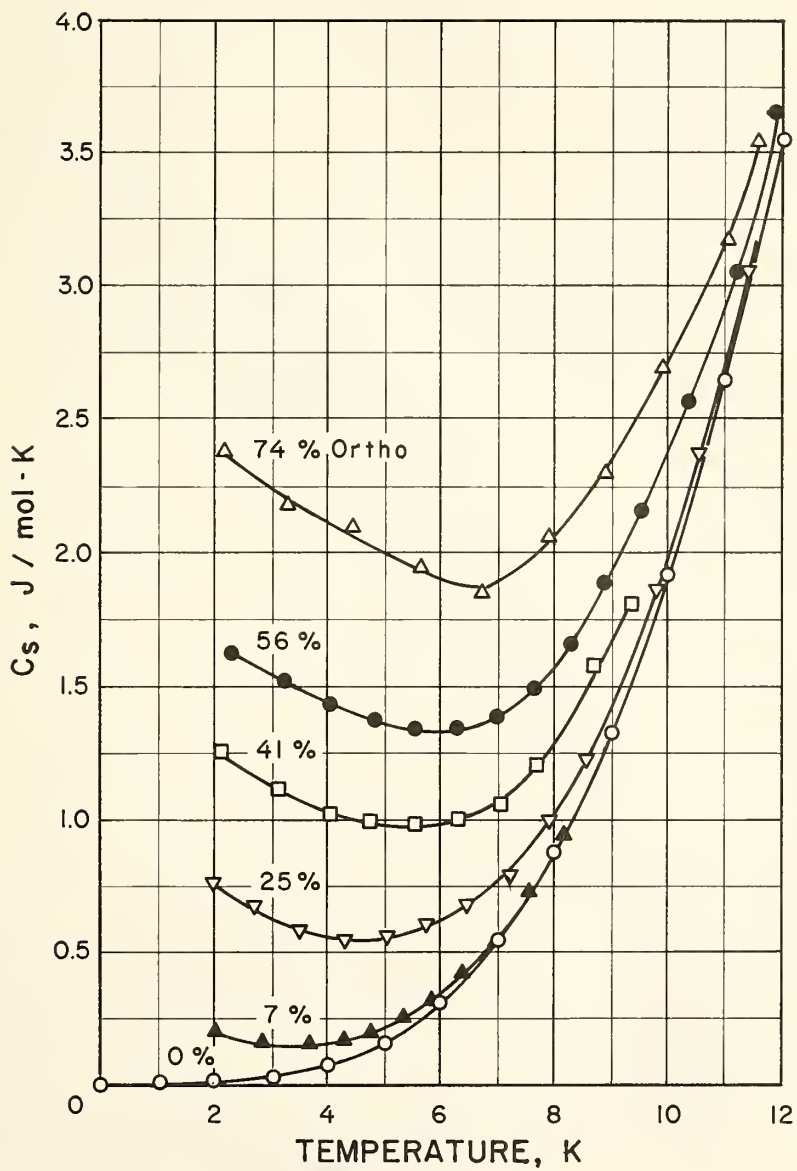


Figure 4-1. The Heat Capacity (C_s) of Solid Hydrogen

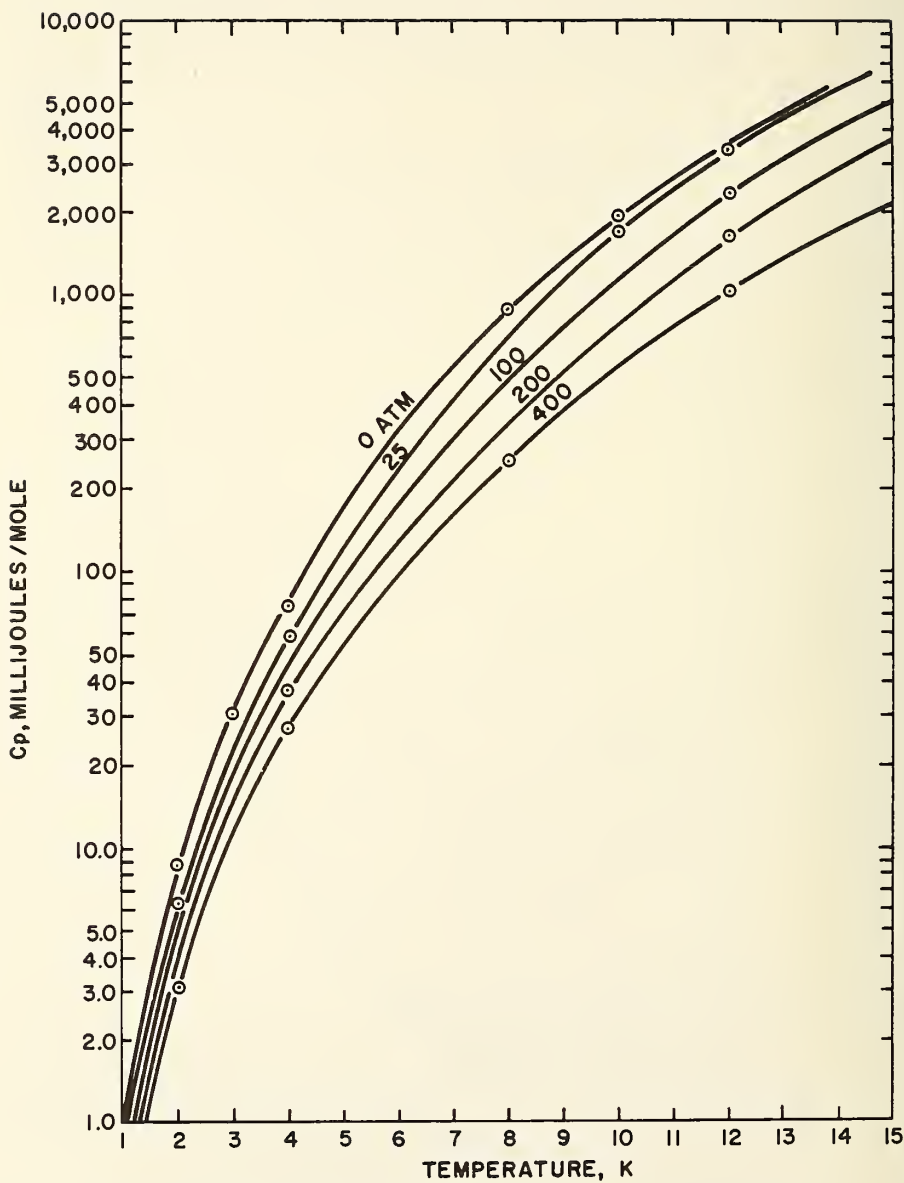


Figure 4-2. Isobars of C_p for Compressed Solid Parahydrogen

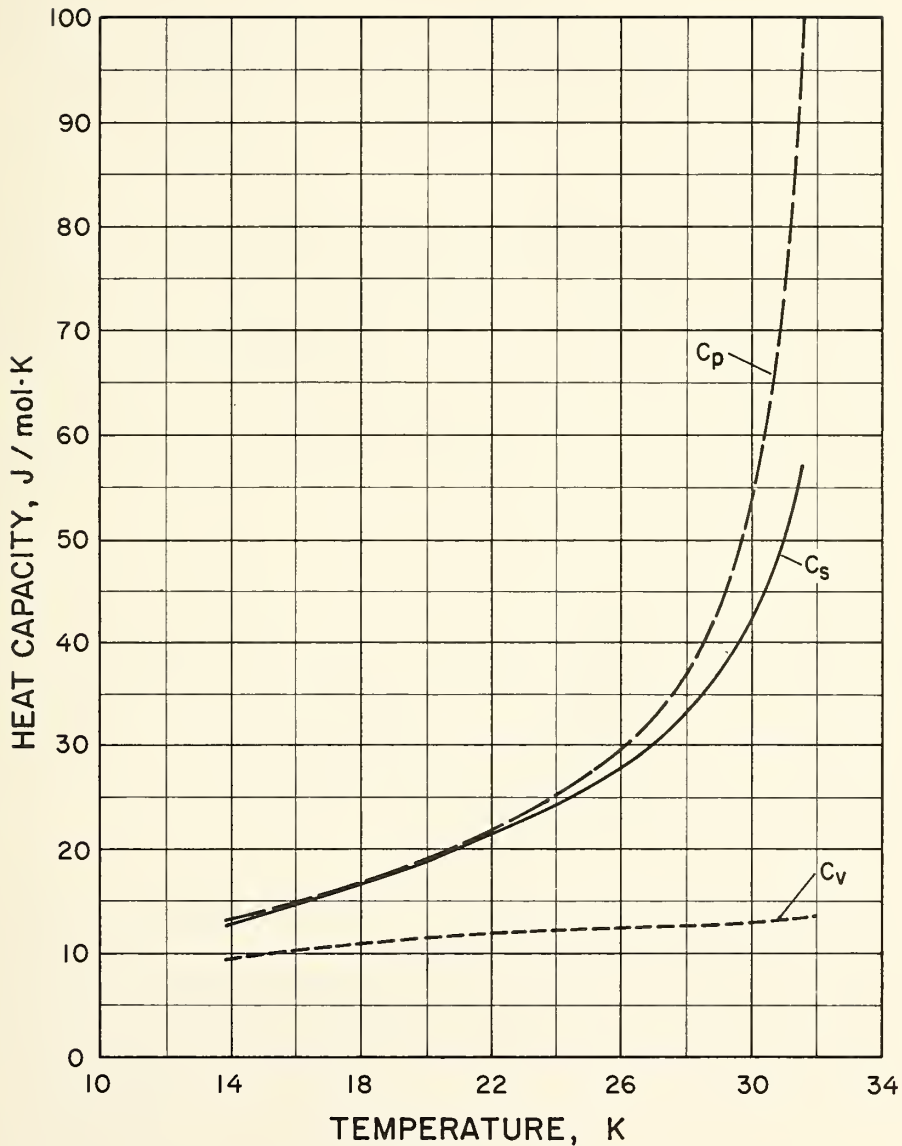


Figure 4-3. The Heat Capacities C_p , C_v , and C_s for Liquid Parahydrogen

Gas

Calculated values for the heat capacities of the gas follow from essentially the same sources as for the liquid above. Differences in specific heats between parahydrogen and other ortho-para compositions exist for the gas states. They are, however, not noticeable, that is less than 1%, for temperatures below 40 K. Specific heat differences if required are best obtained from the tabulations of the ideal gas thermodynamic functions [11271].

4.2 Deuterium

Solid

A composite picture for the heat capacity of solid deuterium (C_s) is given in figure 4-4. The sources of data are Clusius and Bartholome [5595], Kerr, et al. [645], Gonzalez, et al. [8370], Hill and Lounasmaa [6416], and Grenier and White [22677]. Details of the λ -transition are given by Grenier and White [22677]. Calculated values of C_v of the solid and liquid are given in figure 4-5, while values of C_s for liquid deuterium are given in figure 4-6. A good source of data for C_s of the liquid is the paper by Brouwer, et al. [69648], while values of C_v for the liquid are gleaned from the paper of Bartholome and Eucken [5119].

4.3 Other Isotopes

Tritium

For liquid tritium one can estimate values of C_v from the theoretical cell model calculation of Henderson [21016]. For example, the value of C_v at 29.6 K and 38 mol/l, i. e., at saturation conditions, is 15.4 J/mol-K. Values for the ideal gas states have already been introduced. The source of tables and graphs is Jones [18037]. As noted previously there is a 3% difference in C_p^0 between this source and the more recent calculations by Haar, et al. [11271].

HD

Values for solid HD have been measured by Grenier and White [22899]. They are reproduced in table 4-1. White and Gaines [29861] measured values for mixtures of hydrogen and deuterium and found that the results could be represented by a linear combination of the pure components, thus the solid solutions are nearly ideal. The heat capacity of the liquid varied linearly from 4.48 cal/mol-K (18.7 J/mol-K) at the triple point (16.60₄ K) to 6.29 cal/mol-K (26.3 J/mol-K) at 22 K [Brickwedde and Scott, 27091]. Additional values of C_s or C_v could be established from the cell model calculation of Henderson [13537 or 21016], who obtains 6.255 cal/mol-K at 22 K. Heat capacities for the ideal gas are given by Haar, et al. [11271] for temperatures above 50 K.

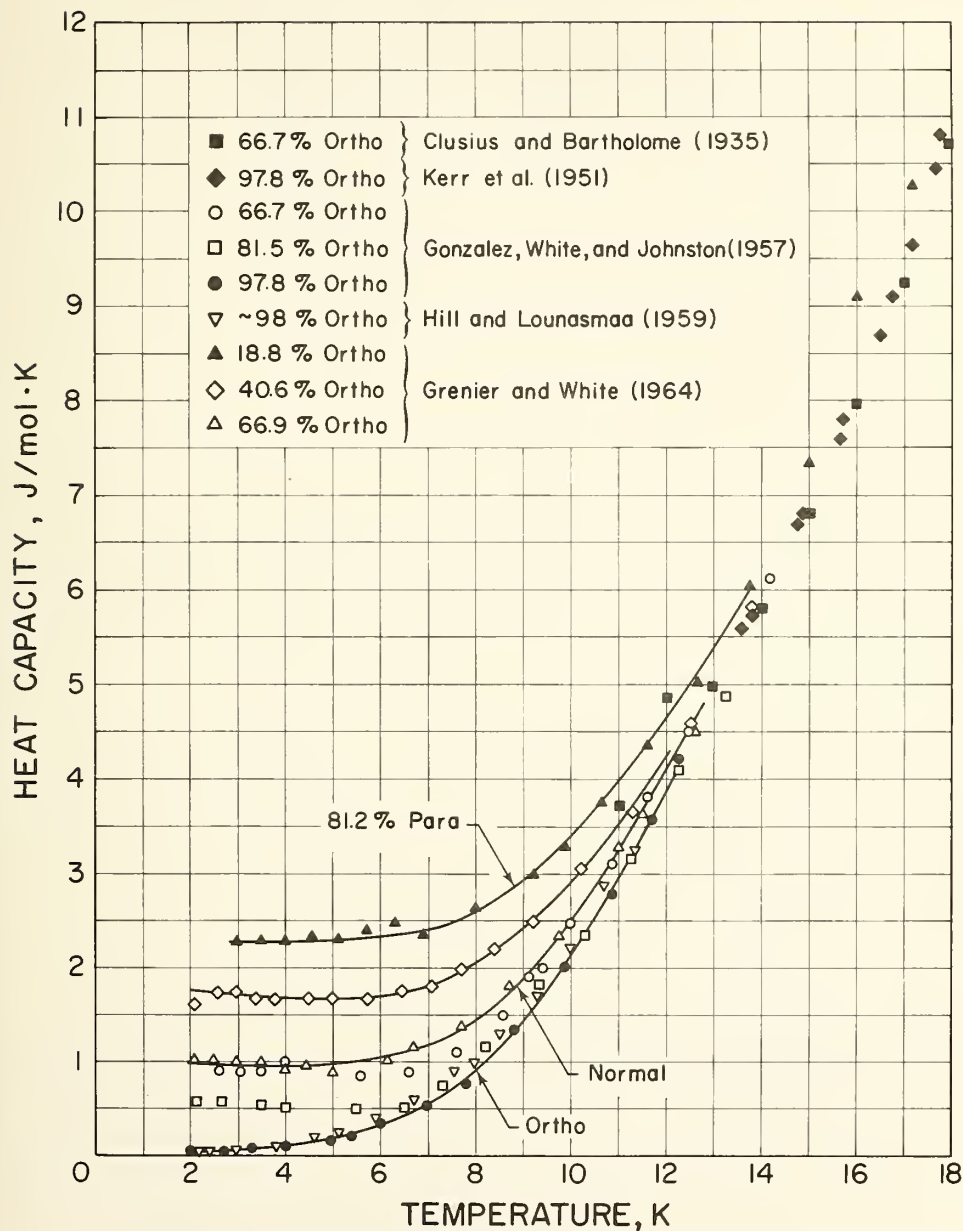


Figure 4-4. The Heat Capacity (C_s) of Solid Deuterium

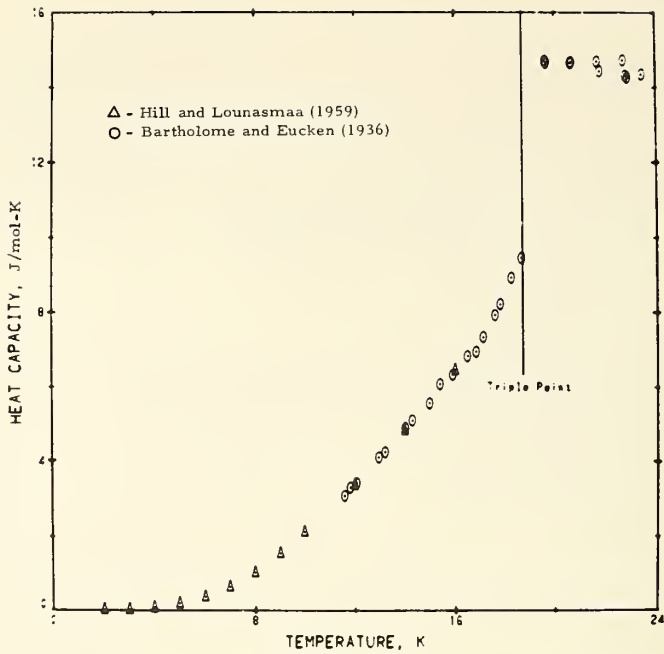


Figure 4-5. The Heat Capacity (C_V) of Solid and Liquid Deuterium

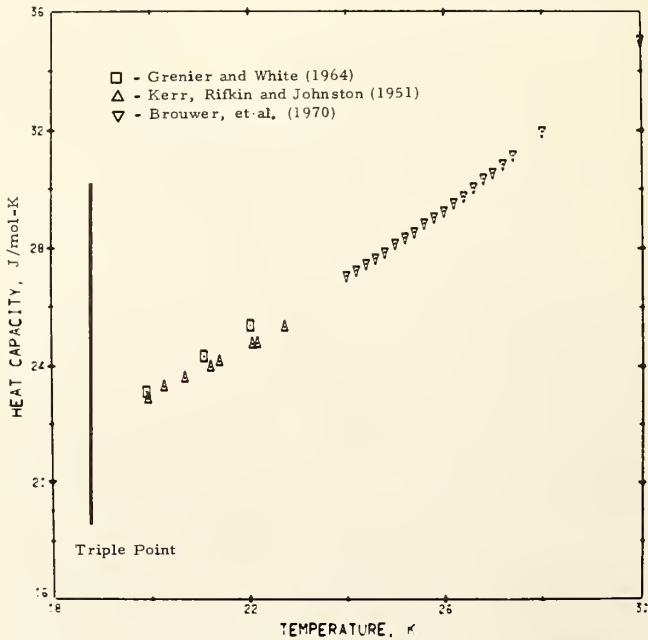


Figure 4-6. The Heat Capacity (C_S) of Liquid Deuterium

Table 4-1. Heat Capacity of Solid HD

Heat capacity of solid HD, 0.06751 moles in calorimeter,
98.58 mole% HD; 1.15% H₂; 0.27% D₂.

T (°K)	ΔT (deg)	C_p (cal mole ⁻¹ deg ⁻¹)	T (°K)	ΔT (deg)	C_p (cal mole ⁻¹ deg ⁻¹)
2.524	0.0380	0.0184	6.469	0.6463	0.117
2.601	0.0918	0.0128	7.014	1.1292	0.156
2.897	0.0732	0.0183	7.714	0.9080	0.216
3.186	0.3192	0.0252	8.274	1.0461	0.282
3.317	0.1414	0.0269	8.904	1.0014	0.367
3.585	0.8249	0.0240	9.857	1.0367	0.510
3.812	0.6505	0.0335	10.815	1.1748	0.672
4.050	0.6325	0.0483	11.888	1.2102	0.889
4.009	0.6525	0.0543	12.935	1.2611	1.093
5.562	0.3815	0.0762	14.132	1.4337	1.369
6.038	0.5347	0.0937	15.381	1.4682	1.679

with permission from G. Grenier, D. White, J. Chem. Phys. 40, 3451 (Jun. 1964).

HT

The only values found in this search are those for the ideal gas calculated by Jones [18037]. The difference between this source and the calculations of Haar, et al. [11271] is 1% in C_p^0 at 50 K. Since the mass of HT is approximately the same as that of D₂, the properties of HT are often taken to be the same as those of D₂. Henderson tabulates cell model calculations for HT [21016], thus one could calculate estimates of C_v for the liquid states provided that one could obtain reasonable estimates for the appropriate values of liquid density.

