

Neutron diffraction by solid ortho-deuterium at pressures up to 25 kbar

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Neutron diffraction investigations have been carried out on solid orthodeuterium at temperatures between 4.2 and 120 K and pressures from 0 to 25 kbar. Unlike the theoretical suggestion [B. L. Holian, Phys. Rev. **B18**, 4780 (1978)] that a structural phase transition induced by pressure is possible, only a hexagonal close packed structure for the ortho-D₂ crystal was observed throughout the whole range investigated, with a ratio of the parameters c/a close to the ideal value. The low-temperature equation of state of orthodeuterium was determined with an accuracy $\Delta V/V$ equal to 0.2–0.4% on the basis of the diffraction results. A comparison with the results for parahydrogen [S. N. Ishmaev *et al.*, Sov. Phys. JETP **57**, 228 (1983)] shows that the results for both isotopes can be represented by a single law of corresponding states $P^i(V)/B_0^i = f(V/V_0^i)$, where B_0^i is the initial bulk modulus and V_0^i is the specific volume at zero pressure for the corresponding isotope. Equations of state for solid deuterohydrogen and tritium are predicted on the basis of a model in which the contribution from the zero point vibrations to the total energy of the crystal is proportional to $(M_i)^{-1/2}$ (M_i is the isotopic mass).

1. INTRODUCTION

Orthodeuterium (molecular spin $I = 0$ or 2), as parahydrogen and paratritium ($I = 0$), is one of the thermodynamically stable modifications of molecular hydrogen at low temperatures (rotational quantum number $J = 0$). Such molecules are spherically symmetrical and isotropic central intermolecular interaction predominates in the cryocrystals formed from them. In spite of the relative simplicity of these molecular crystals, they possess quantum properties: the amplitudes of the zero-point vibrations constitute a noticeable fraction of the periodicity of the crystal lattice, as a result of which it seems to be expanded and has a high compressibility. The hydrogen isotopes are noticeably different from one another in mass, so that the comparative study of their properties in the condensed state, especially on changing the pressure, permits a clearer revelation of the role of zero-point vibrations to be made and the nature of the interaction potential to be studied over a rather wide range of intermolecular distances.

In our earlier paper¹ we reported on neutron-scattering structural investigations of the phase diagram of solid parahydrogen and the determination of its low-temperature ($T \rightarrow 0$) equation of state to rather high accuracy. We give here results for solid orthodeuterium. One of the problems for the experiment was to answer the question: does a structural phase transition take place at high pressures from the hexagonal close packed (hcp) to face centered cubic (fcc) lattice for molecular hydrogen modifications with $J = 0$? The possibility of such a transition was predicted, in particular, by the theoretical work of Holian² (for orthodeuterium, a boundary between the phases was indicated at about 9 kbar). In the case of parahydrogen our diffraction experiments¹ did not confirm these suggestions. As a result, a

search for a phase transition in solid orthodeuterium did not offer much promise. Nevertheless, the results of the present study are very important since deuterium has an appreciably greater coherent-neutron scattering cross-section, and the appearance of a new crystalline phase in this case could be recorded more sensitively. A fairly broad region of its P - T diagram was explored using neutron diffraction: from 4.2 to 120 K and from 0 to 25 kbar. The results obtained were used to construct the low-temperature equation of state of ortho-D₂. A comparative analysis of results for the light and heavy isotopes enable an extrapolation, reasonable in our view, to be made for predicting the equations of state of solid deuterohydrogen and tritium.

2. SOME EXPERIMENTAL DETAILS

The structural investigations of solid orthodeuterium under pressure were carried out by the time-of-flight neutron diffraction method, using the pulsed source of the "Fakel" accelerator (I.V. Kurchatov Institute of Atomic Energy). The experimental method was similar to that used earlier^{1,3,4} except for two improvements.

