

Physics. — “*On the diffraction of Röntgen-rays in liquids*”. II.

By Prof. W. H. KEESOM and Prof. J. DE SMEDT. (Communication N°. 12 from the Laboratory of Physics and Physical Chemistry of the Veterinary College). (Communicated by Prof. H. KAMERLINGH ONNES).

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§ 1. *Introduction.* The experiments on the diffraction of Röntgen-rays described in Comm. N°. 10¹⁾ were all made with K α -rays of copper. No diffraction ring was observed caused by the interference of rays scattered by the separate atoms in the molecules. F.i. in the case of oxygen this might be ascribed to the circumstance, that the distance of the centres of the systems of electrons grouped round the atom nuclei is too small to give an interference ring with rays of that wave length (viz. smaller than 0.95 Å for $\lambda = 1.54$ Å). Therefore it seemed desirable to repeat some of the experiments with rays of a shorter wave length.

We have now made several observations with K α -rays of molybdenum ($\lambda = 0.71$ Å).

§ 2. For method and apparatus see Comm. N°. 10. The rays emitted by the molybdenum anticathode were filtered by 0.35 mm. zirconium.

§ 3. *Results of the observations on the principal diffraction ring.* We now exposed liquid oxygen, argon and nitrogen, also water and carbonic disulphide.

For oxygen, argon, water and nitrogen (investigated for the first time now) we found confirmed that the principal ring is due to neighbouring molecules, which we may consider to be distributed approximately as spheres packed together as closely as possible and filling up the space occupied by the liquid.

This time we obtained a diffraction ring for carbonic disulphide

¹⁾ These Proceedings 25, 1922, p. 118.

too and this gave a deviating value for the distance between the diffracting particles. This is evident from the following table. Here φ is again the half top angle of the cone formed by the diffracted Röntgen rays. M and d have been written for the molecular weight and density, while

$$a = \frac{7,72 \lambda}{4\pi \sin \frac{\varphi}{2}}$$

denotes the distance between the diffracting particles. Here we again have made the assumption that the observed diffraction ring is due to the cooperation of arbitrarily orientated systems each of two particles at that distance from each other.

TABLE I.				
Substance		φ ($\lambda = 0.71 \text{ \AA}$)	a	$1.33 \sqrt[3]{\frac{M}{d}}$
Oxygen	(9 plates)	12.50°	4.0 \AA	4.0 \AA
Argon	(1 plate)	13.0	3.85	4.1
Water	(1 ")	13.44	3.73	3.6
Nitrogen	(1 ")	11.34	4.42	4.4
Carb. disulph.	(1 ")	13.23	3.8	5.2

Instead of formulating a special hypothesis on the deviating behaviour of CS₂, we prefer to postpone this until more substances showing a similar deviation have been investigated.

The diffraction rings obtained now are sharper than the former ones, the liquids being radiated this time in a tube of 1 mm. diameter.

§ 4. *Results of the observations on the second ring.* On six plates of oxygen and on those of argon and nitrogen the second ring is distinctly measurable. The other plates do not show this ring, probably because the obtained films are less blackened. For argon too this ring is very weak.

TABLE II.		
	φ	a
Oxygen	19.5°	2.57 Å
Argon	18.9	2.65
Nitrogen	17.0	2.95

These values of a show a striking agreement with the values obtained in Comm. N°. 6¹⁾ for the diameter of the molecule²⁾ viz. for oxygen $\sigma = 2.65$ Å, for nitrogen $\sigma = 2.98$ Å. This supports the assumption made in Comm. N°. 10 that this diffraction ring should be due to the collaboration of two molecules touching each other.

With this wave length we also found for water at the outside of the principal ring a rather uniform blackening, rather sharply bounded at $\varphi = 24^\circ$, which corresponds with a distance $a = 2.1$ Å.

§ 5. *For oxygen and nitrogen no diffraction by separate atoms in the molecule.* On a well blackened film of oxygen and on that of argon we found indications of a third maximum of blackening, for oxygen at $\varphi = 29^\circ$ and for argon at $\varphi = 30.5^\circ$. We do not pretend the existence of this third maximum to be doubtlessly fixed by these indications. We only draw the following conclusion: If this third maximum really exists, it also does so for argon, so that this maximum cannot be ascribed to the interference of rays that are scattered by the separate atoms in the molecules.

Though on several films the principal diffraction ring is blackened very intensively, no trace of an interference figure of the separate atoms in the molecule was found in these experiments. Yet with the here used wave length a diffraction ring would have been obtained for a distance of the diffracting particles greater than 0,43 Å³⁾.

For a partial verification of the above we made still an exposition

¹⁾ These Proceedings 23, 1920, p. 939.

²⁾ In fact the smallest distance that is possible between the centres of two molecules in the gas.

³⁾ According to the discussion of the band spectra the distances of the atom nuclei would be for oxygen and nitrogen resp. 0,85 and 1,12 Å: A. EUCKEN, Z S. f. Elektrochemie 26, p. 377, 1920. Comp. W. LENZ, Verh. D. physik. Ges. 21, p. 632, 1919.

with Cu-K $_{\alpha}$ rays (9 mA, \pm 25 KV). Though this film is thoroughly blackened, only two rings have been obtained.

It may be suggested, that the rings obtained in these experiments are all due to atoms that temporarily are arranged in a crystal lattice. The values for the diameters of these rings found in this Comm. exclude a cubical arrangement¹⁾. The data are not sufficient to know, whether those temporary arrangements might belong to a crystal structure from an other class of symmetry²⁾. Meanwhile the fact that freezing takes place suddenly at a definite temperature and the possibility of undercooling do not seem to point in the direction of such temporary crystal arrangements.

Lead by these considerations we have made still a plate of water at $\pm 0,5^{\circ}$ C. The obtained interference figure perfectly agreed with that found at room temperature. At the outward side of the nearly uniform blackening only the intensity proved to be somewhat greater. In this way a second ring develops itself there, an indication of the presence of more double molecules at those low temperatures. No indication was found of the presence of more or greater crystal groups.

¹⁾ Comp. Comm. N^o. 10 p. 122, footnote 1.

²⁾ Nitrogen and argon crystallise cubically: W. WAHL, Proc. Roy. Soc. A 87, p. 371, 1912; oxygen below the melting point first hexagonally: W. WAHL, Proc. Roy. Soc. A 88, p. 61, 1913.