DT

Henderson [21016] tabulates cell model calculations for C_v of DT. Thus in principle one could calculate estimates for C_v in the liquid states provided that one could obtain reasonable estimates for appropriate values of the liquid density. Ideal gas thermodynamic values are tabulated for DT only at temperatures above 50 K [Haar, et al., 11271].

5. Transport Properties

5.1 Surface Tension

Surface tension data for hydrogen [43099, 25058, 23183, 27064] prior to 1965 are limited to temperatures below the normal boiling point; the sets disagree by about 10%. Corruccini [30250] has shown that the different sources can be made to agree by applying the capillary corrections in a consistent manner to the older data [43099, 25058]. For temperatures higher than about 21 K only the measurements by Blagoi and Pashkov [33765] on normal hydrogen are available. Comparison of these measurements with the equations proposed by Corruccini shows that extrapolation of the equations to temperatures above about 22 K leads to sizeable differences at the higher temperatures. Blagoi and Pashkov suggest that no single simple empirical equation can represent the temperature dependence of the surface tension

Table 5-1. Surface Tension of the Hydrogens

	p-H ₂		n-H ₂		D ₂		HD					
	T K	Y dyn/cm	T K	Y dyn/cm	T K	Y dyn/cm	T K	Y dyn/cm				
13.803 (t.p.)	2.990		21.15	1.755	24.77	1.171	18.77	3.80	20.45	2.683 [*]	18.72	3.018
13.947 (t.p.)		3.004	21.40	1.715	24.98	1.136	18.86	3.79	20.44	2.686	18.36	3.087
14		2.995	21.98	1.633	25.16	1.100	19.10	3.76	20.44	2.677	18.16	3.114
15		2.792	22.13	1.594	25.25	1.089	19.23	3.72	20.42	2.692	17.92	3.189
16		2.627	22.31	1.575	25.37	1.073	19.62	3.63	20.41	2.699	17.56	3.239
17		2.462	22.43	1.565	26.05	0.969	20.07	3.53	20.41	2.700	17.48	3.255
18		2.298	22.65	1.516	26.72	0.869	20.39	3.46	20.12	2.748	17.43	3.258
19		2.135	22.83	1.502	27.31	0.767	20.49	3.44	19.65	2.837	16.80	3.374
20		1.973	22.84	1.486	28.68	0.540			19.58	2.845	16.76	3.377
20.268 (n.b.p.)	1.930		22.86	1.481	29.49	0.401			19.12	2.942	16.71	3.389
20.38 (n.b.p.)		1.946	23.25	1.431	30.27	0.303			18.84	2.993		
			23.28	1.421	31.23	0.193						
			23.62	1.361	31.49	0.149						
			23.80	1.351	31.90	0.116						
			23.98	1.307	32.57	0.053						
			24.33	1.244	32.77	0.0459						

from triple point to critical point. Apparently neither a linear equation, nor a van der Waals equation with $n = 11/9$, nor inclusion of the quantum parameter (see for example [44771]) yields an acceptable representation of the data.

Values of the surface tension in table 5-1 for normal and parahydrogen up to the normal boiling point were taken from Corruccini. Values for $n\text{-H}_2$ at temperatures above 21 K were taken from Blagoi and Pashkov [33765]. Values for HD are taken from Grigor'ev [22219], values for D_2 from Grigor'ev and Rudenko [23183]. In addition to the values presented here, values for $H_2\text{-}D_2$ mixtures are found in the paper by Grigor'ev and Rudenko while a better choice of quantum mechanical mixing parameter for this paper is discussed by Bellemans and Fuks [34228].

Contact Angle

The liquid vapor contact angle is less than $1' 30''$ according to Blagoi and Pashkov [33765].

5.2 Thermal Conductivity

Solid

The thermal conductivity of the solid hydrogens, similar to the heat capacity, is highly dependent on ortho-para composition. It also depends on crystal quality, and should depend on density for the compressed solid states. However, most of the measurements made were done at or near saturation pressure. Experimental measurements on hydrogen and deuterium have been conducted by Hill and Schneidmesser [8384, 9988], Dwyer, et al. [38751], Bohn and Mate [68582], and Daney [73126]. The experimental results for the solids are plotted in figure 5-1.

Thermal conductivity of solids is usually analyzed in terms of the thermal resistance $W = 1/k$. The intrinsic resistance, W_i , that is phonon-phonon scattering, follows a T^n dependence for nonmetals where n ranges from 1 to 3. For parahydrogen, the value of n is between 3 and 4 if estimated from the results of Hill and Schneidmesser for temperatures between 4 and 11 K. The results of Dwyer, et al. [38751] are in apparent disagreement with this temperature dependence; they are too low in W (too high in k). The disagreement probably occurs because the measurements were taken along the melting curve at elevated pressures, i. e., the densities are higher than those prevailing at the triple point. We recall that for solid helium, thermal conductivity is highly dependent on density [Webb and Wilks, 580].

For deuterium, we note the comment of Hill and Schneidmesser that the conductivity curve for normal deuterium (33% para) was very similar to the corresponding curve of hydrogen (33% ortho). We add an estimated curve for normal deuterium "anchored" by the point published by Hill and Schneidmesser [8384] and by Daney's [73126] values near the triple

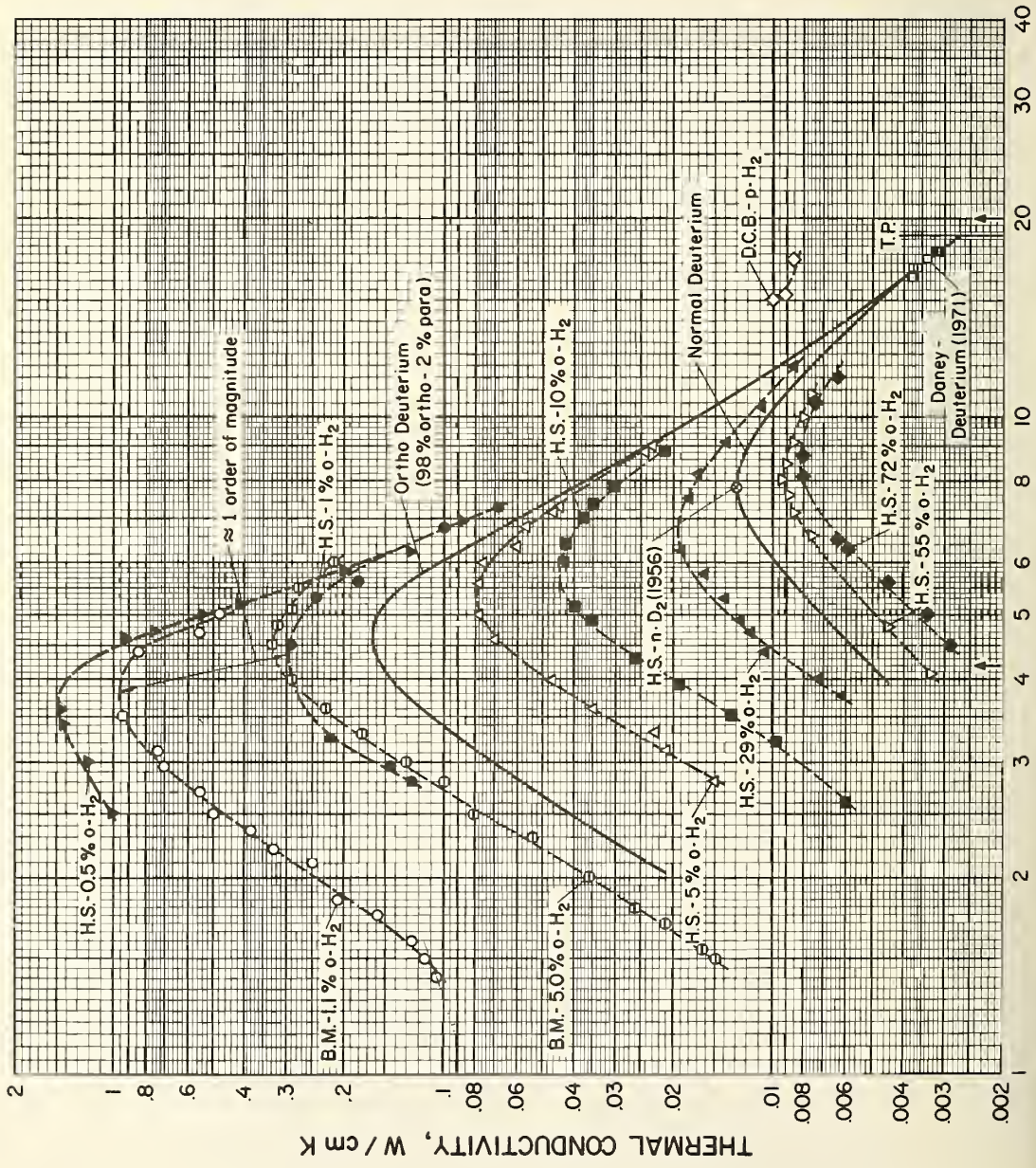


Figure 5-1. Thermal Conductivity of the Solid Hydrogens

point. We have also added a second estimated curve for orthodeuterium, i. e., 20.4 K equilibrium deuterium with about 2% para, by assuming that this curve will be similar to a hypothetical 2% ortho curve for hydrogen. We note that the hydrogen results of Hill and Schneidmesser (1% ortho) and those of Bohn and Mate (1.1% ortho) differ by almost an order of magnitude. This uncertainty should decrease to about 3% as the temperature approaches the triple point.

Liquid

The thermal conductivity of the liquid depends on temperature and is particularly sensitive to density. Its behavior is related to that of C_v and C_p . For temperatures up to the various critical points the thermal conductivity shows very little dependence on ortho-para composition. Experimental values have been measured for hydrogen and deuterium by Powers, et al. [5528, 756], Dwyer, et al. [38751] and Roder and Diller [65729]. A corresponding states estimate was made by Kerrisk, et al. [17393]. We reject the results of Dwyer, et al. because they differ from the others by a factor of nearly 3. If we compare the Powers, et al. [5528] results on hydrogen with those of Roder and Diller [65729], the temperature dependence is similar; the values, however, differ by about 20%. We attribute the difference to convection. Calculation of the Rayleigh numbers for conditions prevailing in the earlier experiment discloses a definite contribution from convection. Experimental conditions in the recent measurements were controlled to limit the contribution from convection to less than 1%. Since the absolute values of thermal conductivity of saturated liquid hydrogen and saturated liquid deuterium from the same laboratory [5528, 756] differ by only 6%, we postulate that a correction for convection is also required for the deuterium results.

The correlation of Kerrisk, et al. [17393] is based in part on the results of Powers, et al. We take the estimates of Kerrisk, et al. for the saturated liquid boundaries of D_2 and T_2 and subtract 0.266 mW/cm-K from all values, about 20%. In addition, we scale the temperature range $0.93 T_c - T_c$ to reflect the rise in thermal conductivity shown by hydrogen near the critical point. The estimates for the boundaries, so adjusted, are plotted along with the one for H_2 [Roder and Diller, 65729] in figure 5-2. The pressure dependence for liquid parahydrogen is illustrated in table 5-2 prepared by Angerhofer and Hanley [74127] from the paper by Roder and Diller [65729]. Additional tables of values for parahydrogen in a different set of units are given by McCarty and Weber [80777].

Values for the thermal conductivity of liquid hydrogen are uncertain by about 3%, the saturation boundary for deuterium could be in error by as much as 25%, while the saturation boundary for tritium simply represents an estimate.

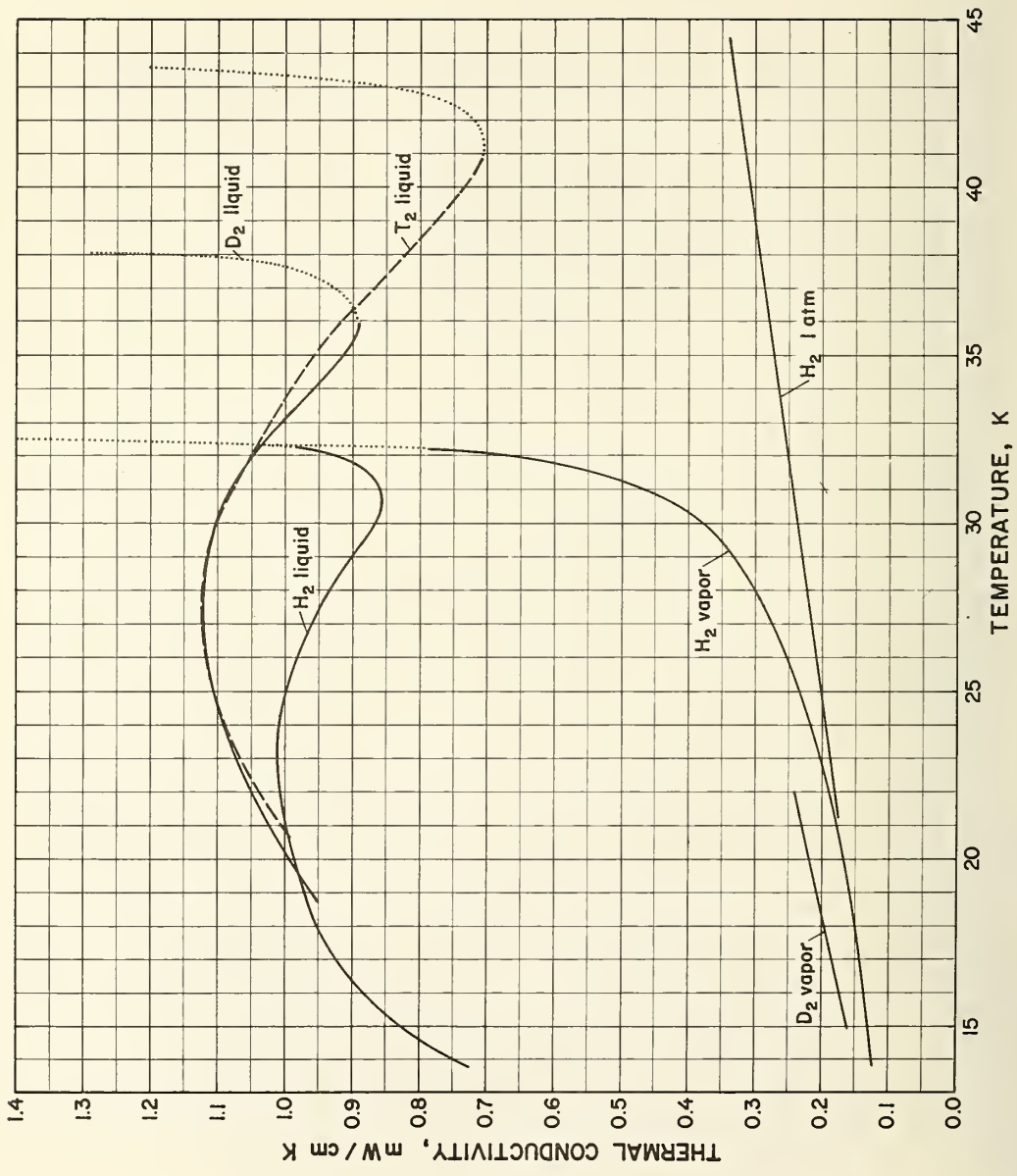


Figure 5-2. Thermal Conductivity of the Hydrogens, Saturated Liquid and Gas

Gas

The thermal conductivity of the gas depends primarily on temperature and on density or pressure. For temperatures below critical the dependence on ortho-para composition is very slight $\sim 0.5\%$. At very low pressures, in the range of "free molecular conduction", the effective thermal conductivity does not depend on the gas at all but is apparatus dependent instead. Figure 5-3 shows the drastic change between ordinary and free molecular conduction for hydrogen [Ubbink, 693].

Major sources of experimental measurements on hydrogen and deuterium are Ubbink [693, 5825], and Roder and Diller [65729]. Results for the saturated vapor of hydrogen are plotted in figure 5-2 along with the 1 atm isobar to illustrate changes in thermal conductivity with density or pressure. The deuterium results plotted in figure 5-2 for Ubbink [5825] are very nearly at saturation conditions. Additional values for parahydrogen are given in the tables of McCarty and Weber [80777].

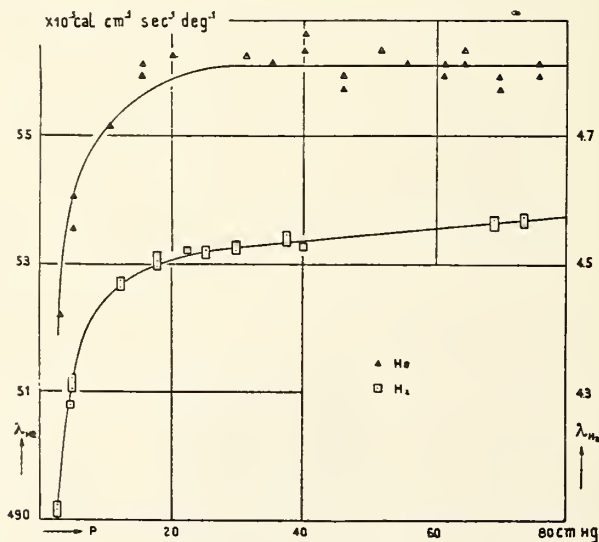


Figure 5-3. Thermal Conductivity of Hydrogen at Low Pressures with permission from J. B. Ubbink, *Physica* 13, 659 (Dec. 1947).

Differences in ortho-para composition have been measured by Müller, et al. [68346] for hydrogen, and by Uebelhack, et al. [77494] for deuterium. The results are given below.

For ortho-para mixtures of hydrogen at 21 K with γ = percent ortho hydrogen and

$$\text{ratio} = - (\lambda / \lambda_P - 1) \times 10^3,$$

γ	0.15	0.22	0.27	0.31	0.50	0.50	0.50	0.50	0.75	0.75	0.75	0.75	0.92	0.92	0.96	1.00
ratio	0.59	1.12	1.42	1.83	3.22	3.32	2.93	3.26	5.92	5.94	5.95	6.10	8.15	8.05	8.41	9.31

For deuterium the results of the measurements over the whole concentration range from pure o-D₂ to pure p-D₂ are represented by

$$\lambda/\lambda(\gamma_p = 0) = 1 - k_1 \gamma_p - k_2 \gamma_p^2 ,$$

with $k_1 = 2.17, 2.43, 2.97 \cdot 10^{-3}$ and $k_2 = 2.37, 2.27, 2.24 \cdot 10^{-3}$ for $T = 18.4, 19.6, 21.0$ K respectively, where γ_p is the molar fraction of p-D₂.

Values of the thermal conductivity of gaseous hydrogen are uncertain by about 3%. The uncertainty for deuterium is estimated to be about the same, because the two sources for hydrogen agree to within 2%.

5.3 Thermal Diffusion

Diffusion is a mixture phenomenon and as such enters into problems of isotope separation in a very practical way. It is perhaps best to first define several of the terms commonly used. The diffusion coefficient is defined as the coefficient relating the flux of a given species in a mixture to the concentration gradient of that species under isothermal conditions. For a two-component mixture of species i and j :

$$\tilde{J}_i = D_{ij} \text{grad } c_i$$

where \tilde{J}_i is the matter flux of i , $\text{grad } c_i$ the gradient of concentration, and D_{ij} the diffusion coefficient. Note that the diffusion coefficient is often referred to as the mass diffusivity. In addition to the diffusion coefficient an artificial quantity, the self-diffusion coefficient is often discussed. Self-diffusion refers to the diffusion of one isotopic form into another. If the system under consideration is subjected to a temperature gradient, then for a binary system the equation above has to be extended to the following

$$\tilde{J}_i = -D_{ij} \text{grad } c_i - D_{ij}^T \text{grad } T$$

where D_{ij}^T is the thermal diffusion coefficient.

Experimentally thermal diffusion measurements are most often made on gases using a two bulb technique. Each bulb is held at a different temperature. Thermal diffusion coefficients are then presented for mean values of temperature. These mean temperatures are almost always higher than critical temperature, and therefore outside of the scope of this report. We restrict ourselves to a brief description of a few of the most pertinent papers.

A summary of experimental results completed by 1966 is given by Grew and Humphreys [40966] who note that discrepancies between different laboratories are large. Calculations of thermal diffusion coefficients from intermolecular potentials have been published by Diller and Mason [35559] and by Monchick, et al. [54131].

5.4 Thermal Diffusivity

Thermal diffusivity is a parameter frequently used in engineering applications. Thermal diffusivity is defined as

$$\alpha \equiv k/\rho C_p .$$

In this equation, α is the thermal diffusivity, k the thermal conductivity, ρ the density, and C_p the heat capacity at constant pressure. The thermal diffusivity can be obtained directly, for example, from a light-scattering experiment. Usually, however, values are calculated from the other properties. Sources of data are Daney [73126] and Ludtke and Roder [78262] for deuterium. For parahydrogen McCarty and Weber [80777] present extensive tables for both liquid and gaseous states.