Deuterium is a relatively expensive material, in contrast to hydrogen. An adsorption chamber containing an intermetallic deuteride (LaNi₅)D₆ was therefore connected to the gas handling system of the apparatus³ to obtain pure gaseous D₂, to reduce its losses and for collection at the end of an experiment. The properties of this compound (see, for example, van Mal *et al.*⁵) enable deuterium gas to be liberated easily on relatively mild heating and to be absorbed efficiently when the temperature is lowered. The deuterium of normal composition (67% ortho-D₂, 33% para-D₂) obtained in this way first entered a catalyst unit (Fe(OH)₃) where its para \rightarrow ortho conversion took place. The ortho-D₂ was then

fed to the specimen chamber where at low temperatures its liquefaction, freezing and compression were carried out. Two apparatus were used:¹ one with helium gas pressure transmission ($P = 0-0.6$ kbar, $T = 4.2-14$ K) and the other with a hydraulic press and a piston-cylinder type high-pressure chamber ($P = 1-25.3$ kbar, $T = 15-120$ K).

On crystallizing hydrogen and deuterium, especially at zero pressure, large-grained specimens with noticeable texture are often obtained. This sometimes raises difficulties for accurate structural analysis. In the apparatus in which pressure was transmitted to the specimen by helium gas along a capillary,¹ a device was made which enabled the chamber with the specimen in the helium cryostat to be rotated automatically through $\pm 350^\circ$ while the neutron diffraction pictures were being taken. This made effective averaging over the volume of the specimen possible with a noticeable reduction in the effect of the texture on the measured diffraction picture. There was also a polycrystalline germanium calibration specimen in the cryostat, so that the time-of-flight scale of the diffractometer could be calibrated to an accuracy better than 0.02% in a separate experiment.

Analysis of the diffraction spectra, measured by the time-of-flight method for 16 scattering angles, was carried out using a specially composed library of programs for an IBM SM-4 minicomputer connected on line. The algorithm made it possible to select the method of analytical approximation of the profile of the diffraction peaks and monitor the calculated parameters and their uncertainty using statistical

criteria. As a result, the monitored accuracy in determining the structural parameters was 0.02-0.03%.

3. RESULTS OF THE MEASUREMENTS

It is known that the observation of structural phase transformations at low temperatures can be complicated as a result of weak diffusion and of the hindering of the kinetics of the process. In order to remove this uncertainty we used various means of preparing the specimen, up to its melting under pressure and subsequent cooling to low temperatures at various rates. In a series of experiments the deuterium specimen was at pressures above 20 kbar for 5 days. However, as in the studies of parahydrogen,¹ no traces of a new structural phase were found.

In the pressure range 0-25 kbar and at temperatures 4.2-120 K, crystals of parahydrogen and orthodeuterium have a hexagonal close-packed lattice.

A typical form of neutron diffraction patterns is shown in Fig. 1, measured on solid orthodeuterium at zero and high (25.3 kbar) pressures. The dashed curves indicate the neutron scattering background from the high-pressure chambers made of aluminum and Ti-Zr alloys. The strong effect of a shift in the diffraction pattern with pressure (the deuterium density is approximately doubled in this range) is seen. Up to 6 reflections corresponding to the hcp orthodeuterium lattice could be resolved accurately. On the upper neutron pattern measured at high pressure, the effect of texture is

