

Thermal Expansion of Liquid Normal Hydrogen Between 18.8 and 22.2 K

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The thermal expansion coefficient α of liquid normal hydrogen ($n\text{-H}_2$) was measured between 18.8 and 22.2 K in the pressure range 5 to 70 bar. The results are compared with those derived from PVT measurements by others on both normal and *para* ($p\text{-H}_2$) hydrogen. Our analysis of the earlier normal data includes fitting an empirical equation of state, and expansion coefficients are derived from this equation by differentiation. We discuss the effects on α and the compressibility β from molecular quadrupole interactions; both theoretical and empirical results suggest these to be on the order of 2% or less for the normal spin mixture. We conclude that our thermal expansion data are consistent with earlier results on both $n\text{-H}_2$ and $p\text{-H}_2$ in this range of pressures and temperatures.

Key words: hydrogen, normal; hydrogen, *para*; pressure, 5 to 70 bar; temperature, 18.8 to 22.2 K; thermal expansion coefficient.

1. Introduction

The prototype, inertial-confinement reactor, designed and built at this laboratory, is fueled with mixtures of D_2 , DT , and T_2 . The fuel is contained in tiny (100 μm diameter), hollow glass microballoons. Its state is either solid or liquid depending on the temperature and pressure, which are fixed somewhere in the range between 18.5 and 23.0 K, and zero and 70 bar.

When the program for target fabrication began, little was known about the behavior of the fuels. Therefore, along with other work, efforts were included to measure and correlate the physical properties of all the hydrogen isotopes. Our part in this program has been to supply thermodynamic data on the solid and liquid phases. One product of this research is a recently published study on deuterium [1]¹. Similar data on tritium will follow. Here,

About the Authors, Paper: The work reported on was performed at the Los Alamos National Laboratory where L. A. Schwalbe, a physicist, remains and from which E. R. Grilly, also a physicist, is retired. The work was supported by the Fusion Target Fabrication Group at Los Alamos.

¹Figures in brackets indicate literature references at the end of this paper.

we consider the equation of state for liquid H_2 with particular emphasis on the thermal expansivity α of the normal spin mixture.

In this range of temperature and pressure the properties of liquid *para* hydrogen ($p\text{-H}_2$) are well described [2,3]. PVT data exist [4] for normal hydrogen ($n\text{-H}_2$), which contains 0.25 *para* ($J=0$) and 0.75 *ortho* ($J=1$) species. While the latter results are less extensive, the combined data are sufficient to describe certain differences between $n\text{-H}_2$ and $p\text{-H}_2$ such as the 0.5% difference in their molar volumes [5]. Usually, the liquid thermal expansion and compressibility β are assumed to be independent of the *ortho-para* composition. Theoretical estimates, discussed below, suggest that the differences, $\Delta\alpha = \alpha(n\text{-H}_2) - \alpha(p\text{-H}_2)$ and $\Delta\beta = \beta(n\text{-H}_2) - \beta(p\text{-H}_2)$, are about 2%.

Our purpose is to examine the effects of the *ortho-para* composition explicitly. We show that compressibilities derived from earlier PVT measurements of $n\text{-H}_2$ and $p\text{-H}_2$ are consistent with theoretical estimates of $\Delta\beta$. However, the precision of these data is not sufficient to permit a similar analysis for $\Delta\alpha$. It was, therefore, necessary to measure the expansivity of $n\text{-H}_2$ directly. These data, presented below, range in temperature from 18.8 to 22.2 K and in pressure from about 5 to 70 bars. We compare them with earlier data on both $p\text{-H}_2$ [3] and

n -H₂ [4], and find the results to be generally consistent with theory.

2. Experimental

The experiments on liquid hydrogen were carried out with the same apparatus as that described and used in the previous work on deuterium [1]. Briefly, the pressure cell consisted of three BeCu diaphragms welded together at their circumference and separated by 0.3 mm gaps. The upper gap, which served as the sample chamber, could be sealed with a low-temperature valve; the lower gap remained open to a room-temperature gas-handling system.

The entire pressure-cell assembly was held immersed in a bath of liquid hydrogen. Temperatures were obtained from measurements of the vapor pressure of the cryogen and the liquid-vapor equation for p -H₂ by Souers et al. [6]. (By strict definition our cryogen was not pure *para*; the equilibrium mixture of hydrogen at this temperature, commonly denoted as e -H₂, contains an *ortho* fraction of about 0.2%. However, for the following discussion the difference between e -H₂ and p -H₂ is insignificant, and both forms will be referred to simply as p -H₂.)