Applicable property values and calculated thermal diffusivities are shown for solid and liquid ortho and normal deuterium in table 5-3 which is taken from Ludtke and Roder. The thermal conductivity of solid deuterium is "anchored" with Daney's values. Therefore, the agreement between his values of thermal diffusivity and those presented in table 5-3 is excellent. The uncertainty in α near the triple point is about 3% as obtained from the errors in the other variables. Since the solid thermal conductivity is very sensitive to para content, the value of thermal diffusivity of ortho deuterium near 4 K has to be considered uncertain by one order of magnitude. Note that values of thermal diffusivity differ by several orders of magnitude for the lower temperatures of the solid.

5.5 Viscosity

Sources of data for various hydrogens are discussed in more detail below. A composite of all saturated liquid curves is given in figure 5-4. In this figure p-H₂, HD, and D₂ are based on experimental measurements. HT and T₂ are based on a corresponding states calculation which as its base used experimental values for D₂ which we now believe to be in error. The corresponding states calculation ought to be redone because better input data are now available. In addition, the correlation should be done in terms of density rather than temperature.

Hydrogen

As source of data we select Diller [27674], who measured viscosity of p-H₂ at temperatures from 14 to 100 K and at pressures to 345 atm. Extensive tables are presented in the reference; we present only the tables of saturated liquid p-H₂ and saturated liquid n-H₂. It can be seen that the viscosity of saturated liquid n-H₂ is up to 5% higher than that of p-H₂ at the same temperature. However, there is virtually no difference in viscosity, if the comparison is made at the same densities.

Table 5-3. Thermal Diffusivity of Deuterium

ORTHO DEUTERIUM				NORMAL DEUTERIUM					
TEMPERATURE		SOLID		TEMPERATURE		SOLID			
KELVIN	DENSITY MOL/CM	HEAT CAPACITY J/MOL-K	THERMAL CONDUCT. W/CM-K	DIFFUSIVITY CM ² /SEC	KELVIN	DENSITY MOL/CM	HEAT CAPACITY J/MOL-K	THERMAL CONDUCT. W/CM-K	DIFFUSIVITY CM ² /SEC
4.000	0.0507	0.111	0.1440	25.58772	4.000	0.0507	0.950	0.0046	0.09551
5.000	0.0506	0.226	0.1580	13.81650	5.000	0.0506	1.030	0.0071	0.14032
6.000	0.0506	0.410	0.1020	4.91661	6.000	0.0506	1.070	0.0094	0.18101
7.000	0.0505	0.688	0.0590	1.69813	7.000	0.0505	1.200	0.0122	0.20132
8.000	0.0504	1.091	0.0368	0.66926	8.000	0.0504	1.400	0.0130	0.18424
9.000	0.0503	1.630	0.0238	0.29028	9.000	0.0503	1.830	0.0116	0.12478
10.000	0.0501	2.260	0.0170	0.15014	10.000	0.0501	2.500	0.0104	0.08303
11.000	0.0500	2.810	0.0125	0.08897	11.000	0.0500	3.230	0.0088	0.05415
12.000	0.0498	3.730	0.0095	0.05114	12.000	0.0498	4.100	0.0075	0.03673
13.000	0.0497	4.650	0.0075	0.03245	13.000	0.0497	5.000	0.0065	0.02616
14.000	0.0495	5.620	0.0060	0.02175	14.000	0.0495	6.000	0.0056	0.01886
15.000	0.0493	7.000	0.0050	0.01449	15.000	0.0493	7.000	0.0048	0.01391
16.000	0.0491	8.040	0.0041	0.01051	16.000	0.0491	8.000	0.0041	0.01044
17.000	0.0489	9.380	0.0035	0.00763	17.000	0.0489	9.400	0.0035	0.00761
18.000	0.0487	11.200	0.0030	0.00550	18.000	0.0487	11.130	0.0030	0.00555
18.691	0.0485	13.000	0.0027	0.00428	18.691	0.0485	13.070	0.0027	0.00428
LIQUID				LIQUID					
18.691	0.0432	21.850	0.0010	0.00106	18.691	0.0432	21.850	0.0010	0.00106
19.000	0.0430	22.120	0.0010	0.00105	19.000	0.0430	22.120	0.0010	0.00105
20.000	0.0425	23.010	0.0010	0.00102	20.000	0.0425	23.010	0.0010	0.00102
21.000	0.0420	23.680	0.0010	0.00100	21.000	0.0420	23.680	0.0010	0.00100
22.000	0.0414	24.760	0.0010	0.00098	22.000	0.0414	24.770	0.0010	0.00098
23.000	0.0408	25.640	0.0011	0.00105	23.000	0.0408	25.640	0.0011	0.00105

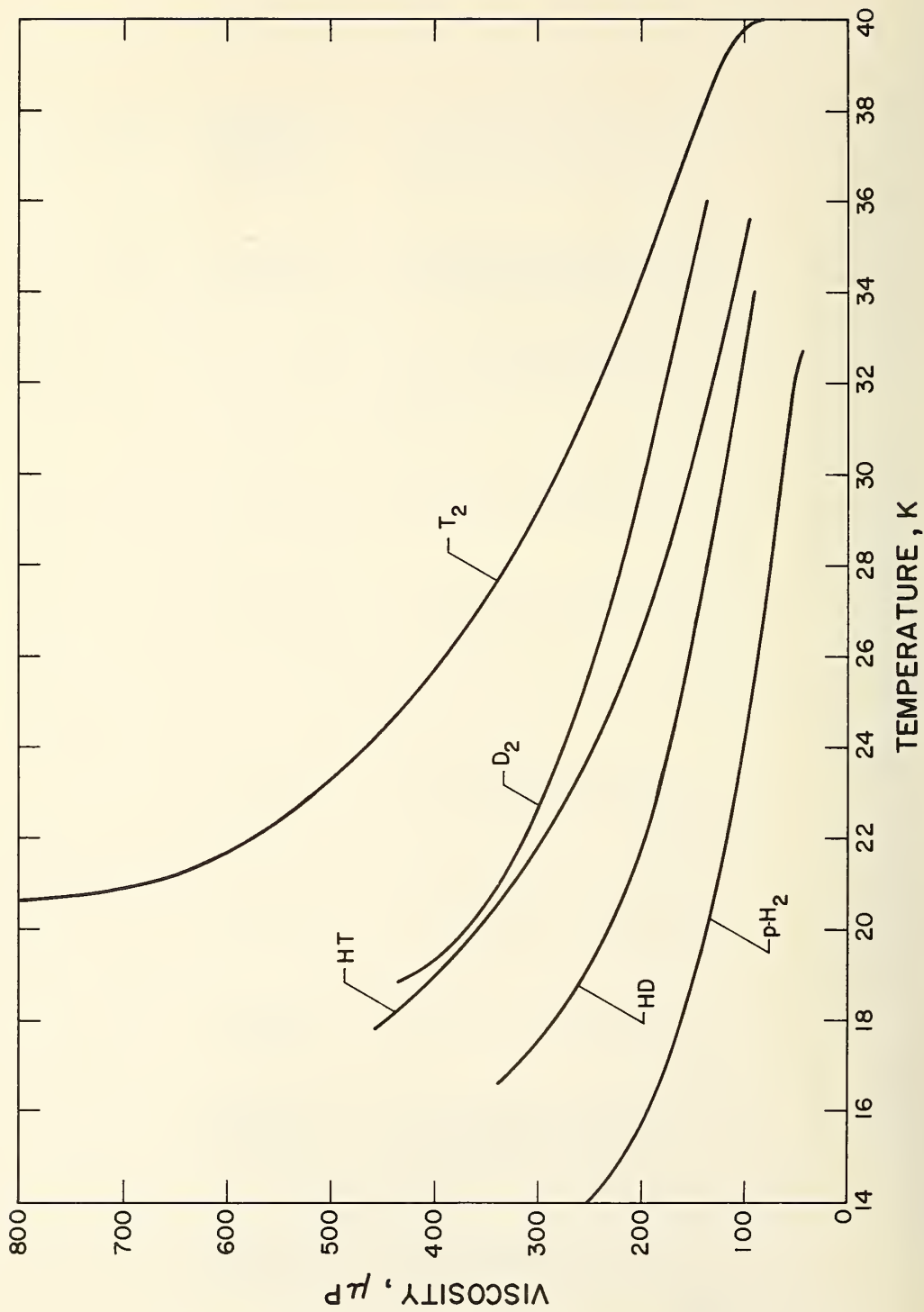


Figure 5-4. Viscosity of the Hydrogens, Saturated Liquid

Table 5-4. Viscosity of Saturated
Liquid Parahydrogen

T (°K)	ρ (g/cm ³)	$\eta \times 10^6$ (g/cm·sec)
14.000	0.07685	250.7
14.500	0.07643	234.1
15.000	0.07599	221.3
15.500	0.07555	207.3
16.000	0.07510	197.5
16.500	0.07464	185.6
17.000	0.07417	177.7
17.500	0.07368	168.5
18.000	0.07319	160.5
18.500	0.07268	153.8
19.000	0.07216	147.0
19.500	0.07163	141.3
20.000	0.07108	135.4
20.500	0.07052	130.6
21.000	0.06994	125.3
21.500	0.06934	120.6
22.000	0.06872	116.1
23.000	0.06741	108.1
24.000	0.06601	100.8
25.000	0.06449	93.5
26.000	0.06283	87.2
26.500	0.06194	84.1
27.000	0.06100	81.0
27.500	0.06001	78.1
28.000	0.05896	75.2
28.500	0.05784	72.4
29.000	0.05664	69.6
29.500	0.05534	67.0
30.000	0.05393	64.9
30.500	0.05236	61.2
31.000	0.05058	58.1
31.500	0.04852	55.7
32.000	0.04599	51.9
32.500	0.04248	47.5
32.700	0.04041	43.9

Table 5-5. Viscosity of Saturated
Liquid Normal Hydrogen

T (°K)	ρ (g/cm ³)	$\eta \times 10^6$ (g/cm·sec)
14.000	0.07717	254.3
15.000	0.07631	230.2
16.000	0.07542	203.9
17.000	0.07449	182.9
18.000	0.07350	165.6
19.000	0.07246	151.5
20.000	0.07136	139.2
21.000	0.07020	128.4
22.000	0.06896	118.7
23.000	0.06763	110.5
24.000	0.06621	102.6
25.000	0.06468	95.7
26.000	0.06302	89.0

with permission from D. E. Diller, J. Chem. Phys. 42, 2089 (Mar. 1965).

with permission from D. E. Diller, J. Chem. Phys. 42, 2089 (Mar. 1965).

Values for the dilute gas viscosities can be obtained from Diller [27674]; extensive tables calculated from the equations by Diller are presented by McCarty and Weber [80777] in engineering units. Differences in viscosity for various ortho-para compositions may approach 1% for the dilute gas; an extensive set of measurements is given by Camani [76374].

Deuterium

Several investigators have measured the viscosity of saturated liquid deuterium. We believe that the earliest of these measurements, those by van Itterbeek and van Paemel [6274], are in error. The more recent data of Rudenko and Konareva in 1963 [22496] and Konareva and Rudenko in 1967 [47292], covering the range from 18.8 to 36 K, have been selected. The selection is based on the higher precision of these measurements, and on the excellent agreement between data taken by these authors on liquid normal hydrogen with the data of Diller in the range from 22 - 32 K. No ortho-para composition of the deuterium is given. The selected experimental values for deuterium are given in table 5-6. We note that the difference in viscosity at the triple point for the two sources is large, 434 micropoises for the Russian sources vs about 530 for van Itterbeek and Paemel.

Table 5-6. Viscosity Values of the Other Hydrogens, Saturated Liquid

Deuterium		HD			HT		
Temperature (K)	Viscosity (micropoise)	T, K	η micropoise	T, K	η micropoise	T, K	η micropoise
20.4	355	14.12	8.18	16.6	338	20.62	819
20.0	368	15.47	9.05	18.0	284	21.0	678
19.5	390 [22496]	17.04	10.04	20.0 [5132]	231	21.5	618
19.0	418	18.70	11.21	22.0	200	22.0	575
18.8	434	20.32	12.17	24.0	180	22.5	542
				26.0	156 [47292]	23.0	517
36.0	136	14.4	8.80	28.0	138	24.0	510
34.0	156	20.4	12.67	30.0 [3948]	120	25.0	432
32.0	177			32.0	104	26.0	397
30.0	198	20.41	12.0	34.0	90	27.0	362
28.0	221 [47292]	26.09	15.8			28.0	331
26.0	246	32.57	20.1 [8330]	16.6	338	29.0	305
24.0	276	41.35	24.9	17.0	321	30.0	283
22.0	318	48.06	28.8	18.0	284	31.0	259
20.0	368			19.0	255	32.0	240
18.8	434			20.0	231	33.0	228
				20.4	223	34.0	205
						35.0	191
						36.0	173
						37.0	159
						38.0	142
						38.5	133
						39.0	123
						39.5	110
						40.0	82

Becker, Misenta, and Stehl [5137] measured the difference in viscosity between normal and various concentrations of ortho deuterium. They present two isotherms of 16.6 and 20.3 K indicating the relative difference in viscosity as a function of ortho concentrations from 66.67 to 98 percent. Their viscosity data is presented in terms of relative viscosity difference, $\Delta \eta/\eta \times 10^3$, defined as

$$\left[\frac{(\eta)_{x\% \text{ oD}_2} - (\eta)_{66.67\% \text{ oD}_2}}{(\eta)_{66.67\% \text{ oD}_2}} \right] 10^3 .$$

The results are listed below:

Temperature (K)	Ortho Composition			
	78.1%	84.9%	88.4%	98.0%
20.3	0.23		0.41	0.56
16.6		0.41		0.62

For a given temperature, the measurements show that ortho D_2 has a slightly higher viscosity than $n\text{-}D_2$. Here we remark again that for hydrogen the saturated liquid viscosities are nearly independent of ortho-para composition if compared at the same density.

HD

The first reported measurements of HD viscosity (gas) were by Becker and Misenta [5132] who measured viscosities of H_2 , HD, and D_2 from just above their triple points to 90 K. They estimate their errors to range from 0.68% at 14.12 K, increasing linearly to 4.12% at 90 K. Rietfield and van Itterbeek [3948] measured η for mixtures of gaseous HD and D_2 , ranging in concentration from 0 to 100% HD. Above 20 K, their reported values of η_{HD}/η_{H_2} were consistently between 1.21 and 1.22 independent of temperature. Coremans, et al. [8330] measured η for $95 \pm 2\%$ pure HD gas. Konareva and Rudenko [47292] measured η for HD at 10 temperatures along the saturated liquid boundary augmenting the earlier results of Rudenko and Konareva [42002]. Selected values for liquid and gaseous HD are given in table 5-6.

T_2 , HD, and DT

As mentioned in the introduction, estimates for the saturated liquid of these hydrogens were taken from the quantum mechanical corresponding states calculation of Rogers and Brickwedde [38388]. This correlation relies heavily on the viscosity data of van Itterbeek and van Paemel [6274] for deuterium, data which we now believe to be in error. As a consequence the values presented here are also thought to be in error, i. e., a new correlation would be desirable.

Rogers and Brickwedde reestimated the force constants, ϵ/k and σ , and the quantum correction parameter Λ^* for T_2 to be 34.5 K, 2.949 Å and 1.029 respectively, and recalculated viscosities for liquid tritium. Values are given in table 5-6. We note that the entry of 510 micropoises for 24 K must be a printing error, according to the graph a value of about 470 is much more consistent with the other values. Comparison with an earlier calculation, i. e., Kerrisk, et al. [17393] shows a large difference for η at the triple point, 739 micropoises vs 819 for Rogers and Brickwedde.

Values for the liquid viscosity of HT are also taken from the paper of Rogers and Brickwedde [38388]. They are based on a Λ^* eff of 1.374 and are presented in table 5-6.

Values of viscosity of DT may also be obtained from the graphs published by Rogers and Brickwedde. We note that the Λ^* eff for DT (1.046) is very close to that of T_2 ($\Lambda^* = 1.029$). At $T^* = 0.8$, η_{DT} is about 2% lower than η_{T_2} . It is therefore suggested that the table entries for liquid T_2 be adjusted (lowered) by 2% to obtain values for DT.

6. Electrical Properties

6.1 Breakdown Voltage

Measurements have been made by Mathes [19687] on liquid and gaseous hydrogen using 60 cycle a-c voltages applied across steel electrodes to obtain values for use in practical applications rather than intrinsic values. His results are shown in figure 6-1.

Note that electrode material and its degree of oxidation have a pronounced effect on such measurements. Also, helium is affected by presence of finely divided solid impurities such as air. Dissipation factors as discussed by Jeffries and Mathes [68981] are normally very low.

6.2 Dielectric Constant

The dielectric constant is used to indicate liquid level, to measure density in gas, liquid, or solid states, and to detect holes or voids in the solid phase. In general, experimental measurements made before 1960 are lacking in both precision and accuracy and will not be considered here. The significant experimental papers are:

1. Stewart [22412] who measured the dielectric constant for parahydrogen over a large range of pressure and temperature in both liquid and gas phases;
2. Younglove [52135] who measured the dielectric constant for highly compressed liquid and solid parahydrogen;
3. Hermans, et al. [32363] who measured for the dilute gas the differences in polarizability between H_2 , HD and D_2 , and also between normal and para H_2 ;
4. Kogan and Milenko [66761] who measured the difference in dielectric constant for various concentrations of o-p H_2 in the liquid state.

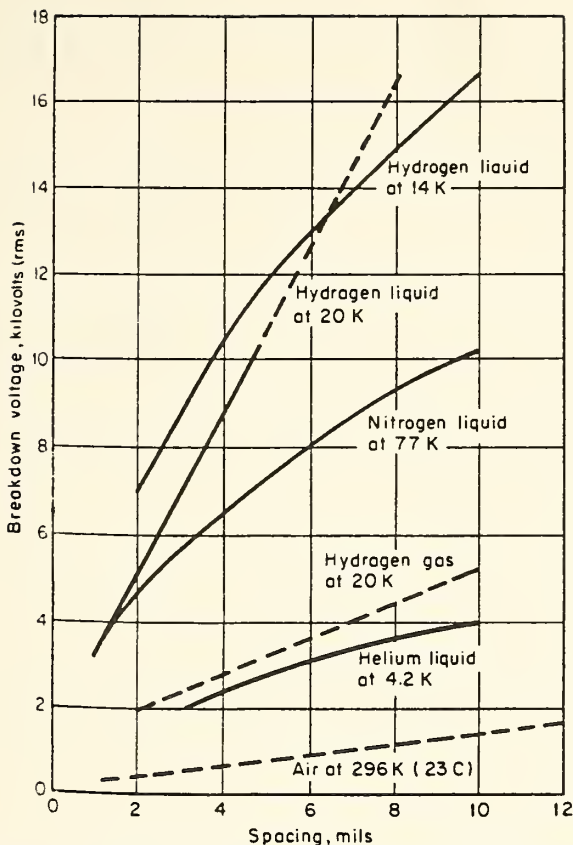


Figure 6-1. Breakdown Voltage of Cryogenic Liquids from K.N. Mathes, *Electro-Technol.* 72, 72 (Sept. 1963).

In addition we will use one of the older papers to indicate a difference in polarizability between H_2 and D_2 in the liquid state:

5. van Isterbeek and Spaepen [684].

It is important for the user to realize that correlation or analysis of the dielectric constant is almost always handled through the Clausius-Mossotti relation $M(\epsilon - 1)/(\epsilon + 2)\rho = P$, or solved for $\epsilon = (1 + 2P\rho)/(1 - P\rho)$, where ϵ is the dielectric constant, M the molecular weight, ρ the density, and P the molar polarizability. Polarizability is most often expressed in cm^3/g ; a nominal value for parahydrogen is $1.004 cm^3/g$, a nominal value for D_2 is $0.494 cm^3/g$. However, to compare polarizabilities of the various hydrogen isotopes it is best if P is expressed in molar units, thus the nominal value for parahydrogen becomes $2.02 cm^3/mol$ and the one for D_2 is $1.99 cm^3/mol$.