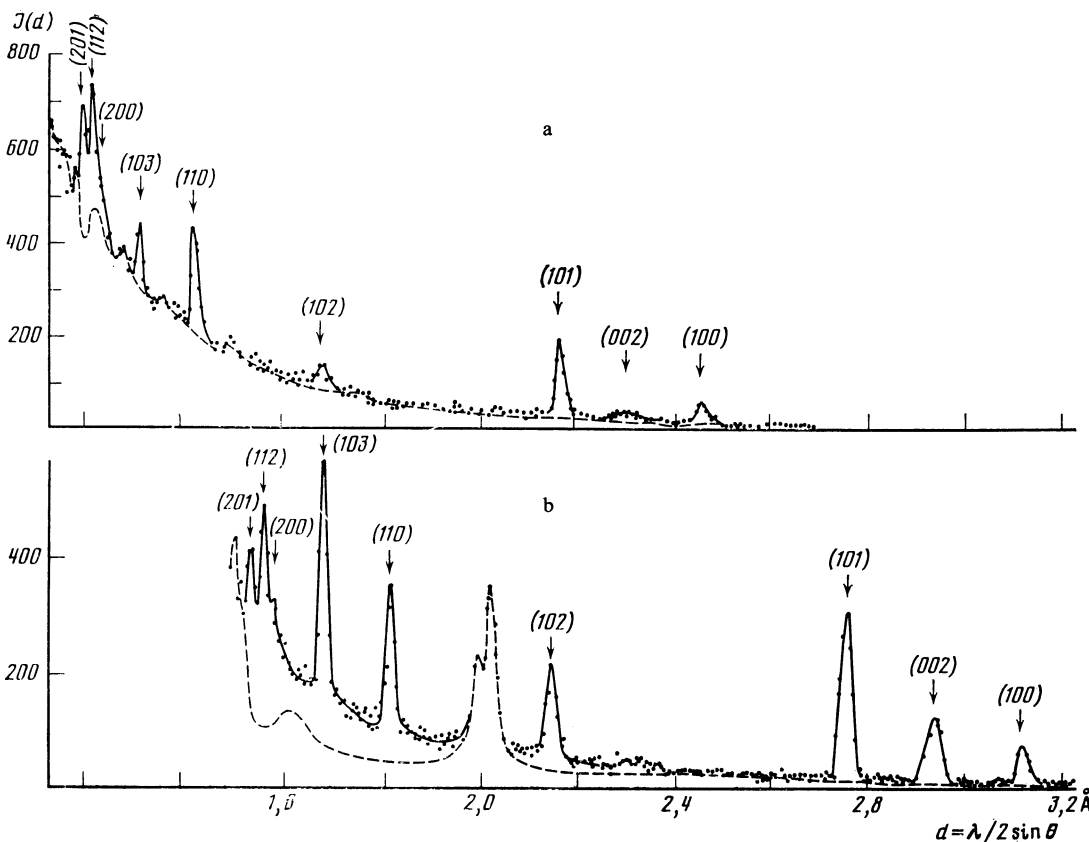


FIG. 1. Neutron diffraction by solid orthodeuterium at $P = 25.3$ kbar, $T = 40$ K (a) and $P = 0$, $T = 4.2$ K (b). The arrows indicate the positions of the reflections from the hcp structure of ortho- D_2 . The background scattering from the chamber material is indicated by the dashed lines.