The volume of the sample is determined by the deflections of the upper and middle diaphragms of the cell. Increased internal pressures P_u in the upper gap increase the sample volume V_u . Increased external pressures, either in the lower gap P_l or in the surrounding cryogen bath P_b , decrease V_u . These impressed changes are expressed quantitatively by

$$\Delta V_u = (S_u + S_l)\Delta P_u - S_l\Delta P_l - S_u\Delta P_b \quad (1)$$

where S_u and S_l are pressure sensitivity factors of the volume to changes in the upper and middle diaphragm displacements, respectively. The sensitivity factors were determined by calibrating the sample volume at $T = 20.00$ K against the density values for p -H₂ tabulated by Goodwin et al. [2].

There is no explicit temperature dependence included in eq (1). In this range, there should be negligible effects from the thermal expansion of the cell material. To our knowledge, there are no measurements of thermal expansion for beryllium-copper, but data for copper exist, and the temperature coefficient for the alloy should be comparable. Rubin et al. [7] reported a linear expansion coefficient of 6×10^{-6} for copper at 25 K. Between 19 and 22 K, the volume coefficient should, therefore, be somewhere between 1 and 2×10^{-5} K⁻¹. The results obtained in this range for α of liquid hydrogen are between 9 and 19×10^{-3} K⁻¹. If accuracy limits for the

hydrogen data are set at about 1%, the effects of thermal expansion from the cell are at least an order of magnitude lower than the uncertainties arising from other sources.

To begin a set of measurements at a given pressure, the sample chamber was first evacuated, then filled with hydrogen, and sealed with the valve. Typically, during each run the temperature was first decreased and then increased in increments of 0.4 K. Each experimental value for α was obtained from the measured change in the lower-cell pressure ΔP_l that was required to maintain constant sample pressure P_u when the temperature of the system was changed by an amount ΔT . From eq (1) it follows that

$$\alpha \equiv \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p = - \frac{S_l \Delta P_l}{V_u \Delta T} - \frac{S_u}{V_u} \left(\frac{dP_b}{dT} \right) \quad (2)$$

where \bar{V}_u is the average sample volume for the measurement, and dP_b/dT is the temperature derivative of the cryogen vapor pressure calculated from the Souers et al. [6] equation for liquid p -H₂.

The total time of each sample confinement at low temperatures was about 12 or 13 h although one sample was kept cold for as long as 100 h. During the course of each set of measurements there was some *ortho-to-para* conversion. Our data were, therefore, not taken on "normal hydrogen" in the strictest sense. But the starting material for each sample was n -H₂, and we estimate the *ortho* fractions of most samples to have been in the range 0.65 to 0.75 at the time of their measurement.

The time for temperature equilibration at each point varied between 20 and 90 min depending on the amount of cryogen in the bath. During the course of each measurement, the effects of *ortho-to-para* conversion were, therefore, about 1 or 2%. Because this amount is roughly equal to the accuracy limits of the data, we corrected all of the measurements for conversion. The correction procedure begins by estimating $x_0(t)$, the *ortho* fraction of the sample at the time of the measurement. An expression for this is obtained by integrating the rate equation

$$\frac{dx_0}{dt} = -0.019 x_0^2 \text{ h}^{-1}, \quad (3)$$

which had been determined from previous measurements in the cell at 15 K and 30 bar using gaseous thermal conductivity [8] for the x_0 determination. The rate constant is somewhat greater than the value, 0.0114 h⁻¹, given by Woolley et al. [9]. The integration constant, $x_0(0) = 0.75$, is set by assuming the normal spin mixture at the time of the sample loading. The calcu-

lated *ortho* fraction in eq (3) gives the rate of conversion, which we multiply by the duration of each measurement and by $\Delta V/V\Delta x_0 = 7.4 \times 10^{-3}$. The latter value was derived from the data of Scott and Brickwedde [5] and checked by direct measurements in this cell at 15 K and 34 bar.

The results of this work are listed in table 1. Included with them are 14 additional data (mostly on *p*-H₂), which were taken about 18 months before the main study on *n*-H₂. We estimate the accuracy of the expansion data to be about ± 1 or 2%. For comparison with results of others, we have also plotted the *n*-H₂ data in figure 1. The solid lines in the plot represent interpolated values for α of *p*-H₂ from the work of Roder et al. [3]. The dashed lines were derived from an equation of state fitted to the PVT data that Johnston et al. [4] reported for *n*-H₂.