Parahydrogen

The most extensive measurements have been made on parahydrogen. A composite plot of the results, figure 6-2, shows that the polarizability is a very slowly varying function

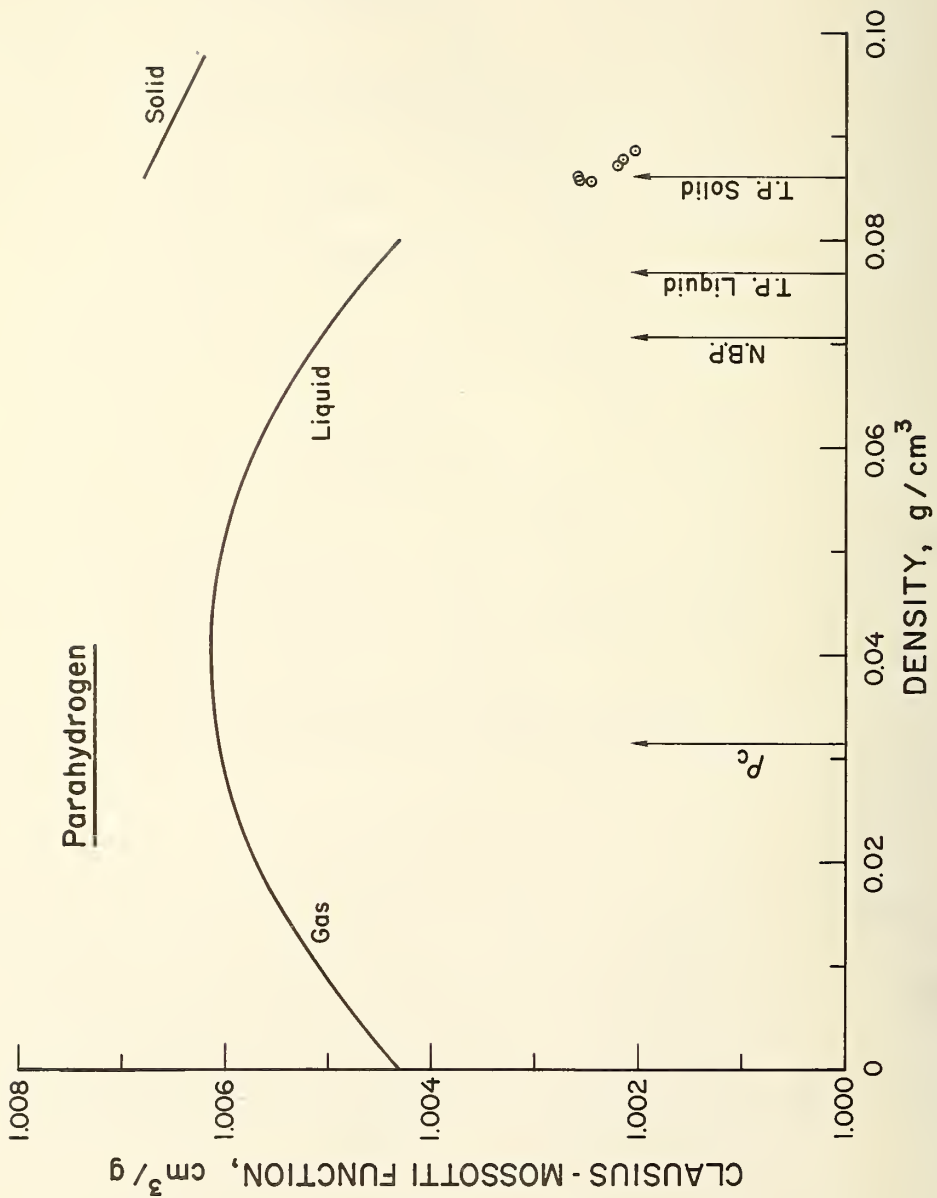


Figure 6-2. The Clausius-Mossotti Function for Parahydrogen

of density. Stewart [22412] is able to correlate his entire set of measurements with a single equation

$$1/P = 0.99575 - 0.09069\rho + 1.1227\rho^2$$

where P is in cm^3/g and ρ in g/cm^3 . Younglove's [52135] measurements extend to higher liquid densities and include values for the solid. He represents his solid measurements by the equation $P = 1.011 - 0.049091\rho$ with units as above. As shown in figure 6-2 Younglove reports a difference of about 0.5% in P between liquid and solid. This value has recently been called to question by Udovidchenko and Manzhelii [68779] and by Wallace and Meyer [Wallace and Meyer, 1972] who report a change of no more than 0.2%.

Since the dielectric constant is proportional to density, a plot of ϵ vs. T is very similar to a density-temperature phase diagram. For parahydrogen, liquid and solid boundaries are shown in figure 6-3 while the vapor-liquid boundary is shown in figure 6-4.

Other Modifications

The other experimental sources cited present dielectric constants for several hydrogen modifications, or differences in polarizability. We have used these data, recalculating as necessary, to prepare the table shown below, where all other modifications are referred to the polarizability of parahydrogen [Stewart, 22412]. For those modifications which have not been measured experimentally we have estimated polarizability differences as shown.

Table 6-1. Polarizabilities of the Hydrogens

molar polarizabilities, cm^3/mole		difference		conditions	source	
n- H_2	2.0273	p- H_2	2.0243	0.0030	dilute gas	Hermans et al.
n- H_2	2.0372	p- H_2	2.0279	0.0093	sat. liquid	Kogan et al.
HD	2.0130	p- H_2	2.0243	-0.0113	dilute gas	Hermans et al.
D_2	1.9988	p- H_2	2.0243	-0.0255	dilute gas	Hermans et al.
D_2	1.9910	p- H_2	2.0231	-0.0321	sat. liquid	van Itterbeek et al.
HT	1.995	p- H_2	2.0243	-0.029	estimated	plot of experimental
DT	1.978	p- H_2	2.0243	-0.046	estimated	polarizability dif-
T_2	1.961	p- H_2	2.0243	-0.063	estimated	ferences above vs. molecular weight

The significant conclusions drawn from this table are that

1. The differences in polarizability are nearly constant in going from gas to liquid states;
2. We can estimate values of the dielectric constant for other hydrogen modifications by using the polarizability differences and the known polarizability of parahydrogen in the

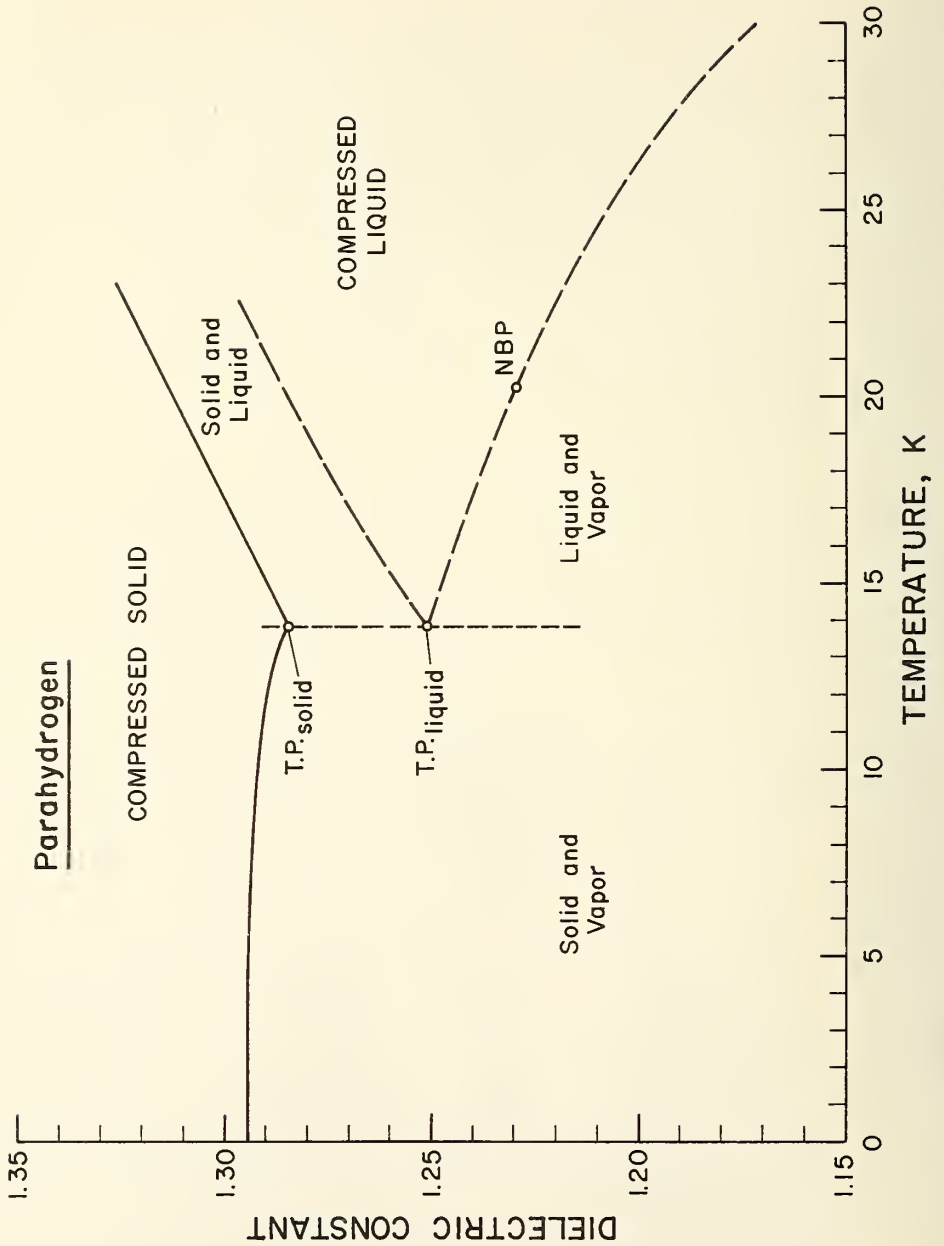


Figure 6-3. Parahydrogen, Dielectric Constant for Saturated Solid and Liquid

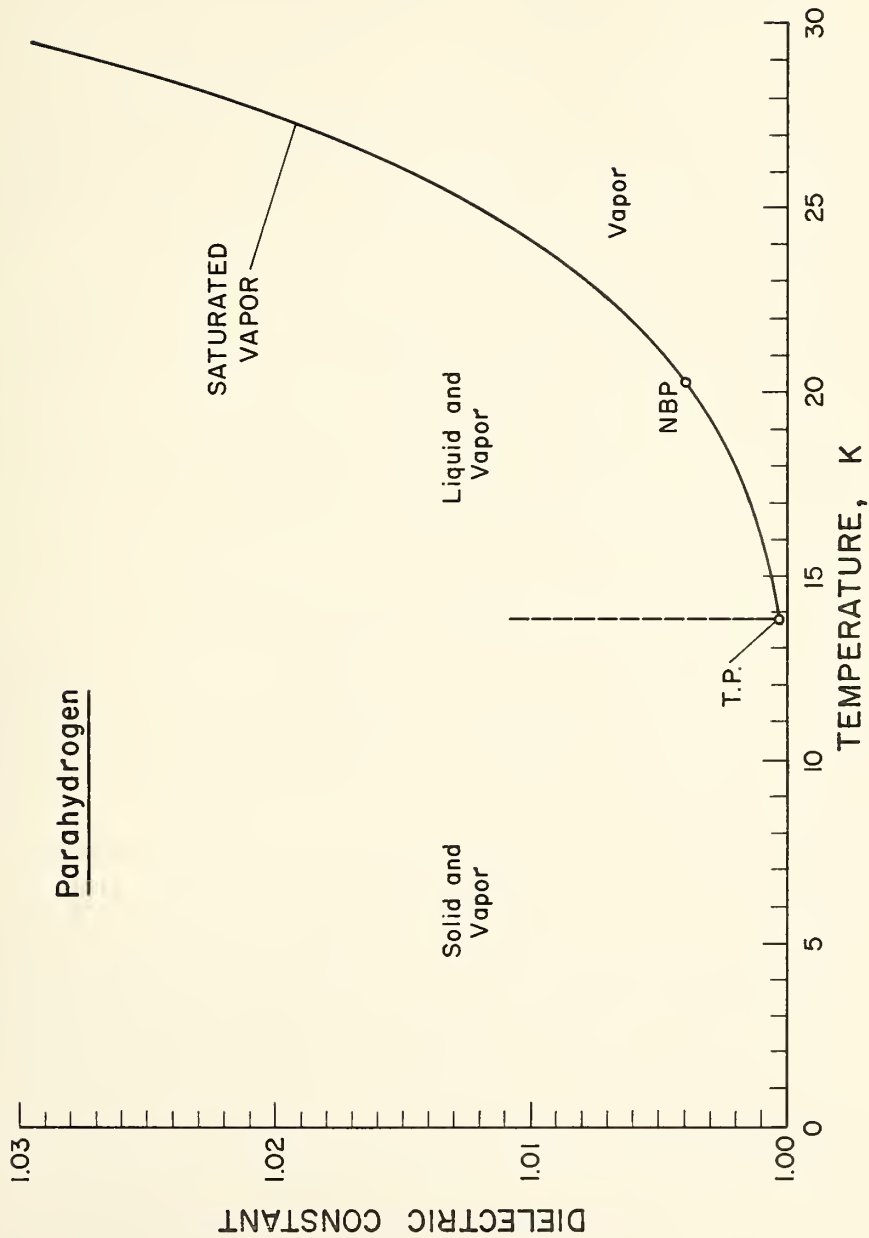


Figure 6-4. Parahydrogen, Dielectric Constant for Saturated Vapor

following sequence of steps:

- a. find density or state at point desired (moles/cm³)
- b. calculate polarizability of p-H₂ at an equivalent density (moles/cm³)
- c. adjust polarizability by appropriate difference
- d. calculate desired dielectric constant.

6.3 Electrical Conductivity

A significant experimental paper is that of Willis [40311]. His conclusions are: "It does not appear to be correct to speak of the electrical conductivity of cryogenic fluids in the sense of Ohm's law. Although a current can be passed through a cryogen, only a weak dependence on the applied gradient exists. In other words, the conductivity is not a constant as in an ohmic material, but a function of the applied potential. The cryogen acts as a constant current source in this respect. This is believed to be so because no intrinsic mechanism of charge carrier formation exists within the cryogen itself. All charge carriers present are generated by the ionizing radiation present as natural background." Note that the conductivity of D₂ is affected by a small amount of T₂. The T₂ contamination provides a large amount of ionization through the emission of beta particles. Values for hydrogen and deuterium are given in table 6-2.

Table 6-2. Electrical Resistivity of Hydrogen and Deuterium

Cryogen	Vapor pressure (mm Hg absolute)	Temperature (°R)	Electrostatic voltmeter measurement (Ωcm)	Electrometer amplifier measurement (Ωcm)
Deuterium	167	35	2.12×10^{18}	7.4×10^{15}
Hydrogen	585	34.8	$3.73, 10.4 \times 10^{18}$	4.6×10^{19}

7. Optical Properties

7.1 Index of Refraction

The refractive index of liquid hydrogen and deuterium is often required for the analysis of data obtained from high-energy physics experiments which use bubble chambers as detectors. The refractive index of hydrogen is also used in densitometry.

Index of refraction measurements are almost always analyzed in terms of the Lorentz-Lorenz function:

$$L-L = r_{\lambda} = \frac{n_{\lambda}^2 - 1}{n_{\lambda}^2 + 2} \frac{1}{\rho}$$

where r_{λ} is the specific refraction at wavelength λ , n_{λ} is the refractive index, and ρ is

the density. The Lorentz theory predicts that the Lorentz-Lorenz function should be nearly independent of temperature and density for a non-polar fluid. For many fluids including hydrogen the function is in fact weakly dependent upon temperature and density.

The specific refraction is related to the specific polarization p of the Clausius-Mossotti function (see section 6.2) as follows: $\epsilon = n_{\infty}^2$, which is equivalent to $p = r_{\infty}$. These relations imply that the temperature and density dependences of the Lorentz-Lorenz function and the Clausius-Mossotti function are similar, and that data of one type may be used to develop values for the other variable provided that the appropriate values of density are available.

Hydrogen

Diller [54546] obtained very precise values on gaseous and liquid para- and normal hydrogen at temperatures between 15 and 298.15 K and at pressures up to 230 atm. The data were analyzed in terms of the density and temperature dependence of the L-L function; the precision and reproducibility of L-L is better than 0.05% in most cases. Diller's results for parahydrogen are summarized in figure 7-1. The large deviations for saturated liquid and vapor near the critical density are attributable to errors in the density, not in the index of refraction. These results in fact indicate that a reexamination of the PVT surface of parahydrogen is in order, and that the value of the critical temperature should be close to 32.95 rather than 32.976. Diller also measured values for normal hydrogen. In general the normal-para differences in the L-L function are about 0.1%, very close to theoretical predictions. The differences for the saturated liquids vary a bit more than 0.1%. Values are shown in figure 7-2. In addition, this figure is perhaps the best graphical illustration of the differences in liquid density between normal and parahydrogen.

Deuterium

Values for liquid deuterium have been calculated by Childs and Diller [57921]. These authors used information on the dielectric constant following the method indicated by Corruccini [30251]. The required dispersion relationship was obtained from the room temperature measurements of the refractive index of deuterium by Larsén [Larsén, 1936]. The values obtained by Childs and Diller are based on the liquid densities of Prydz [43781]. They are tabulated as a function of wave length and density in table 7-1.

HD

Since the differences in dielectric constant between n and pH_2 , HD and D_2 are available (Hermans, et al. [32363]), and since Diller's difference in the Lorentz-Lorenz function between normal and parahydrogen is consistent with the polarizability difference of Hermans, et al., one could in principle obtain index of refraction values for HD. This has not yet been done.

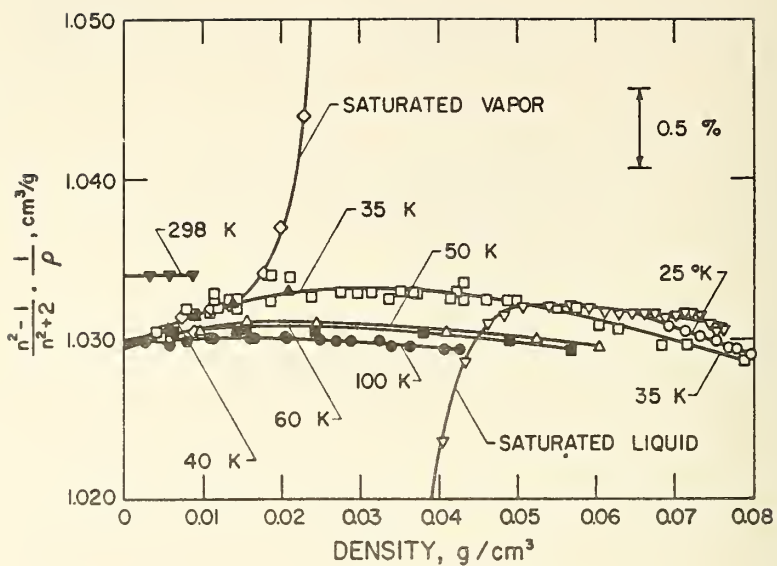


Figure 7-1. The Lorentz-Lorenz Function for Parahydrogen

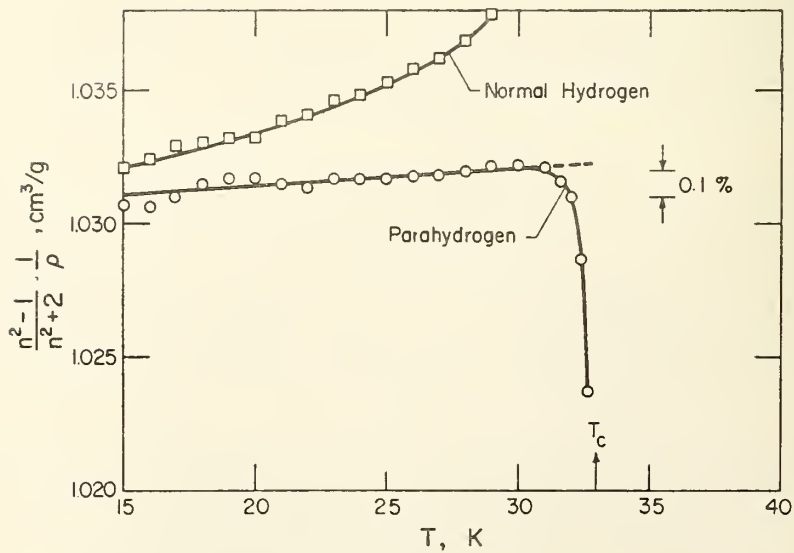


Figure 7-2. The Lorentz-Lorenz Function for Saturated Para and Normal Hydrogen

Table 7-1. The Refractive Index of Liquid Deuterium

T, °K	ρ , g/cm ³	$\lambda = 3200\text{\AA}$		$\lambda = 5461\text{\AA}$		$\lambda = 6328\text{\AA}$	
		r_λ , cm ³ /g	n_λ	r_λ , cm ³ /g	n_λ	r_λ , cm ³ /g	n_λ
18.7	0.1739	0.5358	1.1437	0.5089	1.1363	0.5056	1.1354
19.0	0.1733	0.5358	1.1432	0.5089	1.1358	0.5056	1.1349
20.0	0.1712	0.5358	1.1415	0.5090	1.1342	0.5057	1.1333
21.0	0.1691	0.5358	1.1397	0.5090	1.1324	0.5057	1.1316
22.0	0.1669	0.5359	1.1378	0.5091	1.1307	0.5057	1.1298
23.0	0.1645	0.5359	1.1358	0.5091	1.1288	0.5058	1.1279
24.0	0.1620	0.5360	1.1337	0.5091	1.1268	0.5058	1.1259
25.0	0.1595	0.5360	1.1315	0.5092	1.1247	0.5059	1.1239
26.0	0.1567	0.5360	1.1292	0.5092	1.1225	0.5059	1.1217
27.0	0.1538	0.5361	1.1267	0.5092	1.1202	0.5059	1.1194
28.0	0.1508	0.5361	1.1242	0.5093	1.1178	0.5060	1.1170
29.0	0.1475	0.5361	1.1214	0.5093	1.1152	0.5060	1.1144
30.0	0.1440	0.5362	1.1184	0.5094	1.1124	0.5061	1.1116

with permission from G. E. Childs, et al., Cryogenic Engineering Conference 15th, Los Angeles, Calif. (Jun. 1969), paper D-2.

