

TABLE I. Lattice constants, second moments and linewidths for H₂ and D₂ at zero pressure.

Method	Structure	<i>a</i> (Å)	<i>c</i> (Å)	Volume per molecule (Å ³)	<i>M</i> ₂ (kc/sec) ² (theor)	<i>M</i> ₂ (kc/sec) ² (exper)	Width (G)
H ₂							
Density ^a				37.5			
X rays ^b	hcp	3.75	6.12	37.2	67.6		
X rays ^c	hcp(?)	3.75	6.49	39.5	60.5(?)	75±1	6.0 ±0.2
X rays ^c	bct	4.57	3.75	39.2	62.2		
Neutrons ^d	bct	4.5	3.6	36.5	70.1		
X rays ^e	hcp	3.78	6.16	38.1	64.5		
D ₂							
Density ^a				32.5			
X rays ^c	bct	3.38	5.86	33.5	0.126	0.28±0.02	1.53±0.1
Neutrons ^d	bct	3.38	5.60	31.9	0.135		
X rays ^e	hcp	3.54	5.91	32.1	0.134		

^a Reference 17.^b Reference 20.^c Reference 23.^d Reference 24.^e Reference 25.

concentration.^{13,14} Since the ortho-para conversion depends on magnetic interactions, it is expected to increase strongly with density.

High-pressure experiments were initiated on H₂ by McCormick and Fairbank and preliminary results were reported.¹⁵ In that work the expected effects mentioned above were essentially observed. After an interruption of two years, the work was started again by the present authors who used some of the high-pressure equipment designed and used by McCormick and Fairbank. The new results and their discussion form the subject of this paper.

Section 2 reviews some properties of solid H₂ and D₂. The proposed crystalline structures for both isotopes are described. The different theories for the occurrence of the transition point *T*_λ as a function of ortho concentration and structure are also reviewed. The theoretical second moment for both isotopes is calculated, assuming a rigid lattice. In Sec. 3 a description of the experimental arrangement is given. The method of measuring the ortho-para conversion rate in H₂ and the second moment for a given *c* and density *ρ*, and the technique for determining *T*_λ, are described. Finally, in Sec. 4 the results are presented and discussed for each isotope separately. The line shape in the region between 4.2 and *T*_λ is presented and the second moment is compared to theory. The data of *T*_λ versus density are compared with the recent specific heat data of Ahlers and Orttung.¹⁶

II. REVIEW OF SOME PROPERTIES OF SOLID H₂ AND D₂

A. The Crystalline Structure

Bulk measurements on solid H₂ and D₂ include the density data at 4°K by Megaw¹⁷ for pressures up to

¹³ K. Tomita, Proc. Phys. Soc. (London) A68, 214 (1955).¹⁴ G. M. Bell and W. M. Fairbairn, Mol. Phys. 4, 481 (1961); 5, 605 (1962).¹⁵ W. D. McCormick and W. M. Fairbank, Bull. Am. Phys. Soc. 3, 166 (1958); W. D. McCormick, thesis, Duke University, 1959 (unpublished).¹⁶ G. Ahlers and W. H. Orttung, Phys. Rev. 133, A1642 (1964).¹⁷ H. D. Megaw, Phil. Mag. 28, 129 (1939).

about 100 atm and the compressibility data of Stewart and Swenson^{18,19} for pressures up to 10 000 atm. In Megaw's measurements the initial ortho concentration in H₂ was 0.55. Stewart and Swenson did not specify *c* but presumably it must have been between 0.50 and 0.75. Information on the crystalline structure of these isotopic solids has been gathered from several x-ray and neutron-diffraction experiments as well as from infrared data. Keesom, de Smedt, and Mooy²⁰ reported in 1931 the study of pure para hydrogen. Their results were interpreted as indicating a hexagonal close packed (hcp) structure. Their interpretation was substantiated by recent infrared data.^{21,22}

There have been several x-ray investigations on H₂ and D₂ with *c*=0.75 and 0.67, respectively,²³ which seemed to indicate for both isotopes a body centered tetragonal (bct) structure and neutron-diffraction work confirmed this interpretation.²⁴ However, very recently, x-ray determinations by Kogan and co-workers²⁵ have again been analyzed in terms of a hcp lattice for both H₂ and D₂. Hence, this structure seems well established for both pure para and ortho-para mixtures. The lattice constants from all published sources are presented in the four first columns of Table I.

¹⁸ J. W. Stewart, Phys. Rev. 97, 578 (1955); J. Phys. Chem. Solids 1, 146 (1956).¹⁹ J. W. Stewart and C. A. Swenson, Phys. Rev. 94, 772 1069 (1954).²⁰ W. H. Keesom, J. de Smedt, and H. H. Mooy, Commun. Kamerlingh Onnes Lab. Univ. Leiden, 19, 35 (1931).²¹ H. P. Gush, W. F. J. Hare, E. J. Allin, and H. L. Welsh, Can. J. Phys. 38, 176 (1960).²² J. Van Kranendonk and H. P. Gush, Phys. Letters 1, 22 (1962).²³ V. S. Kogan, B. G. Lazarev, and R. F. Bulatova, Zh. Eksperim. i Teor. Fiz. 37, 678 (1959) [English transl.: Soviet Phys.—JETP 10, 485 (1960)].²⁴ V. S. Kogan, B. G. Lazarev, R. P. Ozerov, and G. S. Zadanov, Zh. Eksperim. i Teor. Fiz. 40, 1022 (1961) [English transl.: Soviet Phys.—JETP 13, 718 (1961)].²⁵ V. S. Kogan, A. S. Bulatov, and L. F. Yakimenko, Zh. Eksperim. i Teor. Fiz. 46, 148 (1964) [English transl.: Soviet Phys.—JETP 19, 107 (1964)].