3. Discussion

3.1 Comparison with Earlier Liquid *n*-H₂ Data

Of the existing PVT work on liquid *n*-H₂, that of Johnston et al. [4] is the most useful for present comparisons. Their data were taken along seven isotherms between

20.3 and 32.6 K. Temperatures were measured with a copper-constantan thermocouple calibrated against the vapor pressure equation for *n*-H₂ by White et al. [10].

The pressure cell used in the PVT experiment contained a constant, known volume of sample. The molar volume along each isotherm was determined by first filling the cell to the highest pressure (ca. 100 bar) and then decreasing the pressure in steps of about 10 or 20 bar. After each decrease in pressure, measurements were made of the amount of material drawn from the cell. When the sample pressure had been lowered to about 10 bar, the entire quantity of remaining liquid was measured to establish absolute molar volumes for the isotherm. We estimate a relative precision of $\pm 0.03\%$ for volume data within each set at constant temperature. The absolute accuracy of the measurements is probably about $\pm 0.15\%$.

Johnston et al. [4] found their PVT results well represented by an equation of the form

$$P = A_v + T B_v \quad (4)$$

where A_v and B_v are functions of the molar volume only. They gave the following analytical expression for B_v :

Table 1. Measurements of the thermal expansivity of liquid hydrogen.

T(K)	P(bar)	x_0	$\alpha(10^{-3}\text{K}^{-1})$	T(K)	P(bar)	x_0	$\alpha(10^{-3}\text{K}^{-1})$	T(K)	P(bar)	x_0	$\alpha(10^{-3}\text{K}^{-1})$
22.0	5.48	0.42	18.86	20.4	28.52	0.69	12.32	21.6	56.05	0.53	10.96
21.6	5.48	0.42	17.63	20.8	28.52	0.69	12.65	21.2	56.05	0.53	10.74
21.2	5.48	0.42	17.12	22.0	28.52	0.53	13.75	20.4	69.81	0.73	9.58
22.1	5.48	0.31	18.42	21.6	28.52	0.53	13.34	20.0	69.81	0.72	9.48
21.9	5.48	0.31	18.12	21.2	28.52	0.53	13.07	19.6	69.81	0.72	9.24
21.7	5.48	0.31	17.64	20.8	42.28	0.72	11.34	19.2	69.81	0.72	9.06
21.5	5.48	0.31	17.36	20.4	42.28	0.72	11.29	19.2	69.81	0.70	9.46
21.3	5.48	0.31	17.29	20.0	42.28	0.71	11.01	19.6	69.81	0.70	9.18
21.1	5.48	0.31	17.10	19.6	42.28	0.71	10.85	20.0	69.81	0.69	9.36
20.8	14.76	0.74	14.94	19.2	42.28	0.71	10.55	20.4	69.81	0.68	9.49
20.4	14.76	0.73	14.26	19.2	42.28	0.67	10.84	20.8	69.81	0.68	9.70
20.0	14.76	0.73	13.82	19.6	42.28	0.66	10.76	20.8	69.81	0.67	10.03
19.6	14.76	0.72	13.45	20.0	42.28	0.65	10.98	20.8	69.81	0.66	9.74
19.2	14.76	0.72	13.01	20.4	42.28	0.64	11.08	20.8	69.81	0.66	10.01
18.8	14.76	0.72	12.60	20.8	42.28	0.63	11.43	14.5	6.16	0.002	10.28 ^a
18.8	14.76	0.69	12.76	20.8	56.05	0.74	10.73	15.5	6.16	0.002	11.03 ^a
19.2	14.76	0.68	12.88	20.4	56.05	0.73	10.32	16.5	6.16	0.002	11.77 ^a
19.6	14.76	0.67	13.25	20.0	56.05	0.73	10.12	14.4	15.43	0.02	9.69 ^a
20.0	14.76	0.66	13.68	19.6	56.05	0.72	9.96	15.5	37.13	0.002	9.01 ^a
20.4	14.76	0.65	14.10	19.2	56.05	0.72	9.70	16.5	37.13	0.002	9.44 ^a
20.8	14.76	0.64	14.45	19.2	56.05	0.69	9.59	15.6	50.66	0.002	8.58 ^a
20.8	28.52	0.74	13.20	19.6	56.05	0.68	9.83	15.6	50.66	0.02	8.51 ^a
20.4	28.52	0.73	12.52	20.0	56.05	0.67	9.94	15.6	50.66	0.20	8.53 ^a
20.0	28.52	0.73	12.22	20.4	56.05	0.66	10.07	16.5	69.81	0.002	8.01 ^a
19.6	28.52	0.73	11.92	20.8	56.05	0.65	10.44	17.5	69.81	0.002	8.37 ^a
19.2	28.52	0.73	11.63	21.2	56.05	0.72	10.62	18.5	69.81	0.002	8.69 ^a
19.2	28.52	0.71	11.49	21.6	56.05	0.72	10.93	19.25	69.81	0.002	9.02 ^a
19.6	28.52	0.70	11.82	22.0	56.05	0.72	11.26	19.75	69.81	0.002	9.32 ^a
20.0	28.52	0.70	12.10	22.0	56.05	0.53	11.12				

