Physics. — *On the Crystal Structure of Methane.* By H. H. MOOY (Communication N°. 213*d* from the Physical Laboratory Leiden). (Communicated by W. H. KEESOM.)

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§ 1. *Introduction.* The density 0.413 deduced from the crystal structure of solid methane at —252°.7 C. published by MC LENNAN and PLUMMER [1]) is in disagreement with the value 0.522 measured directly by HEUSE [2]).

Therefore a renewed X ray analysis seemed to be desirable, extending thereby the temperature range of the experiments also to temperatures distinctly below 20°.4 K. [3]).

We communicate here our results so far obtained.

§ 2. *Experiments.* We made use of the experimental arrangement that served in investigations on the crystal structure of neon and hydrogen [4]).

For with methane it proved to be very difficult to make an exposure with an apparatus of the type described in Comm. Leiden Suppl. N°. 53*a*. As a matter of fact the gas then solidified in a layer of uniform thickness against the inner wall of the Dewar flask as far as it is cooled with liquid hydrogen. Hence big quantities would be necessary to get a layer of sufficient thickness on the capillary tube. This difficulty (which has never been encountered with *Ar*, N_2 etc.) may possibly be explained by the fact that an anomaly in the heat-capacity of solid methane occurs just at the temperature of liquid hydrogen boiling under normal pressure.

With the metal cryostat mentioned above we made six exposures of solid methane at five different temperatures ; radiation *CuKα*.

We used a quantity of gas taken from a gasholder and purified by distilling it once. Film VI was made just for safety with a quantity that had been distilled thrice.

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[1]) J. C. MC LENNAN and W. G. PLUMMER, Phil. Mag. **7**, 761, 1929. An optical investigation of the crystallographical properties has been made by W. WAHL, Proc. Roy. Soc. (A) **87**, 371, 1912. See also W. WAHL, Proc. Roy. Soc. (A) **90**, 1, 1914.

[2]) W. HEUSE, Z. ph. Ch. (A) **147**. 271, 1930.

[3]) K. CLUSIUS, Z. ph. Ch. (B) **3**, 63, 1929 has observed a transformation at 20°.4 K. HEUSE suggests that there is no change in crystal structure; Zs. physik. Chem. (A) **147**, 282, 1930.

[4]) These Proceedings Comm. Leiden N°. 203*b* and N°. 209*d*.

§ 3. *Results.* The line-distances (in $^1/_{10}$ mm) of the different films are given in:

TABLE I.

	I	II	III	IV	V	VI
*	242	244	242	240	238	242
*	284	284	283	283		—
	300	302	301	300	297	300
	342	343	342	340	338	342
+	478	479	478	—	470	478
	561	563	564	560	558	561
	588	590	590	588	574	587
+	679	674	674	—	—	679
	748	748			731	747
	770	770	766	767	750	768
	847	849			830	847
	906	906			889	907
	1065	1066				1062
	1081	1085				1081
						1164

* Parasitic lines.

+ Concording with lines from the copper rod 478 and 679.

Flim		Temp.		tickness sample
I.		20.35° K.,		2.2 mm
II.		18.6		2.2
III.		21.2		2.9
IV.		14		2.2
V.		± 68		3.1
VI.		20.35		2.4

As the films taken below and above 20°.4 K are concordant it is clear that no change in crystal structure takes place at that temperature. We may add that the relative intensities of the lines on all our films are the same, but that they are different from those of Mc LENNAN and PLUMMER.

Spacings have been calculated in the way described previously [1]).

[1]) These Proceedings Comm. Leiden N°. 209d.

TABLE II.

Line-distance	Correction	θ	d
300	28	13°8′	3.39
342	28	15°10′	2.94
478	31	21°36′	2.09
561	31	25°36′	1.78
588	31	26°55′	1.70
679	31	31°18′	1.48
748	31	34°38′	1.35^4
770	31	35°42′	1.31^8
847	26	39°40′	1.20^6
906	21	42°46′	1.13^3
1065	16	50°41′	0.99^4
1081	16	51°27′	0.98^4
1164	14	55°33′	0.93^3

TABLE III.

Indices	d measured	d calculated	Intensities observed	Intensities for a close-packed structure of atoms.
111	3.39	3.40	v.s.	15
200	2.94	2.94	s.	8.6
220	2.09	2.08	w?	7.8
311	1.78	1.78	m	10.6
222	1.70	1.70	w	2.9
400	1.48	1.47	v.w.	1.6
331	1.35^4	1.35^1	w	5.5
240	1.31^8	1.31^8	w	4.9
224	1.20^6	1.20^2	w	4.0
333 511	1.13^3	1.13^4	w	5.0
440		1.04^1	—	1.8
531	0.99^4	0.99^5	m	3.5
600 442	0.98^4	0.98^2	m	4.4
620	0.93^3	0.93^1	w	3.3

§ 4. *Translation-lattice and size of elementary cell.* Using a Hull-Davey graph we found that the observed spacings are compatible with those of a face-centred cubic translation lattice, side of the elementary cube $a = 5.89 \pm 0.01$ A.U. (See Table III).

With four molecules in the elementary cube the calculated density is 0.517 ± 0.006, which is in good agreement with HEUSE's value 0.522 ± 0.001.

The observed intensities disagree slightly from those to be expected from a cubic close-packed structure of atoms; showing that the molecular structure of CH_4 must be taken into consideration. So we can not support the idea of a neon-like constitution of the methane-molecule in the solid phase proposed by MC LENNAN and PLUMMER.

The possibility exists that the parameter distance C-H can be fixed so that a still better agreement between observed and calculated intensities is obtained.