TABLE I. Parameters of the ortho-D₂ hcp crystal lattice

P, kbar	T, K	a, Å	c, Å	c/a	V, cm ³ /mole
0	4.2	3,605 (1)	5,882 (2)	1,632 (1)	19,933 (15)
0,098 (3)	4,2	3,572 (1)	5,825 (2)	1,631 (1)	19,38 (2)
0,140 (3)	6,6	3,558 (1)	5,801 (2)	1,630 (1)	19,15 (2)
0,237 (3)	8,0	3,538 (1)	5,770 (3)	1,631 (1)	18,83 (2)
0,398 (2)	8,8	3,500 (1)	5,709 (2)	1,631 (1)	18,24 (2)
0,547 (3)	12,3	3,474 (1)	5,666 (2)	1,631 (1)	17,83 (2)
1,89 (2)	16,0	3,320 (3)	5,384 (9)	1,622 (4)	15,47 (9)
3,0 (2)	20,0	3,247 (1)	5,284 (4)	1,627 (2)	14,53 (1)
3,5 (1)	22,0	3,218 (1)	5,277 (4)	1,624 (2)	14,11 (1)
4,1 (1)	22,0	3,189 (1)	5,194 (4)	1,629 (2)	13,78 (1)
4,4 (1)	21,0	3,172 (1)	5,162 (3)	1,627 (1)	13,54 (1)
4,9 (2)	22,5	3,159 (1)	5,150 (1)	1,630 (1)	13,40 (1)
7,4 (3)	20,0	3,083 (1)	5,028 (2)	1,631 (1)	12,46 (1)
7,5 (2)	23,0	3,074 (1)	5,010 (2)	1,630 (1)	12,36 (1)
8,1 (3)	23,0	3,067 (1)	4,999 (1)	1,630 (1)	12,27 (1)
9,1 (2)	20,0	3,038 (1)	4,955 (3)	1,631 (1)	11,93 (1)
11,0 (3)	23,0	2,999 (2)	4,888 (4)	1,630 (2)	11,47 (2)
12,0 (3)	23,0	2,982 (1)	4,859 (2)	1,629 (1)	11,26 (1)
12,4 (3)	25,5	2,976 (1)	4,846 (2)	1,628 (1)	11,19 (1)
13,4 (3)	25,5	2,960 (1)	4,822 (2)	1,629 (1)	11,02 (1)
14,5 (3)	22,0	2,942 (1)	4,790 (2)	1,628 (1)	10,81 (1)
18,4 (7)	40,0	2,891 (1)	4,717 (2)	1,632 (1)	10,28 (1)
21,9 (5)	40,0	2,861 (1)	4,656 (2)	1,627 (2)	9,94 (1)
23,4 (6)	40,0	2,835 (1)	4,620 (2)	1,630 (1)	9,68 (1)
25,3 (5)	40,0	2,826 (1)	4,597 (2)	1,627 (1)	9,57 (1)

clearly revealed by comparison of the intensities of the (101), (102) and (002) reflections. The relative widths of the diffraction peaks did not change with pressure. This is evidence of the uniformity of its distribution throughout the specimen volume.

The crystal lattice parameters of orthodeuterium as a function of pressure are shown in Table I. The uncertainties in the corresponding values in the last place after the decimal point are shown in parentheses. It can be concluded that the ratio of the parameters c/a is close to the ideal value $\sqrt{8/3}$ for a hexagonal close-packed structure, as for solid parahydrogen.¹

Several diffraction experiments⁶⁻¹¹ carried out earlier on solid deuterium have been published. They all refer to zero pressure (Nielsen⁸ also made a measurement at $P = 275$ bar). These results are compared with ours at $P = 0$

in Table II. It can be seen that the values of the lattice parameters and molar volume V_0 determined in the later experiments agree well (the scatter of the results is less than or of the order of 0.1%).

It was noted when studying parahydrogen¹ that the value of the "zeroth" volume V_0 determined earlier in macroscopic (thermodynamic) experiments deviates noticeably towards higher values from the microscopic (diffraction) results [(23.14 ± 0.08) cm³ · mole⁻¹ (Ref. 15) and even (23.23 ± 0.05) cm³ · mole⁻¹ (Ref. 16)] compared with (23.00 ± 0.02) cm³ · mole⁻¹ (Ref. 1). The value $V_0 = (20.03 ± 0.07)$ cm³ · mole⁻¹ for solid orthodeuterium recently obtained by analyzing the thermodynamic properties,¹⁷ is not all that much different from our neutron diffraction results: (19.933 ± 0.015) cm³ · mole⁻¹.