^aMeasurements on liquid hydrogen taken about 18 months before the main study.

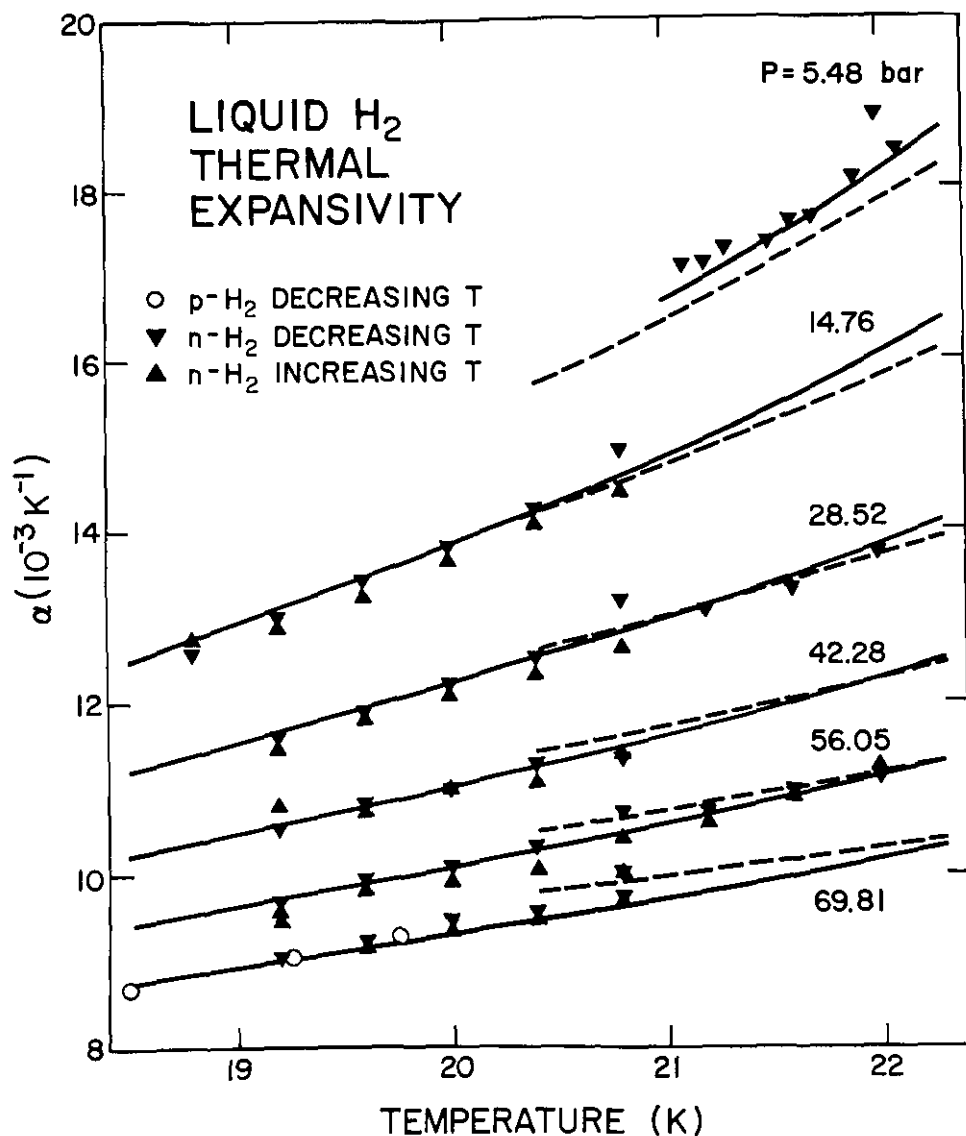


Figure 1--Thermal expansion coefficient of liquid hydrogen. Solid lines represent values interpolated from the p - H_2 data of Roder et al. [3]. The dashed lines were obtained by differentiating eq (4) with B_v and A_v defined by eq (6). The parameter values were derived from a least-squares fit to the data of Johnston et al. [4].