7.2 Infrared Absorption

It is difficult to summarize this subject because so many transitions are possible in hydrogen, i. e., rotational, vibrational, Raman, ultra violet, infrared, far-infrared spectra all have been investigated. The survey uncovered about 150 references of potential interest. From a practical point of view it is of interest to describe the absorption of radiation in solid, liquid, or dense vapor. Unfortunately this becomes involved with system parameters related to the source of radiation. The spectral absorption coefficient for most gases is a strong function of frequency, being zero over large ranges and sharply peaked in certain bands. In the case of oxygen these bands have been identified and a correlation of total band absorbance given [section A-31 and A-42 in 80838]. A similar correlation for hydrogen has not been accomplished. Measurements on liquid hydrogen by Jones [63093, 65842] indicate that the infrared spectrum of liquid hydrogen consists of two distinct parts, each of which is strongly dependent on ortho-para composition. Comparison of these results to those of others show differences of 30 to 40% for the peak absorption. These discrepancies remain unexplained. Jones gives an example calculation for liquid normal and parahydrogen. He notes that the fundamental band of hydrogen is of negligible importance if we consider radiative heat transfer from a room temperature source. Simple definitions are given in [65842] and in [80838]. An extensive survey of induced transitions in liquid and solid hydrogen up to about 1965 is given by Colpa [36823].

8. Mechanical Properties

In earlier sections we have presented values for several mechanical properties such as the velocity of sound of the solid (section 3.4), bulk moduli for liquid (section 2.5), and thermal expansion of the solid (section 2.1). In this section we present the bulk of the mechanical properties of the solid phase.

Experimental measurements of uniaxial loading have been conducted by Bol'shutkin, Stetsenko and their co-workers on polycrystalline parahydrogen [49657], polycrystalline normal hydrogen [78545], and polycrystalline deuterium [68001]. In these papers the authors present values of Young's modulus, of the shear modulus, of tensile strength, of nominal yield stress, and of relative elongation. Values for normal and parahydrogen are summarized in figure 8-1 taken from [78545], and values for deuterium are presented in table 8-1 which is taken from [68001].

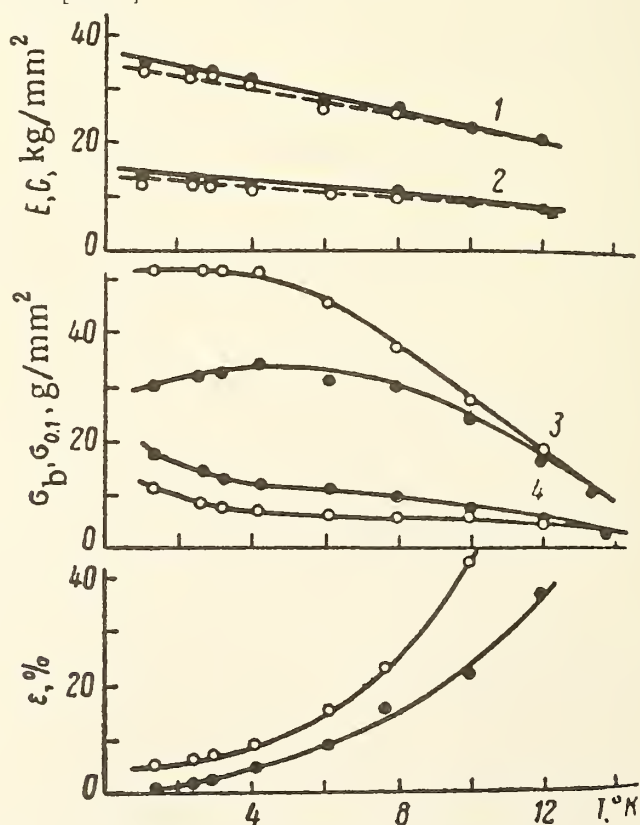


Figure 8-1. Temperature Dependences of the Mechanical Properties of Normal and Parahydrogen

1 - is Young's modulus, 2 - the shear modulus, 3 - tensile strength, 4 - nominal yield stress, ϵ relative elongation.

Table 8-1. Temperature Dependence of Strength, Relative Elongation, Coefficient of Hardening, Young's Modulus, and Shear Modulus of Deuterium on Temperature

T, °K	σ_b , g/mm ²	ϵ , %	$\sigma_{0.1}$	E kg/mm ²	G
1.4	43.0	0.6	51.0	52.0	20.0
3.0	50.0	0.8	34.0	49.6	19.4
4.2	54.0	1.0	29.0	48.2	18.1
5.4	52.0	2.0	27.5	46.5	17.8
8.0	45.0	3.0	26.0	44.0	16.8
11.6	43.3	6.2	22.2	42.0	16.2
15.6	29.1	13.0	14.4	41.1	15.8
16.4	21.4	22.3			

Experimental measurements of the velocity of sound in solid hydrogen and deuterium [78548, 81564] have yielded values of Poisson's ratio as well as values of "adiabatic" and isothermal compressibilities. The values have already been presented in table 3-5. In addition, these papers report values of the Debye θ , of the elastic constants, and of the adiabatic bulk modulus, all of which together with a few calorimetric θ 's are given in table 8-2 below.

Table 8-2. Debye θ , Adiabatic Bulk Modulus and Elastic Constants for H₂ and D₂

	Debye θ		adiabatic bulk modulus [81564]		
normal H ₂	111, single crystal normal [81564]		1740 bar		
	113, polycrystal para [78548]				
	124, para calorimetric [21136]		assuming V = 22.87 cm ³ /mol		
	122, para calorimetric [75211]				
normal D ₂	105, single crystal normal [81564]		3370 bar		
	113.8, ortho calorimetric [75211]		assuming V = 20.19 cm ³ /mol		
Elastic constants [81564], ρ = density					
	C_{11}/ρ	C_{12}/ρ	C_{13}/ρ	C_{33}/ρ	C_{44}/ρ
normal H ₂	4.10	1.35	0.45	4.99	0.94
normal D ₂	3.34	1.17	0.61	3.91	0.80

Note that the calorimetric Debye θ 's are consistent, and the mechanical Debye θ 's are consistent, but that the two methods differ considerably as pointed out in [78548]. The explanation that "This discrepancy is due to incorrect elimination of the anomalies in the temperature dependence of $C_v \dots$ " needs further study, particularly as the recent results of Roberts and Daunt [75211] are in excellent agreement with those of Ahlers [21136]. In addition Ahlers' measurements are at constant volume; technically the correct way to determine a Debye θ is from C_v . Finally Ahlers' θ 's vary from 128 to 106 for his lowest isochore, that is for temperatures from 0 to 15 K, and they vary considerably more for changes in density.

Finally there are measurements of shear strength as a function of applied pressure. Initial values of extrusion pressures were reported by Stewart [5719], 240 kg/cm² for H₂ and 290 kg/cm² for D₂. The measurements have since been refined by Towle [32738] who reports the following values for H₂ at 4 K:

shear strength kg/cm ²	29	41.6	61.4	64.6
mean applied pressure kg/cm ²	1050	2010	3190	3710

9. Modification, Phase Change

9.1 Ortho-Para Conversion

A separate search on ortho-para conversion turned up 241 papers of potential interest. Time and funding for this survey precluded any work beyond that done for the earlier survey [Ludtke and Roder, 78262]. Since many of the references on the earlier report on deuterium also are applicable to hydrogen, we rely heavily on the material of that report.

Hydrogen, deuterium and tritium exist in two modifications. In one the molecule has a rotational energy level $J = 0$, the lowest energy level, with an associated spin quantum number $I = 0$ or 2. In the other state the molecule has a rotational energy level $J = 1$ and a spin quantum number $I = 1$. For hydrogen and tritium the lowest energy level is called para, for deuterium it is called ortho.

We suggest that the heat effects of the ortho-para change can be ignored in most practical applications if the absence of a catalyst can be assured. If not, then we recommend deliberate conversion to the lower energy level as the step to minimize all undesirable side effects. If estimates of ortho-para effects are desired, we suggest that the rate equations of Motizuki and Nagamiya [5439, 5440] be used.

Ortho-Para Composition

The ortho-para composition of hydrogen, deuterium and tritium at equilibrium is temperature dependent, especially below 50 K, as shown in table 9-1, which is taken from Woolley, et al. [6368] and Jones [18037].

Table 9-1. Ortho-Para Composition at Equilibrium

Temperature (K)	Percentage in para form for H ₂	Percentage in para form for D ₂	Percentage in para form for T ₂
10	99.9999	0.0277	97.177
20	99.821	1.998	66.186
20.39	99.789	---	---
23.57	---	3.761	---
30	97.021	7.864	43.306
40	88.727	14.784	33.197
50	77.054	20.718	28.728

Heat of Conversion

The heat of conversion arises from the transition between the different rotational levels of the molecule. When for example para deuterium converts to ortho deuterium, there is a decrease in the energy of the molecule and this energy is dissipated as heat. The amount of heat given off in a conversion process depends on the temperature and on the amount of material requiring conversion. Values of the heat of conversion at a given temperature level should be obtained from the tables of ideal gas thermodynamic functions, Haar, et al. [11271] for hydrogen and deuterium, and Jones [18037] for tritium.

Rate of Self-Conversion

The rate of self-conversion is of particular interest in practical problems where the normal gas, i. e., the gas in equilibrium at room temperature, is liquefied or solidified and self-conversion towards the equilibrium concentration sets in. The earliest vapor pressure papers contain reference to the problem.

For liquid deuterium two order of magnitude estimates for the rate of self-conversion have been published by Brickwedde, et al. [11250] and by Woolley, et al. [6368]. Both are based on the rate of conversion given for normal hydrogen by Woolley, et al. [6368]. The rate of self-conversion of deuterium is plotted in figure 9-1. The rate equations are assumed to be second order differential equations of the form

$$\frac{d[\text{para deuterium}]}{dt} = -K [\text{para deuterium}]^2 ,$$

where K is the velocity constant, and [para deuterium] indicates the mole fraction of para deuterium.

For solid deuterium, Motizuki [Motizuki, 1957, 1962] calculated the rate of para-ortho self-conversion. He assumes that the para-ortho conversion arises from magnetic dipole-dipole

interactions of molecular pairs and the coupling of the nuclear quadrupole of one molecule with the total quadrupole moment of the other. His rate equation is

$$\frac{d[p-D_2]}{dt} = -K[p-D_2]^2 - K'[p-D_2](1 - [p-D_2]) ,$$

where $K' = 1.78 \times 10^{-3} \text{ h}^{-1}$ and $K = 1.27 \times 10^{-3} \text{ h}^{-1}$. Motizuki's rate of self-conversion of solid deuterium is plotted in figure 9-1 for comparison with the liquid estimates. Half-conversion times for the various expressions are also indicated in figure 9-1. A half-conversion time is defined as the time required for 50% of the original para composition (33.33% for this plot) to convert to ortho. As the plot indicates, the half-conversion times for self-conversion of deuterium are extremely long (> 5400 hours).

Grenier and White [22677] measured self-conversion rates of deuterium from the drift curves of their heat capacity measurements. The data are for solid deuterium and cover a wide range of para compositions (33 - 83%). The experimental rates, $d[p-D_2]/dt$, and Motizuki's theoretical rate are compared in figure 9-2. Grenier explains that the large scatter in the data may be due to traces of oxygen that increase the rate of conversion in some of the samples.

For solid hydrogen Ahlers [22667] made measurements of the ortho-para conversion as a function of the molar volume of the solid. These measurements are in excellent agreement with the experimental values obtained at very low pressure by Cremer [Cremer, 1938].

Rate of Conversion of Liquid Using Catalysts

Any of the hydrogens may be readily converted to the equilibrium composition with a catalyst, if conversion is desired. Some very effective catalysts exist. The catalyzed conversion rate for deuterium is comparable to that of hydrogen. It is extremely fast when compared to the self-conversion rate. No special effort was made to gather all references on the conversion using catalysts; the papers mentioned below are included only to indicate the type of information that is available.

An historical work in the field is Farkas' book [2040]. He measured the rate of conversion for deuterium gas at 20.4 K, using activated charcoal and solid oxygen as catalysts. Using solid oxygen, he obtained half-conversion times of 19 minutes, and with activated charcoal, the rate of conversion was too rapid to be measured.

Eberhart [25360] conducted conversion experiments with several catalysts. The catalysts were of the chromia impregnated γ -alumina type. The half-conversion times for deuterium at 20.4 K ranged from 11 to 84 minutes. For hydrogen, the results ranged from 6.7 to 123 minutes.

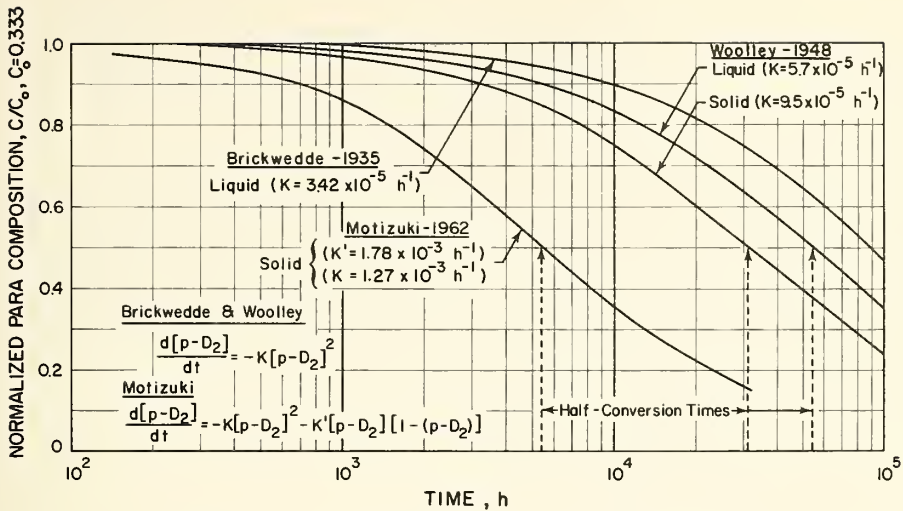


Figure 9-1. Rates of Self-Conversion of Deuterium

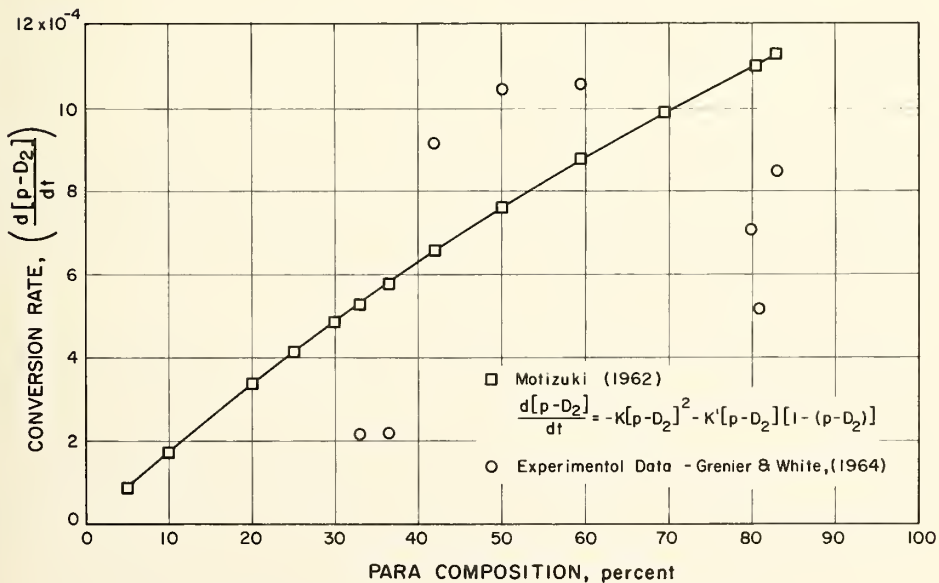


Figure 9-2. Self-Conversion of Solid Deuterium

Albers, Harteck, and Reeves [23788] measured the half-conversion times of tritium, deuterium, and hydrogen gas on coconut charcoal. They found the half-conversion times to be 1 minute for tritium, 19 minutes for deuterium, and 27 minutes for hydrogen.

9.2 Melting Curves

The experimental papers up to 1948 have been summarized by Woolley et al. [6368]. An important paper is that of Mills and Grilly [5449] who measured melting pressures for n-H₂, n-D₂, and n-T₂. For p-H₂ Goodwin [14130] has presented experimental results and an empirical equation. We have taken the values in composite table 9-2 from Goodwin's calculation because he constrained the Simon equation to pass through a selected triple point, thus, negative values in the range of validity of the curve are avoided. The values for HD are taken from Woolley et al. [6368].

Goodwin's equation reads as follows:

$$(P - P_t)/(T - T_t) = A \exp(-\alpha/T) + BT + C$$

where $A = 30.3312 \text{ atm/K}$, $\alpha = 5.693 \text{ K}$, $B = 2/3 \text{ atm/K}^2$, and values of the triple point parameters and the constant C are as follows:

	p-H ₂	n-H ₂	n-D ₂	n-T ₂
C (atm/K)	0	0	3.85	6.4
T _t (K)	13.803	13.947	18.72	20.61
P _t (atm)	0.0695	0.071	0.169	0.213

For parahydrogen, the values in table 9-2 repeat those already given in table 3-2 where the values for the density of the freezing liquid were presented. Additional values for the melting pressures of parahydrogen have been published by Younglove [52135]; these values have not yet been incorporated into a melting pressure analysis.

9.3 Latent Heat of Fusion

The latent heats of fusion are summarized in table 9-3. Most of the experimental work was conducted at essentially triple point temperatures; however some measurements have been made at elevated pressures, and others as a function of ortho-para composition. Grenier and White for example concluded that the heat of fusion of deuterium with para content in the range 78% to 81% is $47.18 \pm 0.10 \text{ cal/mol}$. They also indicate that the heats of fusion are independent of ortho-para composition within experimental error. If the authors indicated the triple point temperature in the paper, then the value of that temperature is given in brackets in the temperature column. For the measurements at elevated pressures, both the temperature and the pressure at which the measurements were made are shown. In the heat of fusion column the values which are underlined are the ones which are given in section 11 in the fixed point tables.