The results of measurements sufficiently far from the

TABLE II. Lattice parameters, molar volume, bulk modulus and its derivative for solid orthodeuterium at $P = 0, T = 0$

Experimental method	a, Å	c/a	V ₀ , cm ³ /mole	B ₀ , kbar	B' ₀
x-ray diffraction ^{6*}	3,54	1,67	19,31		
Neutron diffraction ⁷	3,610	1,625	19,91		
Neutron diffraction ⁸	3,607	1,629	19,94	4,3	
Neutron diffraction ⁹		1,632	19,988 (13)		
Electron diffraction ¹⁰		1,634	19,95 (11)		
x-ray diffraction ¹¹	3,603	1,633	19,930 (16)		
Piston displacement ¹²				3,26	6,18
Piston displacement ¹³				3,15 (6)	6,88
Ultrasonics ¹⁴				3,37 (20)	5,4 ± 2
Measurement of $P_v(T)$ (Ref. 15) + piston displacement ¹³			19,95	3,18 (3)	6,82
Neutron diffraction, results of the present work	3,605 (1)	1,632 (1)	19,933 (15)	3,347 (53)	6,86 (17)

*Normal D₂ (67% ortho + 33% para)

melting curve, $T/T_m \lesssim 0.3$, are shown in Table I. Under these conditions the contribution to the pressure from thermal lattice vibrations is relatively small. Estimates on the basis of measurement of the $T_v(T)$ isochore¹⁵ show that it is less than or comparable with the accuracy in determining the pressure in the experiment. These neutron diffraction results were used to determine the "zero" ($T \rightarrow 0$) isotherm of the equation of state of orthodeuterium.

4. LOW-TEMPERATURE EQUATION OF STATE OF ORTHODEUTERIUM

The $P(V)$ dependence for $T \rightarrow 0$, obtained experimentally (Table I), was approximated by the Birch expression,¹⁸ using the method of least squares:

$$P(V) = (V_0/V)^{5/3} \sum_{n=1}^N A_n [(V_0/V)^{2/3} - 1]^n. \quad (1)$$

Taking account of more than the first two terms in the series was pointless since all coefficients with $n \geq 3$ turned out to be essentially equal to zero. As a result, the zero isotherm of the equation of state of orthodeuterium was determined by the following equation:

$$P(V) = \alpha\beta(A_1 + A_2\beta), \quad (2)$$

where

$$\begin{aligned} \alpha &= (V_0/V)^{5/3}, & \beta &= (V_0/V)^{2/3} - 1; \\ A_1 &= (5.02 \pm 0.08) \text{ kbar}; & A_2 &= (10.78 \pm 0.2) \text{ kbar} \\ V_0 &= (19.933 \pm 0.015) \text{ cm}^3/\text{mole} \end{aligned}$$

We also give the formula for calculating the bulk modulus

$$B(V) = -V \frac{\partial P}{\partial V} = 3.347\alpha + \alpha\beta(26.087 + 32.34\beta) \text{ kbar}. \quad (3)$$

The uncertainty in determining Eq. (2), expressed in units of $\Delta V/V$, lies between $\pm 2.3 \times 10^{-3}$ and $\pm 4 \times 10^{-3}$ in the pressure range 0–100 kbar.

The parameters A_1 and A_2 can be expressed in terms of such physical quantities as the initial bulk modulus B_0 and its derivative B_0'

$$B_0 = -V \left. \frac{\partial P}{\partial V} \right|_{P=0, T=0} = \frac{2}{3} A_1; \quad (4)$$

$$B_0' = \left. \frac{\partial B}{\partial P} \right|_{P=0, T=0} = - \left. \frac{\partial \ln B}{\partial \ln V} \right|_{P=0, T=0} = \left(4 + \frac{4}{3} \frac{A_2}{A_1} \right). \quad (5)$$

TABLE III. Parameters of the low-temperature equation of state (6) for isotopic modifications of molecular hydrogen

Isotope	V_0 , cm ³ /mole	B_0 , kbar	B_0'	
H ₂	23.00 (2)	1.557 (30)	7.03 (28)	Experiment ¹
HD	20.96 (3)	2.71 (3)	6.93 (18)	Estimate (12)
D ₂	19.933 (15)	3.347 (53)	6.86 (17)	Experiment (present work)
T ₂	18.86 (5)	4.23 (20)	6.79 (52)	Estimate (12)