$$B_v = -7.204 + \frac{442.8}{V} \quad (5)$$

where V is taken in cm^3/mole and B_v is given in bar/K . The corresponding volume relationship of A_v appears in tabular form.

Thermal expansivities can be calculated from eq (4) by taking ratios of the temperature and volume derivatives of the pressure. To do this accurately, though, it is necessary to have both A_v and B_v expressed as analytic functions. We, therefore, fit low-order polynomials in both V and $1/V$ to the tabulated A_v data. Close fits were obtained with several different forms, but none of these curves when combined with eq (5) could accurately reproduce the PVT measurements.

Better results were obtained by fitting the original

PVT data to eq (4) with the following parametric functions:

$$B_v = a_1 + \frac{a_2}{V} \quad (6a)$$

$$A_v = a_3 + \frac{a_4}{V} + \frac{a_5}{V^2} + \frac{a_6}{V^3} \quad (6b)$$

The analysis began by converting the original pressure units to bars and by adjusting the temperatures from the NBS-55 scale to the IPTS-68 [11]. A nonlinear least-squares fit then gave the following parameter values: $a_1 = -7.42290$, $a_2 = 4.53373 \times 10^2$, $a_3 = -3.19037 \times 10^2$, $a_4 = 5.67951 \times 10^4$, $a_5 = -3.02764 \times 10^6$, and

$a_6 = 0.435304 \times 10^8$. The standard and average absolute deviations of the volume measurements from the fitted equation of state are 0.033 and 0.085 cm³/mole, respectively. The deviations are comparable to the $\pm 0.15\%$ accuracy limits estimated for these types of measurements [1]. Similar results are obtained by applying the same analysis to earlier data on liquid *n*-D₂ [12].

Thermal expansivities were calculated from eqs (4) and (6) at pressures corresponding to our isobars. The results, plotted in figure 1 as dashed lines, agree well with our measurements except at low pressures where the calculated values are smaller by about 3%. We regard the overall consistency to be quite satisfactory, particularly in view of the uncertainties expected in this type of analysis. The lowest temperature measurements that Johnston et al. made were at 20.329 K. Fitted functions, and especially quantities calculated from these by differentiation, are subject to large uncertainties near the endpoints of the data domain.

3.2 Comparison with Earlier Liquid *p*-H₂ Data

Goodwin et al. [2] published PVT measurements on liquid *p*-H₂. From these results Roder et al. [3] derived pressure functions of density at integral values of temperature. The latter report also contains tabulated values of temperature and density derivatives of the liquid pressure. We calculated thermal expansion coefficients from these tables and interpolated the results to our measured pressures. The calculations agree with our direct measurements on *p*-H₂. Also, as seen in figure 1 where the calculated values are plotted as solid lines, there is a close correspondence with our measurements on *n*-H₂.

Scott and Brickwedde [5] measured liquid volumes of both normal and *para* hydrogen along the vapor pressure curve below 20.4 K. The differences observed between the two data sets are well understood. Attractive forces among the *ortho* fraction introduce an effective pressure that reduces the molar volume *V*. To our knowledge, there are no direct measurements of molecular quadrupole effects on α or β , but it is instructive to consider

$$\ln V(n\text{-H}_2) \approx \ln V(p\text{-H}_2) - \frac{\Delta V}{V(p\text{-H}_2)}, \quad (7)$$

which is correct to first order in the absolute volume difference ΔV . Differentiation of eq (7) gives

$$\Delta\alpha \equiv \alpha(n\text{-H}_2) - \alpha(p\text{-H}_2) = -\frac{\partial}{\partial T} \left(\frac{\Delta V}{V} \right)_p, \quad (8a)$$

$$\Delta\beta \equiv \beta(n\text{-H}_2) - \beta(p\text{-H}_2) = \frac{\partial}{\partial P} \left(\frac{\Delta V}{V} \right)_T. \quad (8b)$$