Table 9-2. Melting Pressures of the Hydrogens

Temperature K	p-H ₂ atm	n-H ₂ atm	n-D ₂ atm	n-T ₂ atm	Temperature K	HD P kg/cm ²
13.803	0.0695				16.60	0.13
13.947	4.312	0.071				
14.	5.887	1.64				
15.	36.880	32.45				
16.	70.191	65.60				
17.	105.676	100.9				
18.	143.217	138.3			17.	14.2
18.720			0.169			
19.	182.717	177.7	11.09			
20.	224.096	218.9	51.37			
20.610				0.213		
21.	267.286	261.9	93.60		18.	52.6
22.	312.230	306.8	137.7			
23.	358.880	353.3	183.6			
24.	407.196	401.5	231.3			
26.	508.684	502.7	331.8			
30.	616.466	723.9	552.2		19.	92.9
35.	730.37	1034	862.4			
40.	976.02	1380	1209			
45.	1387.8	1762	1592			
50.	2186.4	2178	2010			

Table 9-3. The Latent Heat of Fusion

Hydrogen modification	Author	Year	Reference	Temperature K	ΔH fusion
n-H ₂	Simon and Lange	1923	[6375]	T. P. (13.93)	<u>28.0</u> \pm 0.15 cal/mol
94% p-H ₂	Clusius and Hiller	1929	[426]	T. P. (13.95)	28.03 \pm 0.4 cal/mol
99.8% p-H ₂	Johnston, et al.	1950	[9219]	T. P. (13.845)	<u>28.08</u> * \pm 0.15 cal/mol
99.79% p-H ₂	Ahlers	1963	[21136]	T. P. (13.803)	<u>118.</u> \pm 1 J/mol
99.79% p-H ₂	Dwyer, et al.	1965	[30327]	T. P. (13.803)	27.75 cal/mol
				T. P. (13.803)	28.00 cal/mol
				13.808 (0.203 atm)	27.87 cal/mol
				14.653 (25.85 atm)	29.37 cal/mol
				15.535 (54.4 atm)	30.80 cal/mol
				17.813 (136.05 atm)	34.21 cal/mol
				19.481 (202.38 atm)	36.95 cal/mol
				19.522 (204.08 atm)	36.91 cal/mol
				21.186 (275.51 atm)	40.28 cal/mol
				(338.10 atm)	42.87 cal/mol
normal D ₂	Clusius and Bartholome	1935	[5595]	T. P. (18.65)	<u>47.0</u> * \pm 0.4 cal/mol
ortho D ₂	Kerr, Rifkin and Johnston	1951	[645]	T. P. (18.63)	<u>47.07</u> * \pm 0.1 cal/mol
80.7% para D ₂	Grenier and White	1964	[22677]	T. P. (18.787)	
78.7% para D ₂				(18.781)	
78.5% para D ₂				(18.698)	
78.4% para D ₂				(18.626)	
HD	Brickwedde and Scott, as reported by Woolley, et al.	1948	[6368]	T. P. (16.604)	<u>38.1</u> cal/mol

* Average values for more than one run.

9.4 Latent Heat of Sublimation

The latent heat of sublimation like the heat of vaporization is temperature dependent. As far as we are aware, actual experimental measurements of the latent heat of sublimation have not been conducted. Instead the property is usually calculated from the Clausius-Clapeyron equation requiring values of the solid volumes, the vapor volumes, and the derivative of the vapor pressure curve. The difficulty is usually to select a proper equation of state for the vapor, because the vapor volumes predominate in the calculation. For the temperatures involved the extrapolation of the virial B is quite extended, therefore the ideal gas equation is often used for the vapor volumes. At the triple point the latent heat of sublimation should equal the sum of the latent heat of vaporization and the latent heat of fusion. The equality offers a severe test of thermodynamic consistency, in particular, of the various vapor pressure derivatives used in the calculations.

The heat of sublimation at 0 K is of particular interest theoretically, and a number of papers are concerned with this extrapolation. Clusius, for example, in a survey article [16376] offers calculated or extrapolated values of 183.4 cal/mol for H_2 , 228 cal/mol for HD, and 274 cal/mol for D_2 at 0 K. More recently Eyring, et al. [19187] have calculated the following values from the hole theory of liquids:

181.56 cal/mol	for p- H_2
182.92	n- H_2
234.57	H-D
276.32	o- D_2
277.90	n- D_2

The only modern thermodynamically consistent calculation, which by the way included data on the heat capacity of the solid, is that by Mullins, et al. [12596] on parahydrogen. Values of the heat of sublimation for parahydrogen have already been presented in the composite table 2-3.

9.5 Latent Heat of Vaporization

The heat of vaporization is a function of temperature. If sufficiently precise values of the saturated liquid and vapor densities and the derivative of the vapor pressure are available, then one can calculate the heat of vaporization from the Clausius-Clapeyron equation. For parahydrogen and normal deuterium, the heat of vaporization can be established directly from the enthalpies of saturated liquid and vapor given in tables 2-2 and 2-7. For the other hydrogens, values near the normal boiling point are collected in table 9-4 as available. A comparison between normal hydrogen and parahydrogen has been calculated from the values given by Stewart and Roder [23790] and is presented in table 9-5. A

Table 9-4. The Latent Heat of Vaporization

Hydrogen modification	Author	Temperature K	ΔH vaporization
n-H ₂	Simon and Lange	1923 [6375] n. b. p. - 20.35	215.9 \pm 1.1 cal/mol
	Clusius and Bartholomé	1935 [5595] 19.65 (194.5 mm)	219.7 cal/mol
	Johnston, et al.	1950 [9219] 20.261	219.3* cal/mol
n-D ₂	Clusius and Bartholomé	1935 [5595] 19.65	302.3 cal/mol
	Kerr, et al.	1951 [645] n. b. p. - 23.59	293.93 cal/mol
T ₂	Grilly	[444] n. b. p. - 25.04	333 cal/mol
HD	Brickwedde and Scott as reported by		
	Woolley, et al.	[6368] 22.54	257 cal/mol

* calculated from a measured value of p-H₂.

method for estimating desired values of the heat of fusion, both as a function of temperature and by species, has been indicated by White, et al. [9624], and the pertinent graph is reproduced in figure 9-3. This method, reciprocal of the square root of the mass, has been criticized and other mixing rules are preferable [see for example, 32140], but it is the only one that has been worked out at present.

Table 9-5. Latent Heats for n- and p-H₂

T, K	Latent Heat of Vaporization, cal/mol		
	Normal	Para	Difference
14	219.9	217.1	2.8
15	220.7	218.3	2.4
16	221.1	218.5	2.6
17	221.1	218.4	2.7
18	220.6	217.9	2.7
19	219.6	216.8	2.8
20	218.0	215.2	2.8
21	215.7	212.5	3.2
22	212.7	209.5	3.2
23	208.9	205.6	3.3
24	204.2	200.8	3.4
25	198.5	195.0	3.5
26	191.5	187.8	3.7
27	183.1	179.2	3.9
28	173.0	168.7	4.3
29	160.6	155.8	4.8
30	145.2	140.1	5.1
31	125.4	119.8	5.6
32		90.8	

9.6 Solid Transitions

Solid transitions are known to occur for H₂ and D₂. The transition is dependent on ortho-para composition and involves a change in structure from hcp to bcc. Typical transition temperatures range from 1.2 to 4 K. The subject is covered in more detail in section 2.1.

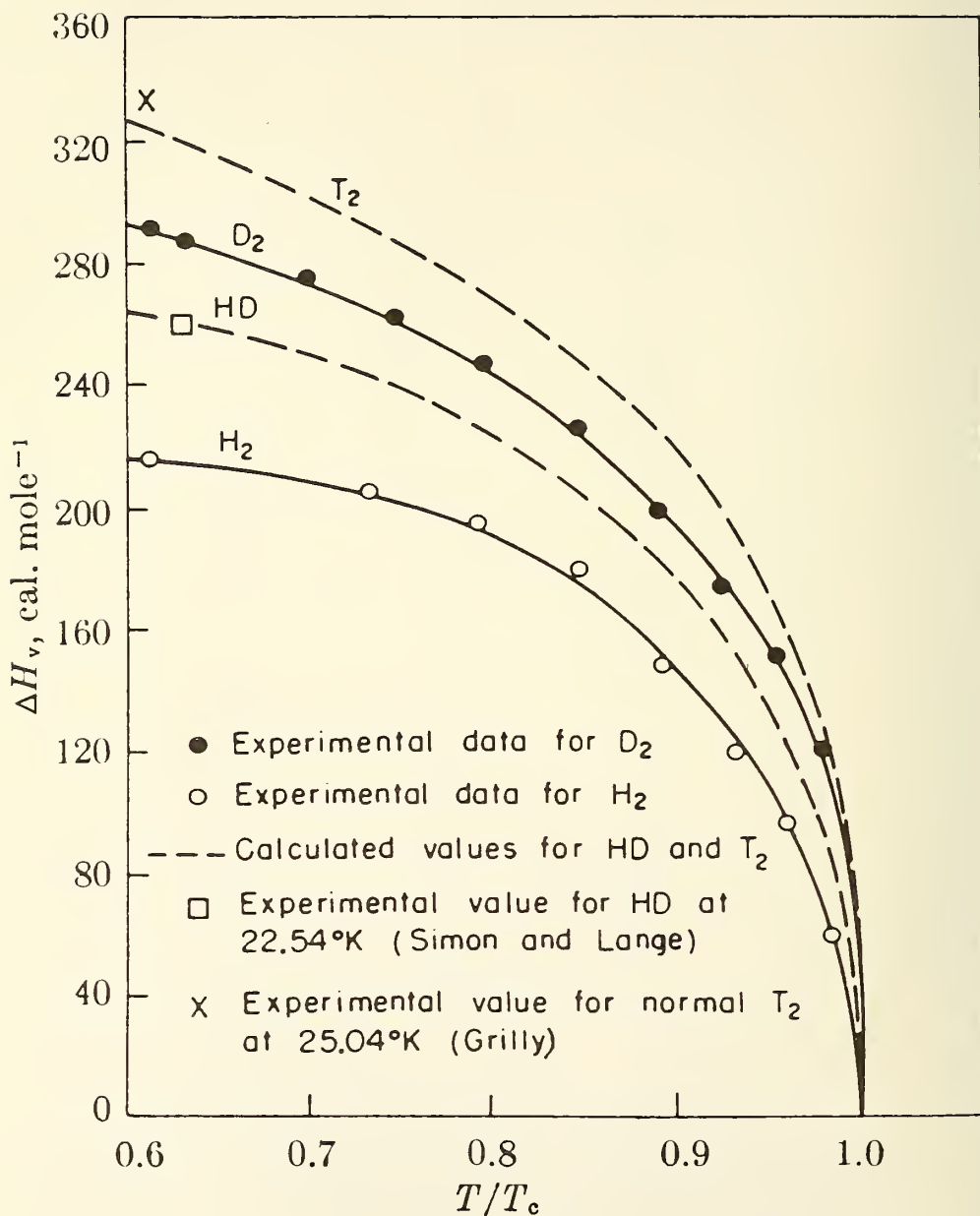


Figure 9-3. Diagram for Estimating Heats of Vaporization with permission from D. White, et al., J. Phys. Chem. 63, 1181 (1959).

10. Vapor Pressure

Vapor pressure measurements on liquid and solid hydrogen, its isotopes, and its isotopic mixtures have been made since 1900. However, for the purposes of this report, all measurements or correlations prior to 1948 are not discussed. We note that differences in vapor pressure measurements by different observers are nearly always attributable to differences in the temperature scales used.

10.1 Hydrogen

Solid p-H₂

For practical purposes 20.4 e-H₂, which is 99.79% para, is the same as para-hydrogen. We recommend the calculation by Mullins, et al. [12596], the values of which have already been presented in table 2-3.

Solid n-H₂

Direct experimental vapor pressure measurements on solid n-H₂ between 3.4 and 4.5 K were made by Borovik, Grishin, and Grishina [6759]. Hydrogen gas was condensed on a cooled surface in a high vacuum. After establishment of equilibrium, the pressure was measured at which the rate of condensation and the rate of evaporation were equal. Errors in the measurement of temperature did not exceed ± 0.02 K, and pressure measurement error did not exceed 10% as reported by the authors. Results are given in table 10-1.

Harrison, Fite, and Guthrie [13770] experimentally determined the vapor pressure of solid n-H₂ from 4.7 K to 11.1 K. For the two experimental temperature ranges, 4.7 - 5.2 K and 6.5 - 11.1 K, the pressure readings were taken at room temperature using a gauge connected by a tube to the low temperature region containing the solid hydrogen, and corrections were made in the calculations when necessary. The data are given in table 10-1. The authors represent the data by the equation

$$\log P = -\frac{43.39}{T} + \frac{5}{2} \log T + 2.047$$

which is valid for the temperature range 4.7 to 13.95 K.

Liquid p-H₂

For maximum consistency with the thermal data such as entropy, enthalpy, etc., we recommend the vapor pressures given by Roder, et al. [29210] already presented in table 2-2. The values are identical to those given by Weber, et al. [13704] and are in very good agreement with those of Hoge and Arnold [454] and those of Woolley, et al. [6368].

The measurements by Hoge and Arnold [454] on liquid equilibrium hydrogen (0.979 p-H₂, 0.021 o-H₂ at 20.4 K) were made from near the triple point to the critical point. They noted that while the e-H₂ was in ortho-para equilibrium only at 20.4 K, the composition did not change as the temperature was raised or lowered during the experiment. Their smoothed experimental data have an accuracy of 0.3 mm Hg up to 1 atm, and the uncertainty above 1 atm pressure gradually increases, reaching ± 8 mm Hg near the critical point. The uncertainty in the temperature scale is perhaps ± 20 mK.

Weber, et al. [13704] presented experimental data on 20.4 K equilibrium H₂ (called parahydrogen) from the boiling point to the critical point. The measurements were done in a P-V-T cryostat, and the overall uncertainty of the pressure measuring system was ± 0.003 atm. The authors present the data with the following equation valid between the boiling point and 29 K,

$$\log P_a \text{ (atm)} = 2.000620 - \frac{50.09708}{T + 1.0044} + 0.01748495T .$$

Above 29 K an adjustment function must be used:

$$P = P_a + 0.001317 (T-29)^3 - 0.00005926 (T-29)^5 + 0.000003913 (T-29)^7$$

The function

$$\log P \text{ (atm)} = 1.772454 - \frac{44.36888}{T} + 0.02055468T$$

was developed by Weber, et al. to represent the values of Hoge and Arnold within experimental deviations from the triple point to about 21 K. The equations join smoothly at 20.268 K.

Liquid n-H₂

Van Itterbeek, et al. [23435] have made the most recent and most extensive measurements of the vapor pressure of n-H₂ and the difference in vapor pressure between normal- and equilibrium (para) hydrogen in the temperature range from 20 K up to 32 K. The data for n-H₂ are shown in table 10-1. The difference of normal- and para-H₂ were smoothed by the function

$$\log P \text{ (kg/cm}^2\text{)} = A + B/T + C \log T$$

where the constants for the n-H₂ are: A = -1.55447, B = -31.875, C = 2.39188; and the constants for the differential pressure are: A = -3.945, B = -14.520, and C = 2.470. The normal boiling point data for n- and e-H₂ were determined from the equation and are in agreement with accepted values within the limit of experimental error. Most of the experimental data in table 10-1 agree with smoothed data calculated from the equations; however above 30 K the equation for the vapor pressure differences is not reliable.

For values between the triple point and the normal boiling point we recommend the values given in the compilation of Woolley, et al. [6368].

Liquid o-H₂

For liquid o-H₂, the only data available are the estimates of Woolley, et al. [6368] who measured values for liquid e-H₂ and liquid n-H₂. The values for o-H₂ are based on an extrapolation with respect to composition; they are given in table 10-1.

10.2 Deuterium

Solid D₂

In accordance with the earlier survey of Ludtke and Roder [78262] we recommend the equations by Woolley, et al. [6368] for the vapor pressure of solid D₂. The equations are:

$$\text{for solid n-D}_2 \quad \log_{10} P = 5.1626 - \frac{68.0782}{T} + 0.03110T ,$$

$$\text{for solid o-D}_2 \quad \log_{10} P = 5.1625 - \frac{67.9119}{T} + 0.03102T .$$

Calculated values for n-D₂ are given in table 10-1.

Liquid D₂

For liquid normal deuterium we must recommend the vapor pressures by Prydz [43781] in order to be consistent with all of the other thermal quantities presented. Values for the vapor pressure of liquid normal deuterium have already been given in table 2-7.

For liquid ortho deuterium, the most extensive measurements are those of Hoge and Arnold [454] whose values are given in table 10-1. The authors indicate that the vapor pressure measurements on deuterium below one atmosphere are accurate to 0.3 mm Hg and that the uncertainty in temperature is ± 20 millidegrees K. The authors also determined the triple point temperature and pressure of ortho deuterium as well as the normal boiling point. The values agree with those of Woolley, et al. [6368] for triple point temperature and pressure, but disagree for the normal boiling temperature. A significant difference

between this work and others is the use of a platinum resistance thermometer to determine temperatures.

Meckstroth and White [71082] measured the vapor pressure differences between ortho deuterium (2.15% para) and varying compositions of para deuterium (up to 80%), over the temperature range from 17.2 to 22 K. They explain that relatively few reliable vapor pressure measurements were obtained for the solid due to plugging of the small capillary tubes whenever the apparatus was maintained below 17 K for any appreciable length of time. Their results are shown in figure 10-1 along with those of Brickwedde, et al. [11250]. The older studies were limited to para compositions in the range 2.1 to 33.3 percent. For the newer results the major impurity of the deuterium samples never exceeded 0.1 mole percent, the change in ortho-para composition during any run was less than 0.4 percent.

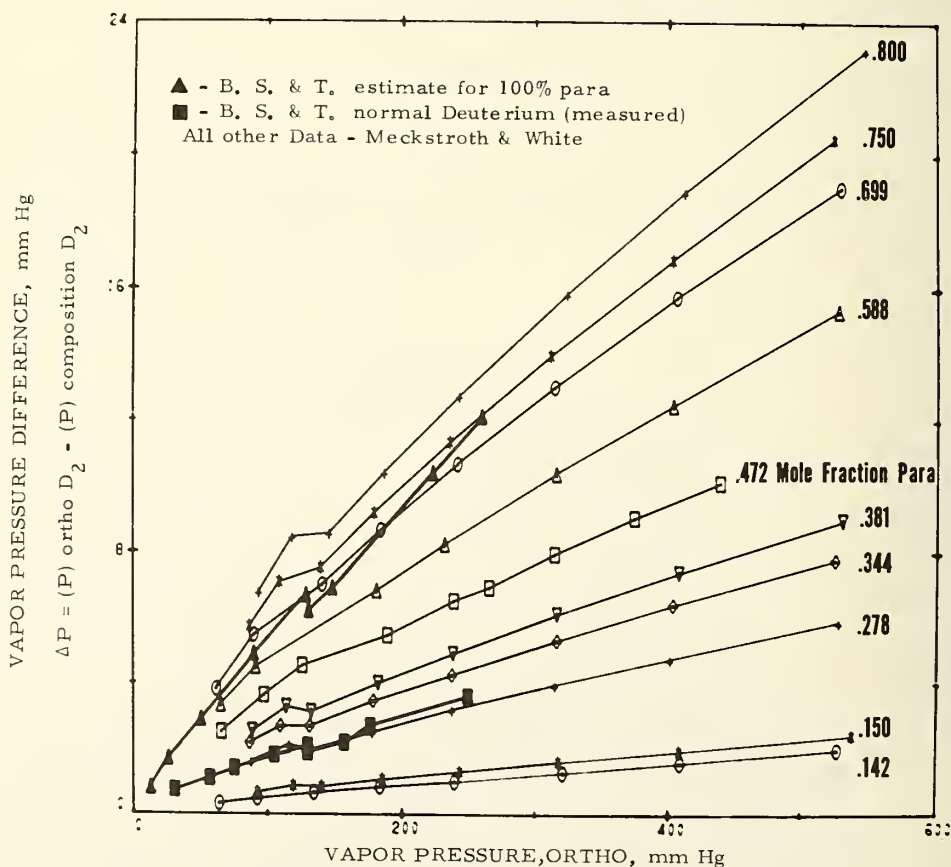


Figure 10-1. Vapor Pressure Differences vs. the Vapor Pressure of Ortho Deuterium

10.3 Tritium

Experimental determination on the vapor pressure of solid and liquid tritium between 14 and 29 K was made by Grilly [444], who also measured hydrogen and deuterium. The author felt that the purity was sufficient to yield fair accuracy in the temperature range. The data are reproduced in table 10-1. The author represented the data by the following two equations:

$$\text{solid:} \quad \log P = 6.4773 - 88.002/T$$

$$\text{liquid:} \quad \log P = 6.0334 - 78.925/T + 0.0002 (T-25)^2$$

Maximum deviation for the solid data is 3.8%, whereas for the liquid it is 0.34%.

10.4 Hydrogen Deuteride

Woolley, et al. [6368] measured the vapor pressure of HD from 10.4 to 20.4 K. For solid HD they used the following equation,

$$\log P = 4.70260 - \frac{56.7154}{T} + 0.04101T$$

Calculated values are given in table 10-1.

Hoge and Arnold [454] made extensive measurements of the vapor pressure of liquid HD. Their smoothed data are given in table 10-1.