Equation (2) then takes the following form:

$$P(V) = \alpha\beta B_0 [^{3/2} + ^{9/8} (B_0' - 4)\beta]. \quad (6)$$

The values of B_0 and B_0' , together with other parameters of the ground state of orthohydrogen at $P = 0$ are shown in Table II. The results obtained do not agree badly with earlier experimental determinations^{13,15} of the zero isotherm of deuterium, mainly based on the piston displacement method. At $V = 15 \text{ cm}^3 \cdot \text{mole}^{-1}$ the difference in determining the pressure is 0.1 kbar, and is about 1 kbar for $V = 10 \text{ cm}^3 \cdot \text{mole}^{-1}$.

Comparison of our equation of state with the theory,¹⁹ based on a semi-empirical isotropic pair potential with a correction for three-particle interaction, shows that the departure does not exceed 0.5% in units of $\Delta V/V$ in the pressure range 0–100 kbar.

5. THE LAW OF CORRESPONDING STATES FOR SOLID HYDROGEN ISOTOPES

If the law of corresponding states is valid for a certain class of materials $\{i\}$, i.e., the volume dependence of their total binding energy is of a universal form:

$$E^i(V) = E_0^i(V_0^i) f(V/V_0^i), \quad (7)$$

then the ratio of the values of pressure for a given V to the initial bulk modulus is a function which depends only on the volume ratio V/V_0 :

$$\frac{P^i(V)}{B_0^i} = - \frac{\partial E^i}{\partial V} \frac{1}{B_0^i} = - \frac{f'(V/V_0^i)}{f''(1)} \equiv \varphi \left(\frac{V}{V_0} \right). \quad (8)$$

The satisfaction of this law can easily be tested using the Birch equation of state as given by Eq. (6). In this case the value of the derivative of the initial bulk modulus B_0' should be one and the same for the given class of materials.

We shall test the applicability of this law to solid hydrogen and deuterium. Using the data of Table II of Ishmaev *et al.*¹ and Eq. (5), we find $B_0' = (7.03 \pm 0.28)$ for parahydrogen, which on taking account of the errors is very close to the value (6.86 ± 0.17) determined for orthodeuterium in the present work. The manifestation of quantum effects due to the large contribution of zero-point vibrations in molecular hydrogen and deuterium crystals is fairly evident: their equilibrium volumes at $P = 0$ differ by 15%, and the initial compressibility of parahydrogen is 1.8 times greater than that of

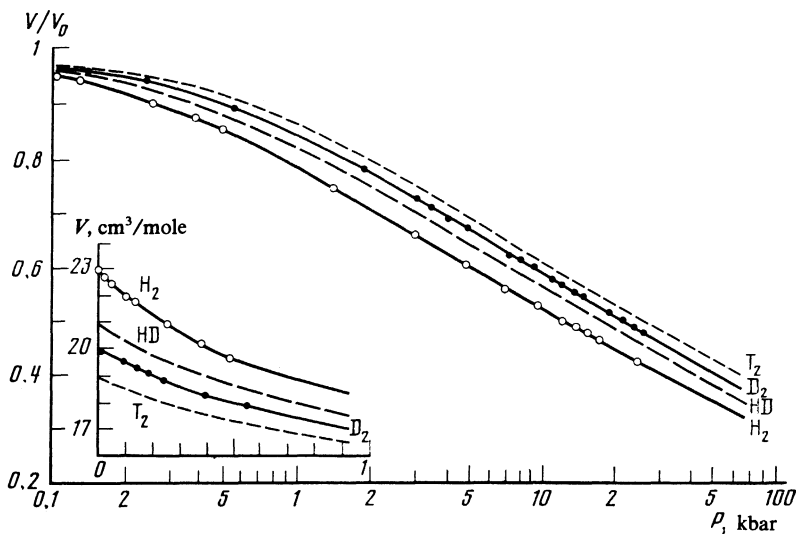


FIG. 2. The compressibility of the solid molecular hydrogen isotopes at $T=0$. The low-pressure region is shown on the inset in a linear scale.