Figure 2 is a plot of $\Delta V/V$, the fractional volume difference between the *n*-H₂ data of Johnston et al. [4] and the interpolated smooth functions for *p*-H₂ by Roder et al. [3]. We believe the *n*-H₂ data are sufficiently precise that empirical estimates of $\Delta\beta$ can be obtained from this plot. In the noncritical regime all data follow the same general trend. The slope of the 20.4-K isobar, in particular, suggests that $\Delta\beta = -0.025 \times 10^{-3} \text{ bar}^{-1}$. For an average compressibility of $1.3 \times 10^{-3} \text{ bar}^{-1}$, the effect is about 2%. We measured liquid compressibilities of both *n*-H₂ and *p*-H₂ at various pressures and temperatures and confirmed that $\Delta\beta$ is no larger than this amount. Similar estimates for $\Delta\alpha$ are not possible because of the inaccuracies involved in measuring the amounts of *n*-H₂ used in different sample loadings [4].

It is interesting to compare these empirical results with theoretical estimates of $\Delta V/V$. Driessen et al. [13] provide a prescription for calculating the effective pressure P_Q introduced by the electric quadrupole interactions of the *ortho* species. From this and the compressibility, the fractional change in the liquid volume is calculated from

$$\frac{\Delta V}{V} = -\beta P_Q. \quad (9)$$

Although Driessen et al. [13] developed their formula to calculate fractional volume changes in the solid, we shall assume the procedure is valid for the liquid systems as well.

We calculated $\Delta V/V$ for liquid hydrogen along the five isotherms shown in figure 2. At low pressures the calculated isotherm at 20.359 K extrapolates to 5.6×10^{-3} , which agrees with the experimental result of Scott and Brickwedde [5]. At pressure, the calculated results are generally higher by about 2.5×10^{-3} than those found empirically. At 100 bar, for example, the theoretical values fall in the range 3 to 4×10^{-3} . Despite this difference the empirical and theoretical results are reasonably consistent. A discrepancy of 0.25% is roughly equal to the combined limits of error for the two liquid PVT experiments [3,4].

The slopes of the calculated $\Delta V/V$ versus *P* are qualitatively similar to those observed experimentally. In the noncritical regions, the calculated slopes are consistently smaller although probably not significantly so. The average theoretical $\Delta\beta$ indicated for the 20.359-K isotherm is about $-0.018 \times 10^{-3} \text{ bar}^{-1}$, compared to the experimental value -0.025×10^{-3} found above.