10.5 Hydrogen Tritide and Deuterium Tritide

No experimental measurements on solid HT or DT were found in this survey, therefore no recommendations can be made for the solid vapor pressures. For liquid HT and DT only the single values at 20.4 K obtained by Libby and Barter [14954] from the evaporation of very dilute solutions are available. The values are HT: 254 ± 16 mm Hg and DT: 123 ± 6 mm Hg. This experiment indicates that the vapor pressure of HT is approximately the same as that of D_2 . A subsequent experiment by Bigeleisen and Kerr [417] showed that the vapor pressure of HT is greater than that of D_2 at 20 K. One interpretation of the latter experiment is to lend credence to the rule of the geometric mean for vapor pressures. It is the geometric mean that is used by Mittelhauser and Thodos [32140, see also summary] to calculate estimates of vapor pressure for these species.

10.6 Summary

Except for parahydrogen the most important experimental vapor pressures are reproduced in table 10-1. A composite picture of the vapor pressure curves of the hydrogens based on the sources selected above is given in figure 10-2. The selection of triple point pressure and temperature, of the normal boiling temperature, and of the critical parameters, which are presented in section 11, is also based on the vapor pressures selected here.

As an alternate, particularly if highly precise values are not required, we include a table of values calculated by Mittelhauser and Thodos [32140]. The table, 10-2, is highly compact and convenient. It is based on an evaluation of very nearly the same vapor pressure data selected above. However, the user should keep in mind the following:

1. The vapor pressure equation, the Frost-Kalkwarf equation, is singularly awkward to use, it is transcendental in nature.
2. Vapor pressure accuracy is on the order of $\pm 1\%$ in pressure. The departures are analyzed in terms of pressure, not in terms of temperature, while the major discrepancies, as noted in the introduction, are ordinarily caused by use of different temperature scales.
3. The critical constants used for para and normal hydrogen are much too high. As the method is essentially a corresponding states analysis, the critical points of the other hydrogens are suspected to be too high.

Table 10-1. Recommended Vapor Pressures of the Hydrogens

p-H ₂ solid	n-H ₂ solid		n-H ₂ liquid		n-H ₂ [2-35]		
	T (K)	P (mm Hg)	T (K)	P (mm Hg)	T (K)	P (mm Hg)	
see table 2-3	3.4	1.0×10^{-9}	13.957	54.	27.256	3766.5	
	3.6	5.5×10^{-9}	14.	55.4	27.479	3929.7	
	3.8	2.9×10^{-8}	[6759]	95.	27.540	3958.4	
	4.0	9.5×10^{-8}		153.3	27.964	4287	
	4.2	3.5×10^{-7}	17	235.2 [6368]	27.970	4293	
	4.5	1.8×10^{-6}	18	345.9	28.289	4552	
	see table 2-2	4.71	4.52×10^{-6}	19	490.8	28.301	4549
		4.78	5.73×10^{-6}	20	675.7	28.464	4682
		4.85	7.28×10^{-6}	20.390	760	28.888	5032
		4.92	1.02×10^{-5}	21.023	909.1	29.178	5292
		5.01	1.37×10^{-5}	21.298	981.1	29.207	5312
		5.0	1.89×10^{-5}	21.607	1067.9	29.288	5343
		7.43	4.02×10^{-2}	21.835	1137.1	29.500	5579
7.74		6.72×10^{-2}	22.089	1202.5	29.771	5834	
8.11		1.294×10^{-1}	22.242	1261.4	29.979	6057	
8.76		3.18×10^{-1}	22.772	1445.2	29.996	6049	
9.29		7.32×10^{-1}	23.085	1557.0	30.137	6216	
9.91		1.55	23.537	1732.1	30.172	6234	
10.67		3.62	24.680	2244.0	30.601	6683	
11.11	5.70	25.929	2369.0	30.971	7064		
		25.209	2513.9	31.119	7220		
		26.328	3147.9	31.146	7255		
		26.721	3400.9	31.28	7363		
		26.791	3459.8	31.352	7481		
		27.072	3633.4	31.720	7912		
				32.276	8596		

Table 10-1. Recommended Vapor Pressures of the Hydrogens (Continued)

o-H ₂ [6368]		n-D ₂ [6368]		o-D ₂		Tritium [444]		HD	
T (K)	P (mm Hg)	T (K)	P (mm Hg)	T (K)	P (mm Hg)	T (K)	P (mm Hg)	T (K)	P (mm Hg)
solid		solid		solid		solid		solid	
None		10.000	0.05	10	0.05	13.968	1.5	10	0.28
	liquid	11.000	0.21	11	.21	15.274	5.4	11	.99
14.05	55.1	12.000	0.73	12	.75	16.122	10.2	12	2.94
15	92.2	13.000	2.14	13	2.20	17.314	24.2	13	7.46 [6368]
16	149.1	13.813	4.61	14	5.57 [6368]	18.054	40.2	14	16.8
17	229.2	13.957	5.24	15	12.6	18.214	43.5	15	34.4
18	337.8	14.000	5.44	16	26.0	18.534	53.9	16	65.2
19	480.7	14.050	5.68	17	49.6	18.952	68.5		liquid
20	662.6	15.000	12.32	18	88.7	19.023	71.1	17	112.4
		16.000	25.43		liquid	19.400	87.3	18	176.2
		16.604	37.92	19	147.1	19.556	94.8	19	264.2
		17.000	48.61	20	222.3	19.746	105.1	20	382.0
		18.000	87.15	21	324.1	20.020	120.9	21	534.9
		18.691	126.35	22	457.8	20.216	133.4	22	728.5
		18.723	128.47	23	628.8	20.547	158.7	23	969.0
			liquid	24	843.1		liquid	24	1262.3
			see table 2-7	25	1106.6	20.647	163.7	25	1614.5
				26	1425.6	20.985	188.6	26	2031.5 [454]
				27	1806.1	21.637	245.1	27	2519.4
				28	2253.5 [454]	22.032	283.8	28	3084.6
				29	2774.3	22.431	329.3	29	3732.8
				30	3374.7	22.776	370.5	30	4470.8
				31	4062.0	23.181	425.9	31	5306
				32	4842.4	23.557	483.2	32	6246
				33	5723	23.941	545.8	33	7301
				34	6712	24.337	618.2	34	8480
				35	7819	24.632	675.3	35	9796
				36	9051	25.182	792.0		
				37	10418	25.700	917.3		
				38	11943	26.367	1096.5		
						27.207	1356.0		
						27.929	1617.9		
						28.586	1884.2		
						29.191	2152.9		

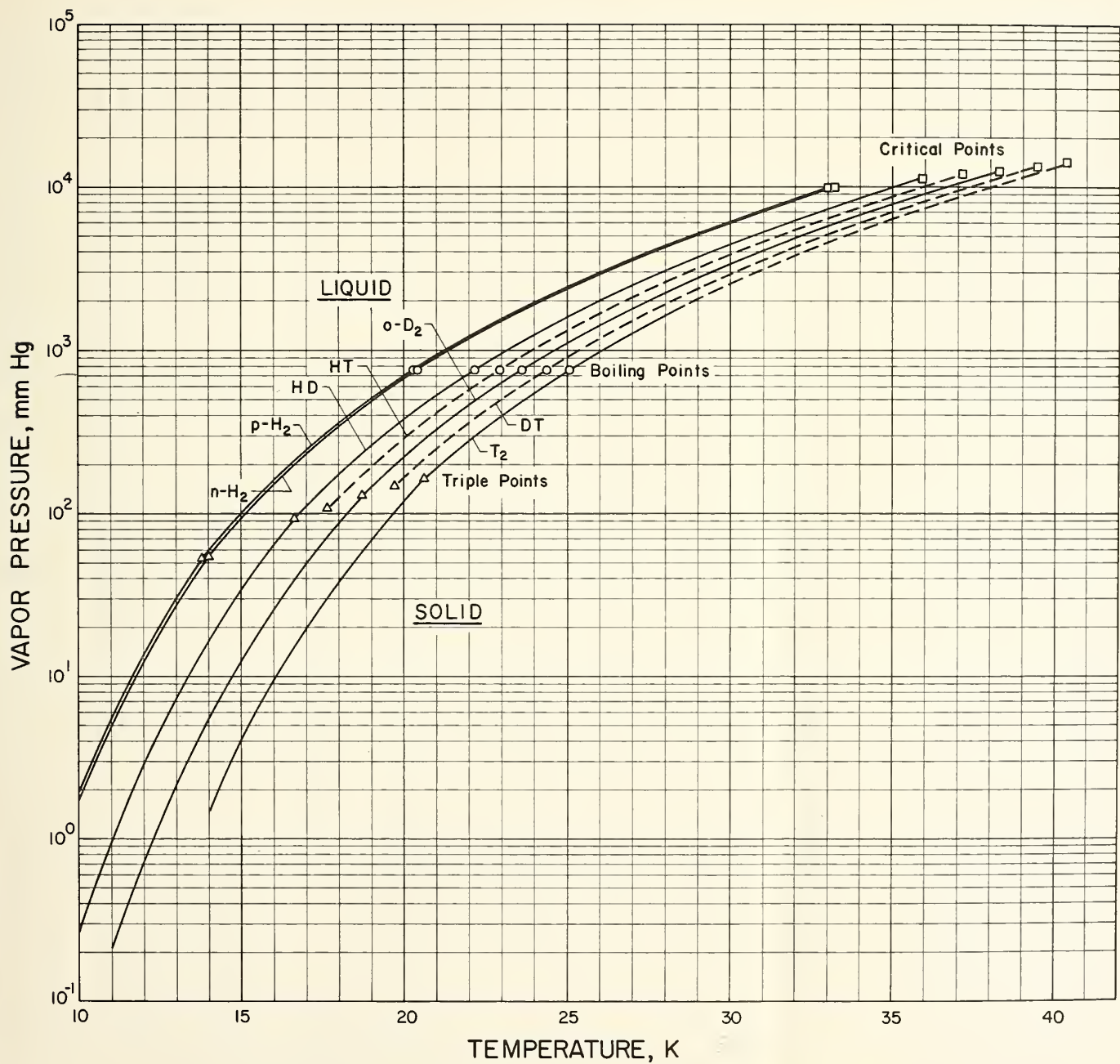


Figure 10-2. Solid and Liquid Vapor Pressures of the Various Hydrogens

Table 10-2. Calculated Vapor Pressures of the Hydrogens

T (K)	$e\text{-H}_2$	$n\text{-H}_2$	HD	HT	$e\text{-D}_2$	$n\text{-D}_2$	DT	$n\text{-T}_2$
Triple point (K)	13.81	13.96	16.60	17.62	18.69	18.73	91.71	20.62
P (mm)	52.8	54.0	92.8	109.5	128.5	128.6	145.7	162.0
14.0	57.2	55.5						
14.5	75.8	73.5						
15.0	98.7	95.7						
15.5	126.4	122.6						
16.0	159.5	154.7						
16.5	198.7	192.7						
17.0	244.7	237.3	112.0					
17.5	298.1	288.9	141.3					
18.0	359.5	348.5	176.1	125.2				
18.5	429.6	416.4	217.0	156.9				
19.0	509.1	493.4	264.7	194.3	146.8	145.5		
19.5	598.8	580.1	319.6	238.0	181.6	180.1		
20.0	699.2	677.4	382.7	288.8	222.5	220.6	164.9	
20.5	811.1	785.7	454.4	347.2	269.9	267.6	202.8	153.7
21.0	935.3	905.7	535.5	413.9	324.6	321.8	246.9	189.5
21.5	1072.4	1038.3	626.7	489.8	387.2	383.6	298.0	231.4
22.0	1223.3	1184.0	728.7	575.3	458.3	454.2	356.6	280.0
22.5	1388.5	1343.7	842.3	671.2	538.7	533.8	423.5	336.0
23.0	1568.9	1517.9	968.0	778.2	628.9	623.3	499.3	400.0
23.5	1765.2	1707.3	1106.8	897.2	729.9	723.2	584.8	472.7
24.0	1978.3	1912.8	1259.2	1028.7	842.3	834.5	680.6	554.8
24.5	2208.8	2135.1	1426.1	1173.6	966.7	957.7	787.5	647.1
25.0	2457.6	2374.9	1608.3	1332.6	1104.0	1093.5	906.2	750.3
25.5	2725.5	2632.9	1806.5	1506.4	1255.0	1242.9	1037.4	865.2
26.0	3013.4	2910.0	2012.4	1695.8	1420.3	1406.4	1181.9	992.3
26.5	3322.1	3207.0	2254.0	1901.6	1600.7	1584.9	1340.7	1132.7
27.0	3652.7	3524.6	2504.9	2124.5	1797.1	1779.1	1514.2	1286.9
27.5	4005.9	3863.8	2775.1	2365.4	2010.3	1989.8	1703.5	1455.9
28.0	4382.6	4225.3	3065.5	2625.1	2241.1	2217.9	1909.2	1640.3
28.5	4783.9	4610.2	3376.7	2901.1	2490.2	2464.0	2132.2	1841.1
29.0	5211.2	5019.2	3710.0	3203.9	2758.6	2729.1	2373.3	2058.9
29.5	5665.2	5453.5	4065.9	3524.6	3047.2	3013.9	2633.5	2294.6
30.0	6147.2	5914.2	4445.9	3867.7	3356.9	3319.5	2913.4	2549.3
30.5	6658.5	6402.3	4850.6	4233.7	3688.5	3646.7	3214.2	2823.4
31.0	7200.7	6919.2	5281.0	4623.6	4043.1	3996.3	3536.5	3118.0
31.5	7774.9	7465.8	5738.5	5038.7	4421.5	4369.3	3881.5	3434.1
32.0	8382.9	8043.7	6224.1	5479.5	4825.2	4766.7	4250.1	3772.3
32.5	9026.4	8654.4	6739.3	5947.3	5254.8	5189.8	4643.2	4113.9
33.0		9299.0	7285.1	6443.2	5711.6	5639.2	5061.9	4519.5
33.5			7863.1	6968.1	6169.6	6116.4	5507.2	4930.3
34.0			8475.0	7523.4	6711.4	6622.1	5980.5	5367.1
34.5			9122.1	8110.7	7257.3	7158.1	6482.7	5831.1
35.0			9806.5	8730.8	7835.7	7725.7	7015.1	6323.5
35.5			10529.0	9385.5	8448.0	8325.6	7579.1	6845.3
36.0				10076.0	9095.7	8960.2	8176.3	7397.7
36.5				10805.0	9781.2	9630.7	8808.0	7981.7
37.0				11573.0	10505.0	10339.0	9475.8	8599.2
37.5					11272.0	11087.0	10181.0	9251.1
38.0					12083.0	11877.0	10927.0	9938.8
38.5							11718.0	10664.0
39.0							12547.0	11429.0
39.5								12236.0
40.0								13086.0
Critical point (K)	32.99	33.24	35.91	37.13	38.26	38.35	39.42	40.44
P (mm)	9705.2	9735.6	11134.0	11780.0	12373.0	12487.0	13300.0	13878.0

with permission from H. M. Mittlehauser, et al., *Cryogenics* 4, 368 (1964).

11. Fixed Points

In table 11-1 we present temperatures and pressures for the triple point, the normal boiling point, and the critical point of the various hydrogens. For normal hydrogen, the values of Woolley, et al. [6368] are adopted. In addition we present a most extensive table of values for the many different properties at the fixed points of normal hydrogen, table 11-2 [Cryogenics Division, NBS, unpublished]. Values are taken from the many sources given in the auxiliary Notes and Reference sheet.

For parahydrogen we select the values given by Roder et al., [29210] in order to be consistent with the values of entropy, enthalpy, etc., which have been presented. A second extensive table of property values at the fixed points of parahydrogen is given in table 11-3 which is quite similar to the one for normal hydrogen.

For the other hydrogens we first list in table 11-1 the values adopted by Woolley et al., [6368] who summarized the literature up to 1948. In addition, all papers are listed which present results of direct experimental measurements. The listing is rounded out by a limited number of values which have been calculated from theory or estimated in various ways. By presenting values from different sources in table 11-1, we give the reader an opportunity to see how much variation is possible in any given value. The preferred values of this survey are underlined.

The temperatures and pressures given in table 11-1 are those of the original articles. The user should be aware that much of the variation in values is due to the use of different temperature scales. Scales such as the NBS-39, the NBS-55, and the IPTS-68 have been in common use. These scales differ by about 0.015K for temperatures up to 40K. Scales in use at some laboratories which are based on thermocouple thermometry occasionally display larger differences. Reconciliation of differences in property values by adjustment of temperatures is beyond the scope of this survey and has not been accomplished here.

Table 11-1. Fixed Point Values for the Hydrogens

Hydrogen Modification	Author	Triple Point		Normal Boiling Point (760 mm Hg)		Critical Point	
		T, K	P, mm Hg	T, K	P, mm Hg	T, K	P, mm Hg
o-H ₂	Woolley, et al. 1948 [6368]	<u>(14.05)</u>	<u>(55.1)</u>	<u>(20.454)</u>			
n-H ₂	Woolley, et al. 1948 [6368]	<u>13.957</u>	<u>54.04</u>	<u>20.39₀</u>		<u>33.19</u>	<u>986.5</u>
p-H ₂ or e-H ₂	Roder, et al. 1965 [29210]	<u>13.803</u>	<u>52.82</u>	<u>20.268</u>		<u>32.976</u>	<u>9696.8</u>
o-D ₂ or e-D ₂	Woolley et al. 1948 [6368]	<u>18.691</u>	<u>128.5</u>	<u>23.52</u>			
	Kerr et al. 1951 [645]	<u>18.63</u>	<u>127.5</u>	<u>23.59₀</u>			
	Hoge and Arnold 1951 [454]	<u>18.691</u>	<u>128.5</u>	<u>23.63₇</u>			
	Hoge and Lassiter 1951 [6084]						
n-D ₂	Woolley et al. 1948 [6368]	<u>18.72₃</u>	<u>128.5</u>	<u>23.57</u>		<u>38.262</u>	<u>12374.</u>
	Grilly 1951 [444]	<u>18.73</u>	<u>128.6</u>				
	Friedman et al. 1951 [16332]	<u>18.72_*</u>	<u>128.5_*</u>	<u>23.57_*</u>		<u>38.35</u>	<u>12488.</u>
	Grenier and White 1964 [22677]	<u>18.698</u>					
	Prydz† 1967 [43781]	<u>18.71</u>	<u>128.5</u>	<u>23.66</u>		<u>38.34</u>	<u>12488.</u>
p-D ₂	Woolley et al. 1948 [6368]	<u>(18.78)</u>	<u>(128.5)</u>	<u>(23.66)</u>			
75.1% p	Grenier and White 1964 [22677]	<u>18.781</u>					
85 % p	Grenier and White 1964 [22677]	<u>18.787</u>					
T ₂	Grilly 1951 [444]	<u>20.62</u>	<u>162.0</u>	<u>25.04</u>			
	Mittelhauser and Thodos 1964 [32140]	<u>(21.65)</u>	<u>(188.0)</u>	<u>(25.57)</u>		<u>(40.44)</u>	<u>(13878)</u>
	Rogers and Brickwedde 1965 [28408]	<u>(20.62)**</u>	<u>(162.)**</u>	<u>(25.04)**</u>		<u>(40.)</u>	
HD	Woolley et al. 1948 [6368]	<u>16.60₄</u>	<u>92.8</u>	<u>22.133</u>		<u>35.908</u>	<u>11130.</u>
	Hoge and Arnold 1951 [454]	<u>16.60₄</u>	<u>92.8</u>	<u>22.143</u>			
	Hoge and Lassiter 1951 [6084]						
HT	Friedman et al. 1951 [5677]	<u>(18.5)</u>	<u>(124.6)</u>	<u>(23.6)</u>		<u>(38.3)</u>	<u>(12464.)</u>
	Mittelhauser and Thodos 1964 [32140]	<u>(17.62)</u>	<u>(109.5)</u>	<u>(22.92)</u>		<u>(37.13)</u>	<u>(11780.)</u>
DT	Friedman et al. 1951 [5677]	<u>(19.7)</u>	<u>(142.9)</u>	<u>(24.3)</u>		<u>(39.5)</u>	<u>(13148.)</u>
	Mittelhauser and Thodos 1964 [32140]	<u>(19.71)</u>	<u>(145.7)</u>	<u>(24.38)</u>		<u>(39.42)</u>	<u>(13300.)</u>

* taken from Woolley et al. [6368] ** taken from Grilly [444]