orthodeuterium. It turns out, nevertheless, that their zero-point isotherms can be represented, with good accuracy, within the framework of a single law of corresponding states. It should be noted in this connection that the values of B'_0 are close to 7 not only for more classical inert gas crystals [$B'_0 = 7.2$ (Ref. 20)] but also for solid helium [$B'_0 = 6.9$ (Ref. 21)] which is more quantumlike than hydrogen.

6. ESTIMATE OF THE EQUATIONS OF STATE OF OTHER HYDROGEN ISOTOPES

It is now possible to try to predict the equations of state at $T=0$ for other isotopic modifications of molecular hydrogen which have not been investigated: deuterohydrogen HD and paratritium T_2 . If the interaction potential is identical, the total energies of crystals of different isotopes $E^i(V)$ at $T=0$ only differ in the contribution of the zero-point vibrations:

$$E^i(V) = E_s(V) + E_z^i(V), \quad (9)$$

where $E_s(V)$ is the potential or static lattice energy, while $E_z^i(V)$ is the zero point energy of a crystal of the i th isotope. In the harmonic approximation

$$E_z^i(V) \propto \hbar \langle \omega \rangle^i(V)/2, \quad (10)$$

where $\langle \omega \rangle^i(V)$ is the mean frequency of the excitation spectrum. Assuming that $\langle \omega \rangle^i \propto (M_i)^{-1/2}$, where M_i is the mass of the i th isotope, we obtain the following expression for the total energy:

$$E^i(V) = E_s(V) + E_z(V) (M_i)^{-1/2} \quad (11)$$

and, correspondingly, for the pressure:

$$P^i(V) = -\frac{\partial E^i}{\partial V} = P_s(V) + P_z(V) (M_i)^{-1/2}. \quad (12)$$

It is easy to determine the contributions to the pressure from the potential energy $P_s(V)$ and the zero point vibrations $P_z(V)(M_i)^{-1/2}$, using the results obtained for hydrogen

and deuterium. It is also not difficult to estimate the parameters B_0 and B'_0 of the low-temperature equation of state for HD and para- T_2 on the basis of Eqs. (6) and (12). These values are given in Table III together with the experimental results for para- H_2 (Ref. 1) and ortho- D_2 . The zero point isotherms of the equation of state for isotopic modifications of molecular hydrogen, obtained in this way, are given in Fig. 2.

Values of the equilibrium zero-point volume V_0^i were found from the condition $P^i(V) = 0$. The value of V_0 found by us for solid HD agrees well with existing electron diffraction experimental results:¹⁰ (20.58 ± 0.2) and $(20.95 \pm 0.11) \text{ cm}^3 \cdot \text{mole}^{-1}$. The value of V_0 found for solid para- T_2 is close to a similar recent calculated value:²² $18.77 \text{ cm}^3 \cdot \text{mole}^{-1}$. The single experimental result so far for solid tritium, calculated from results on x-ray diffraction,⁶ gave a value $18.21 \text{ cm}^3 \cdot \text{mole}^{-1}$.

It follows from our results that the minimum of the lattice potential energy ($P_s(V) = 0$) for the solid hydrogen isotopes occurs at a value $V_s = (15.56 \pm 0.2) \text{ cm}^3 \cdot \text{mole}^{-1}$, which is appreciably less than their equilibrium values at $P = 0$. The value of the bulk modulus of hydrogen at $V = V_s$ is comparable with typical values for the solid inert gases at $P = 0$. The zero-point vibrations thus expand the crystal appreciably and determine the main differences in compressibility of the solid isotopes of molecular hydrogen.

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