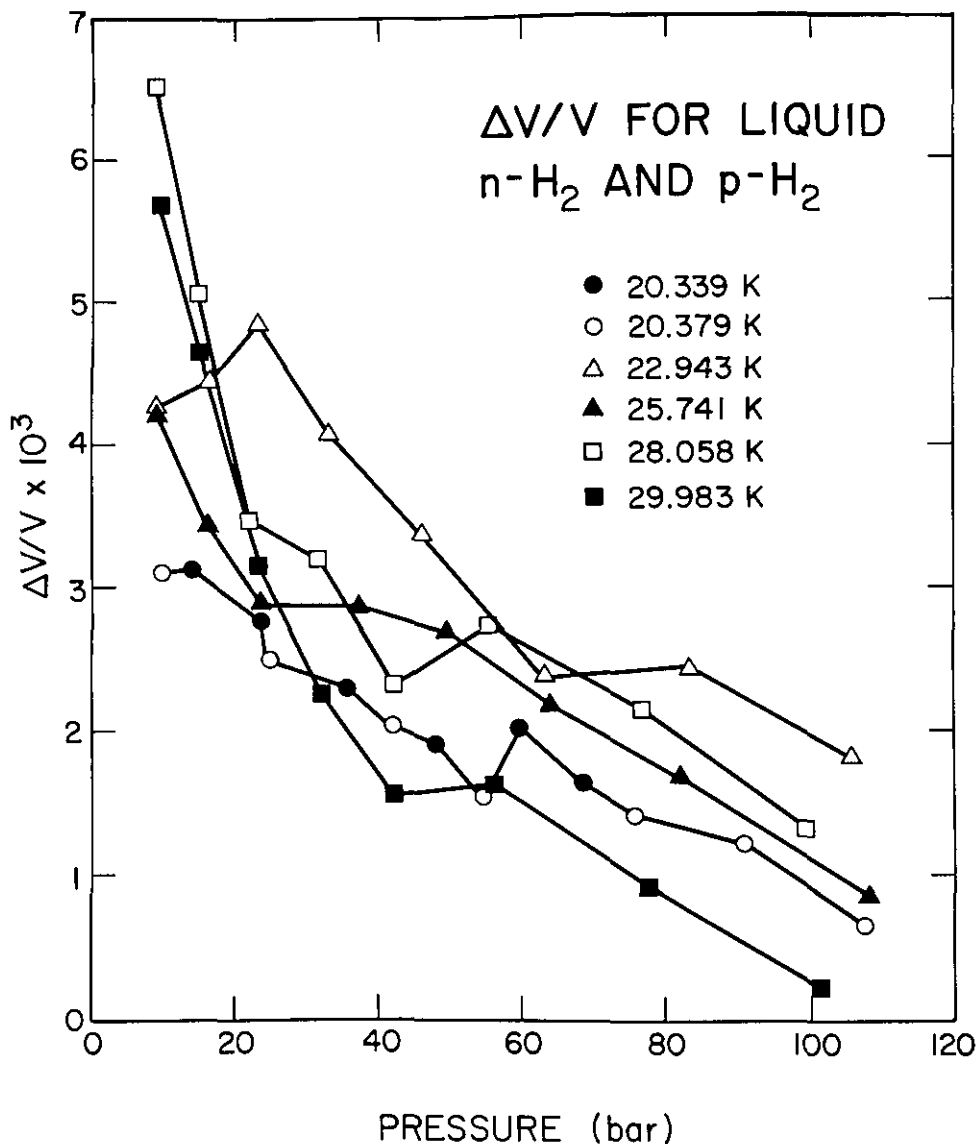


Figure 2—Plot of the fractional difference between the molar volumes of liquid normal and *para* hydrogen. The *n*-H₂ data were taken from [4]. The *p*-H₂ volumes were interpolated from the smooth curves of [3]. Isotherm temperatures are given on the IPTS-68.

The difference between $\alpha(n\text{-H}_2)$ and $\alpha(p\text{-H}_2)$ can be calculated from eqs (8a) and (9). Resulting values for $\Delta\alpha$ are on the order of $0.10 \times 10^{-3} \text{ K}^{-1}$ or less in the temperature range 19 to 22 K for pressures below 20 bar. At higher pressures, 40 to 80 bar, the difference increases to between 0.10 and $0.16 \times 10^{-3} \text{ K}^{-1}$, which is still only about a 1.5% effect. If the empirical $\Delta\beta$ at higher temperatures suggests that our theoretical estimates for quadrupole effects on α and β are too small, even by a factor of two, the magnitude of $\Delta\alpha$ would still not exceed 2 or 3% for the range of temperatures and pressures of the present investigation. Therefore, within the combined limits of experimental error, the expansion data for *p*-H₂ agree with our direct results on *n*-H₂.

4. Summary and Conclusion

We measured thermal expansion coefficients of liquid normal hydrogen in the temperature range 18.8 to 22.2 K and at pressures between 5 and 70 bar. The data are corrected for volume changes due to *ortho-para* conversion. Included with the tabulated expansivity data are calculated estimates of the *ortho* fractions of the samples at the time of the measurements.

To compare our results with the *n*-H₂ data of Johnston et al. [4], we first converted the pressure units of the earlier data to bars and adjusted the temperatures to the IPTS-68. A least-squares fit defines an empirical

function for the liquid pressure. Thermal expansivities calculated from this function agree with present direct measurements except at the lowest pressures.

We discussed the effects of molecular quadrupole interactions among the *o*-H₂ fraction of the normal spin mixture. Comparisons between earlier data on normal [4,5] and *para* [3,5] hydrogen show that experimental $\Delta V/V$ are reasonably consistent with theoretical estimates based on the prescription of Driessen et al. [13]. Empirical and theoretical estimates of $\Delta\beta$ also agree; both suggest that quadrupole interactions decrease the compressibility by about 2%. Comparable estimates of $\Delta\alpha$ are not possible because of the inaccuracies in the *n*-H₂ data. However, calculated estimates in our temperature and pressure domain suggest that $\Delta\alpha$ is no larger than 2 or 3%. The observed agreement between our measured α on *n*-H₂ and corresponding *p*-H₂ data derived by Roder et al. [3] corroborate theoretical estimates of $\Delta\alpha$.

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