† taken from Friedman et al. [16332] with a change in temperature scale

() calculated or estimated values

— preferred values

Table 11-2. Fixed Point Properties of Normal Hydrogen

PROPERTIES ↓	CONDITIONS →				Normal Boiling Point	Critical Point ††	Standard Conditions		References and Notes ‡
	Solid	Liquid	Vapor	Vapor			STP(0°C)	NTP(20°C)	
Temperature (K)	13.957 [†]				20.390 [†]	33.19 [†]	273.15	293.15	5
Pressure (mmHg)	54.04				760	9865	760	760	5
Density (mole/cm ³) × 10 ³	43.01 ^[2]	38.30	0.0644	35.20	0.6604	14.94	0.04460	0.04155	5, 17
Specific Volume (cm ³ /mole) × 10 ⁻³	0.02325 ^[2]	0.026108	15.519	0.028409	1.5143	0.066949	22.423	24.066	5, 17
Compressibility Factor, $Z = \frac{PV}{RT}$	—	0.001621	0.9635	0.01698	0.9051	0.3191	1.00042	1.00049	a
Heats of Fusion & Vaporization (J/mole)	117.1	911.3			899.1	0	—	—	4, h
Specific Heat (J/mole-K)	5.73	13.85	-46.94	18.91	-33.28	(very large)	—	—	5, 6
C_p @ constant pressure	—	13.23	21.22	19.70	24.60	(very large)	28.59	28.89	5, 6, 17
C_v @ constant volume	—	9.53	12.52	11.60	13.2	(19.7)	20.30	20.40	5, 6, 17
Specific Heat Ratio, $\gamma = C_p/C_v$	—	1.388	1.695	1.698	1.863	(large)	1.408	1.416	c
Enthalpy (J/mole)	321.6	438.7	1350.0	548.3	1447.4	1164	7749.2	8324.1	5, 17, d
Internal Energy (J/mole)	317.9 ^e	435.0	1234.8	545.7	1294.0		5477.1	5885.4	5, 17, d
Entropy (J/mole-K)	20.3	28.7	93.6	34.92	78.94	54.57	139.59	141.62	5, 17, d
Velocity of Sound (m/sec)	—	1282	307	1101	357		1246	1294	6, 18
Viscosity, μ (N-sec/m ²) × 10 ³ (centipoise) ††	—	0.0256	0.00074	0.0133	0.00111	(0.0035)	0.00834	0.00875	18
Thermal Conductivity (mW/cm-K), k	—	0.0256	0.00074	0.0133	0.00111	(0.0035)	0.00834	0.00875	18
Prandtl Number, $N_{pr} = \mu C_p/k$	9.0 ^[11]	0.74	0.105	1.00	0.165	(*)	1.739	1.842	6, 9
Dielectric Constant, ϵ	1.287	2.27	0.742	1.30	0.821		0.680	0.681	c
Index of Refraction, $n = \sqrt{\epsilon}$ †	1.134	1.253	1.00039	1.231	1.0040	1.0937	1.000271	1.000253	13
Surface Tension (N/m) × 10 ³	—	1.119	1.000196	1.1093	1.0020	1.0458	1.000136	1.000126	e
Equiv. Vol./Vol. Liquid at NBT	0.8184	3.00	—	1.94	—	0	—	—	15
† Long Wavelengths		0.9190	546.3	1	53.30	2.357	789.3	847.1	f

Gas Constant: $R = 62, 362.7 \text{ cm}^3\text{-mmHg/mole-K}^\dagger$

† Values in parenthesis are estimates

* Anomalous Large

‡ Apply to all items on the line except for specific values separately referenced by superscript

Molecular Weight = 2.015946g

"mole" = gram mole

** Units for poise are: g/cm-sec

Table 11-3. Fixed Point Properties of Parahydrogen

PROPERTIES ↑	CONDITIONS →				Normal Boiling Point	Critical Point ††	Standard Conditions STP(0°C)	References and Notes ‡
	Solid	Liquid	Vapor	Vapor				
Temperature (K)	13.803	20.268			32.976	273.15	293.15	1
Pressure (mmHg)	52.82	760			9696.8	760	760	1
Density (mole/cm ³) x 10 ³	42.9 [2]	38.207	0.0623	0.6636	15.59	0.05459	0.04155	1, 3
Specific Volume (cm ³ /mole) x 10 ⁻³	0.02330 [2]	0.026173	16.057	1.5069	0.064144	22.425	24.069	1, 3
Compressibility Factor, $Z = \frac{PV}{RT}$	—	0.001606	0.9850	0.9061	0.3025	1.0005	1.0006	a
Heats of Fusion & Vaporization (J/mole)	117.5 [4]	905.5			0	—	—	b
Specific Heat (J/mole-K)	5.73 [5]	13.85	-46.94	18.91	(very large)	—	—	5, 6
C_p , @ constant pressure	—	13.13	21.20	19.53	(very large)	30.35	30.02	1, 3
C_v , @ constant volume	—	9.50	12.52	11.57	19.7	21.87	21.70	1, 3
Specific Heat Ratio, $\gamma = C_p/C_v$	—	1.382	1.693	1.688	(large)	1.388	1.383	c
Enthalpy (J/mole)	-740.2 [4]	-622.7	282.8	-516.6	77.6	7656.6	8260.6	1, 3, d
Internal Energy (J/mole)	-740.4°	-622.9	169.8	-519.5	5.7	5384.5	5822.0	1, 3, d
Entropy (J/mole-K)	1.49 [4]	10.00	75.63	16.08	35.4	127.77	129.90	1, 3, d
Velocity of Sound (m/sec)	—	1273	305	1093	350	1246	1294	1, 7
Viscosity, μ (N-sec/m ²) x 10 ³ (centipoise) ††	—	0.026	0.00073	0.0133	0.0011	0.0035	0.00875	8, 9, 10
Thermal Conductivity (mW/cm-K), k	9.0 [1]	0.026	0.00073	0.0133	0.0011	0.0035	0.00875	8, 9, 10
Prandtl Number, $N_{pr} = \mu C_p/k$	—	0.73	0.104	1.00	0.163	(*)	1.897	8, 9, 12
Dielectric Constant, ϵ	1.286	2.33	0.738	1.288	0.8202	0.6873	0.6867	c
Index of Refraction, $n = \sqrt{\epsilon}$ †	1.134	1.252	1.00038	1.230	1.0040	1.00027	1.00026	13
Surface Tension (N/m) x 10 ³	—	1.119	1.00019	1.109	1.0020	1.00013	1.00012	14, e
Equiv. Vol./Vol. Liquid at NBT	—	2.99	—	1.93	0	—	—	15
Equiv. Vol./Vol. Liquid at NBT	0.8181	0.9190	563.8	1	52.91	787.4	845.1	f

Gas Constant: $R = 62,362.7 \text{ cm}^3\text{-mmHg/mole-K}^{(1)}$

† Values in parenthesis are estimates

‡ Apply to all items on the line except for specific values separately referenced by superscript

Molecular Weight = 2.01594 [2e]

* "mole" = gram mole

†† Units for poise are: g/cm-sec

Notes and References for Tables 11-2 and 11-3

<p>a. Compressibility factor calculated from the tabulated data using the gas constant, R, from reference 1.</p> <p>b. Heats of vaporization calculated from enthalpy differences given in references 1 and 3.</p> <p>c. Calculated from property values given in this table.</p> <p>d. Base point (zero values) for enthalpy, internal energy, and entropy are 0 K for the ideal gas at 1 atmosphere pressure.</p> <p>e. Index of refraction calculated from the dielectric constant data given in reference 13, in good agreement with the values reported in reference 14.</p>	<p>f. Ratio of specific volumes reported in reference 1 and 3, or 5 and 17.</p> <p>g. These temperatures given in reference 5 are based on the IPTS-1968 temperature scale. To compare with the corresponding temperatures given in reference 1 for para-hydrogen which are based on the NBS-1955 temperature scale, 0.01 K should be subtracted, i. e., use 13.947, 20.380 and 33.18 respectively for the triple point, normal boiling point and critical point when determining property differences between normal and para hydrogen.</p> <p>h. Heats of fusion and vaporization calculated from enthalpy differences and as reported in references 4, 5, and 17.</p>
<p>Tables 11-2, 11-3</p>	
1.	not used
2.	[5729]
3.	not used
4.	[54546]
5.	not used
6.	not used
7.	[6560]
8.	not used
9.	not used
10.	not used
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Table 11-4. Fixed Point Properties of Hydrogen Deuteride

PROPERTIES ↓	CONDITIONS →				References
	Solid	Liquid	Vapor	Normal Boiling Point Liquid Vapor	
Temperature (K)	16.60 ₄		22.14 ₃		[6368] [454]
Pressure (mmHg)	92.8		760		[6084]
Density (mole/cm ³)	0.04579	0.04062	0.03782		[6368] [59955]
Specific Volume (cm ³ /mole)	21.84	24.62	26.44		[6084]
Compressibility Factor, $Z = \frac{PV}{RT}$	—				
Heats of Fusion & Vaporization (J/mole)	159.4	(1179.5)			[6368]
Specific Heat (J/mole-K)	C_s , @ saturation				[27091]
	C_p , @ constant pressure				(26.4)
	C_v , @ constant volume				(very large)
Specific Heat Ratio, $\gamma = C_p/C_v$	—				(large)
Enthalpy (J/mole)	—				
Internal Energy (J/mole)	—				
Entropy (J/mole-K)	—				
Velocity of Sound (m/sec)	$\Delta = 9.6$	$(\Delta = 71.04)$		$\Delta = 48.56$	calculated
Viscosity, μ (N-sec/m ²) (micropoise) **	—				
	—	338	198.5		13.3
Thermal Conductivity (mW/cm-K), k	—				[42002] [47292] [8330]
Prandtl Number, $N_{pr} = \mu C_p/k$	—				(*)
Dielectric Constant,	—				
Index of Refraction,	—				
Surface Tension $d\gamma/cm$	—				sec. 6.2
Equiv. Vol./Vol. Liquid at NBT	—	3.389 @ 16.71	—		[22219]

Values in parenthesis are estimates

* Anomalous Large

Molecular Weight = 3.023

** Units for poise are: g/cm-sec

Table 11-5. Fixed Point Properties of Hydrogen Tritide
(Properties are often assumed to be the same as D₂)

PROPERTIES ↓	CONDITIONS →						References
	Solid	Liquid	Vapor	Normal Boiling Point Liquid	Normal Boiling Point Vapor	Critical Point	
Temperature (K)	(17.62)						(37.13)
Pressure (mmHg)	(109.5)						(11780)
Density (mole/cm ³)							760
Specific Volume (cm ³ /mole)							
Compressibility Factor, $Z = \frac{PV}{RT}$	—						
Heats of Fusion & Vaporization (J/mole)							
Specific Heat (J/mole-K)	C_s , @ saturation						
	C_p , @ constant pressure						(very large)
	C_v , @ constant volume						
Specific Heat Ratio, $\gamma = C_p/C_v$	—						(large)
Enthalpy (J/mole)							
Internal Energy (J/mole)							
Entropy (J/mol ^o -K)							
Velocity of Sound (m/sec)							
Viscosity, μ	(N-sec/m ²)						
	(micropoise) **						(275.6)
Thermal Conductivity (mW/cm-K), k							[38388]
Prandtl Number, $N_{pr} = \mu C_p/k$							(*)
Dielectric Constant, n							
Index of Refraction, ϵ							
Surface Tension (N/m)	—						
Equip. Vol./Vol. Liquid at NBT							1

Values in parenthesis are estimates

* Anomalously Large

Molecular Weight = 4.025

** Units for poise are: g/cm-sec

Table 11-6. Fixed Point Properties of Ortho or e-Deuterium

PROPERTIES ↓	CONDITIONS ↔					References
	Solid	Liquid	Vapor	Liquid	Vapor	
Temperature (K)	18.691					38.262 [454]
Pressure (mmHg)	128.5					12374 [6084]
Density (mole/cm ³)	0.04859	0.042988		0.040072		0.01658 [13537]
Specific Volume (cm ³ /mole)	20.58	23.262		24.955		60.3 [6084]
Compressibility Factor, $Z = \frac{PV}{RT}$	—					
Heats of Fusion & Vaporization (J/mole)	196.9			1229.8		0 [645]
Specific Heat (J/mole-K)	—					(very large)
Specific Heat Ratio, $\gamma = C_p/C_v$	—					(large)
Enthalpy (J/mole)						
Internal Energy (J/mole)						
Entropy (J/mole-K)	($\Delta = 10.53$)			($\Delta = 52.028$)		calculated
Velocity of Sound (m/sec)						
Viscosity, μ (N-sec/m ²)	—					
(centipoise) **	—					
Thermal Conductivity (mW/cm-K), k						(*)
Prandtl Number, $N_{Pr} = \mu C_p/k$						
Dielectric Constant, n						
Index of Refraction, ϵ						
Surface Tension dyn/cm	—					
Equiv. Vol./Vol. Liquid' at NBT				1		

Values in parenthesis are estimates

* Anomalously Large

Molecular Weight = 4.0282

** Units for poise are: g/cm-sec

Table 11-7. Fixed Point Properties of Normal Deuterium

PROPERTIES ↓	CONDITIONS →						References
	Solid	Triple Point		Normal Boiling Point		Critical Point	
Temperature (K)		18.71		23.66		38.34	[43781]
Pressure (mmHg)		128.5		760		12488	
Density (mole/cm ³)	0.04883	0.043174		0.0404446		0.00056607	0.017328
Specific Volume (cm ³ /mole)	20.48	23.162		24.7252		1766.58	57.710
Compressibility Factor, $Z = \frac{PV}{RT}$	—	—		—		—	—
Heats of Fusion & Vaporization (J/mole)	196.6	1287.6		1210.7		0	[5995] [43781]
Specific Heat (J/mole-K)	C_s , @ saturation	(23.0)		(26.66)		(very large)	Fig. 4-6
	C_p , @ constant pressure	—		—		—	—
	C_v , @ constant volume	—		(14.7)		—	Fig. 4-5
Specific Heat Ratio, $\gamma = C_p/C_v$	—	—		—		(large)	—
Enthalpy (J/mole)	-845.5	-648.9		638.7		-509.4	701.3
Internal Energy (J/mole)	—	—		—		-512.0	522.3
Entropy (J/mole-K)	7.76	18.27		87.09		24.78	75.97
Velocity of Sound (m/sec)	—	1070@19		255.3@18.9		(965.1)	47.38
Viscosity, μ (N-sec/m ²)	(micropoise) **	—		—		—	—
		434@18.8		283		—	—
Thermal Conductivity (mW/cm-K), k	2.73	—		(1.08)		(0.26)	(*)
Prandtl Number, $N_{pr} = \mu C_p/k$	—	—		—		—	—
Dielectric Constant, n	—	—		—		—	—
Index of Refraction, ϵ	—	—		—		—	—
Surface Tension dyn/cm	—	—		—		—	—
Equiv. Vol./Vol. Liquid at NBT	—	3.80@18.77		—		—	—

Values in parenthesis are estimates

* Anomalous Large

Molecular Weight = 4.0282

** Units for poise are: g/cm-sec

Table 11-8. Fixed Point Properties of Deuterium Tritide

PROPERTIES ↓	CONDITIONS →		Triple Point		Normal Boiling Point		Critical Point	References
	Solid	Liquid	Liquid	Vapor	Liquid	Vapor		
Temperature (K)		(19.71)			(24.38)		(39.42)	[32140]
Pressure (mmHg)		(145.7)			760		(13300)	
Density (mole/cm ³)					(0.0418)			
Specific Volume (cm ³ /mole)					(23.9)		(57.8)	[32140]
Compressibility Factor, $Z = \frac{PV}{RT}$	—							
Heats of Fusion & Vaporization (J/mole)							0	
Specific Heat (J/mole-K)	C_s , @ saturation							
	C_p , @ constant pressure	—					(very large)	
	C_v , @ constant volume	—						
Specific Heat Ratio, $\gamma = C_p/C_v$	—						(large)	
Enthalpy (J/mole)								
Internal Energy (J/mole)								
Entropy (J/mole-K)								
Velocity of Sound (m/sec)	(N-sec/m ²)							
	(micropoise) **	—		(803)		(423)		sec. 5.5
Thermal Conductivity (mW/cm-K), k							(*)	
Prandtl Number, $N_{pr} = \frac{\mu C_p}{k}$								
Dielectric Constant, n						(1.2703)		sec. 6.2
Index of Refraction, ϵ								
Surface Tension (N/m)	—							
Equiv. Vol./Vol. Liquid at NBT						1		

Values in parenthesis are estimates

* Anomalous Large

Molecular Weight = 5.032

** Units for poise are: g/cm-sec

Table 11-9. Fixed Point Properties of Tritium

PROPERTIES ↓	CONDITIONS →				References
	Solid	Liquid	Vapor	Normal Boiling Point Liquid Vapor	
Temperature (K)		20.62		25.04	[444]
Pressure (mmHg)		162.0		760	[32140]
Density (mole/cm ³)	(0.0537)@4	0.04539	0.00013	0.04262	(0.01806)
Specific Volume (cm ³ /mole)	(18.62)@4	22.03	(7692)	23.46	(55.3)
Compressibility Factor, $Z = \frac{PV}{RT}$	—				[28408]
Heats of Fusion & Vaporization (J/mole)				1393.3	[444]
Specific Heat (J/mole-K)					
C_s , @ saturation					
C_p , @ constant pressure	—				(very large)
C_v , @ constant volume	—			(15.4@29.6)	[21016]
Specific Heat Ratio, $\gamma = C_p/C_v$	—				(large)
Enthalpy (J/mole)					
Internal Energy (J/mole)					
Entropy (J/mole-K)				($\Delta = 55.64$)	calculated
Velocity of Sound (m/sec)					
Viscosity, μ (N-sec/m ²)	—				
(micropoise) ##	—	(819)		(432@25)	[38388]
Thermal Conductivity (mW/cm-K), k				(1.11)	(*) Fig. 5-2
Prandtl Number, $N_{pr} = \mu C_p/k$					
Dielectric Constant, n				(1.2735)	sec. 6.2
Index of Refraction, ϵ					
Surface Tension (N/m)	—				
Equiv. Vol./Vol. Liquid at NBT				1	

Values in parenthesis are estimates

* Anomalous Large

Molecular Weight = 6.034

Units for poise are: g/cm-sec

12. Mixture Properties

The essential data elements for mixture problems are: 1. properties of the pure components, 2. equilibrium properties of mixtures, and 3. transport properties of mixtures. The properties of the pure components have been described in the present report. The equilibrium properties of mixtures include excess volumes, excess Gibbs free energies, and excess enthalpies. In a bibliography on equilibrium properties which is nearly completed, Hiza, Kidnay and Miller [Hiza, et al.] provide the designer with a complete reference to available experimental data on mixtures of cryogenic interest. The data are categorized according to the various phase equilibria, gas or liquid mixture densities, and mixture enthalpies; solubilities are thus covered in this volume. For the transport properties it would be desirable to conduct a similar survey, however, such a survey is not planned at present.

A few references that may be of particular value are the following: Hoge and Arnold [454] who report dew and bubble points of H_2 -HD and D_2 mixtures; Technical Note 621 by Hiza [77791] on the He^4 - nD_2 , the He^3 - nD_2 , the He^4 - nH_2 , and the He^3 - nH_2 systems. Sherman [Sherman, 1972] is presently investigating the He^4 and He^3 systems with nH_2 , nD_2 , and nT_2 . A number of hydrogen mixtures with other gases have been surveyed by Esel'son, et al. [Esel'son, et al., 1971]. Several references for diffusion coefficients in binary liquid systems will be available when the compilation of Bailey, et al. [Bailey, et al.] is published.

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U.S. DEPT. OF COMM. BIBLIOGRAPHIC DATA SHEET	1. PUBLICATION OR REPORT NO. NBS TN-641	2. Gov't Accession No.	3. Recipient's Accession No.
4. TITLE AND SUBTITLE Survey of the Properties of the Hydrogen Isotopes Below Their Critical Temperatures		5. Publication Date October 1973	6. Performing Organization Code
7. AUTHOR(S) H. M. Roder, G. E. Childs, R. D. McCarty, and P. E. Angerhofer		8. Performing Organization	
9. PERFORMING ORGANIZATION NAME AND ADDRESS NATIONAL BUREAU OF STANDARDS, Boulder Labs. DEPARTMENT OF COMMERCE Boulder, Colorado 80302		10. Project/Task/Work Unit No. 2752450	11. Contract/Grant No. AEC AT(04-3)-905
12. Sponsoring Organization Name and Address University of California Lawrence Livermore Laboratory Livermore, California 94550		13. Type of Report & Period Covered	
15. SUPPLEMENTARY NOTES		14. Sponsoring Agency Code	
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17. KEY WORDS (Alphabetical order, separated by semicolons) vapor pressure. Compilation; density; deuterium; electrical properties; enthalpy; entropy; fixed points; hydrogen; mechanical properties; optical properties; specific heat; thermophysical properties; transport properties; tritium;			
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