levels. This ratio was found to be 19 : 40.2 : 43.1 at the L_I limit. This does not agree with the value 19 : 32 : 49 given by Patten[2] for the five elements Au(79) to Bi(83), and neither does it compare well with the ratio interpreted from Wolf's curve for gold; 19 : 70 : 72.

I gratefully acknowledge my obligation to Professor S. K. Allison for the suggestion of the problem and for his helpful suggestions throughout the course of the investigation.

[2] C. G. Patten, Phys. Rev. **45**, 131 (1934).

The Nuclear Spin of Deuterium

G. M. Murphy and Helen Johnston, *Department of Chemistry, Columbia University*

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By the methods of photographic photometry, the alternating intensities in emission of the $\Delta v = 0$ sequence of the Fulcher bands of deuterium have been investigated in order to determine the nuclear spin. The alternation of intensities was determined for the P, Q and R branches of each of the 5 bands studied. The average value of g_s/g_a obtained from 4 plates is 1.97 ± 0.03. This value agrees best with the theoretical ratio of 2 and a consequent nuclear spin of 1. Since the even rotational levels are more intense than the odd levels, the nucleus obeys Bose-Einstein statistics.

THE nuclear spin of rare isotopes such as H^2, N^{15}, O^{17}, O^{18} is of the utmost importance in any theory of the nucleus but because of the low abundance of such nuclear species, it will probably be rather difficult to obtain this information. This is not true, however, for deuterium since it is possible to separate it almost completely from the more abundant protium. Measurements of the alternation of intensities in the molecular spectrum of deuterium seemed to offer the simplest approach to the problem of the determination of its nuclear spin. We[1] have therefore made such measurements on the P, Q and R branches of 5 bands in the $\Delta v = 0$ sequence of the Fulcher bands of deuterium as analyzed by Dieke and Blue.[2]

The nuclear spin of protium is well established as $\frac{1}{2}$. Relative intensity measurements have been made on the protium molecular spectrum by Kapuscinski and Eymers[3] and on the acetylene spectrum by Childs and Mecke.[4] Specific heat measurements at low temperature also give the nuclear spin.[5]

These methods are all applicable to deuterium and in addition we may mention the magnetic deflection method of Breit and Rabi[6] and the study of scattering as proposed by Sexl.[7] Hyperfine structure cannot be used.

Since it is known[8] that the lines in the molecular spectrum of deuterium alternate in intensity, it follows at once that the spin cannot be zero. By analogy with N^{14}, deuterium would probably have a spin of 1, and if its nucleus consists of one proton and one neutron[9] this would be consistent with a spin of $\frac{1}{2}$ for each. This result is also indicated by the conservation of spin momentum in nuclear disintegrations involving deutons[10] and by a study of the ortho-para deuterium conversion.[11] From the latter experiment, it is also concluded that the deuton obeys Bose-Einstein statistics.

[1] Murphy and Johnston, Phys. Rev. **45**, 550 (1934).
[2] Dieke and Blue, Nature **133**, 611 (1934).
[3] Kapuscinski and Eymers, Proc. Roy. Soc. **A122**, 58 (1929).
[4] Childs and Mecke, Zeits. f. Physik **64**, 162 (1930).
[5] Dennison, Proc. Roy. Soc. **A115**, 483 (1927).
[6] Breit and Rabi, Phys. Rev. **38**, 2082 (1931).
[7] Sexl, Naturwiss. **22**, 205 (1934).
[8] Lewis and Ashley, Phys. Rev. **43**, 837 (1933).
[9] Heisenberg, Zeits. f. Physik **77**, 1 (1932).
[10] Raether, Naturwiss. **22**, 151 (1934).
[11] Farkas, Farkas and Harteck, Proc. Roy. Soc. **A144**, 481 (1934).

Apparatus and Procedure

The bands were photographed in the second order of a 21-foot concave grating in a Paschen mounting, giving a dispersion of about 1.3A per mm. A Jena glass filter, OG–1, was placed before the slit to remove overlapping orders. Eastman spectroscopic plates type III–F and IV–F were used since the bands lie in the orange region of the spectrum. The discharge tube, which was 60 cm long and 8 mm in diameter was of the usual H-type with 2 aluminum electrodes, the potential applied being about 4100 volts and the current 0.75 ampere. The latter was kept constant to within 1 percent during an exposure by manipulating an adjustable rheostat in series with the tube. Two large electric fans blew air across the tube during an exposure in order to keep it cool. Before use a solution of silver nitrate had been allowed to evaporate on the glass of the tube. The coating thus formed gave a catalytic surface for recombination of hydrogen atoms and the discharge showed a satisfactory "white stage" with most of the intensity in the molecular spectrum. The gas used was prepared by the electrolysis of water containing more than 90 percent deuterium.

Density marks were put in the center of each plate by means of a tungsten filament lamp burning with constant current of 0.85 ampere and a set of neutral wire screens.[12] These were prepared as described by Murphy and Urey[13] but had been recently recalibrated.[14] A system of slits placed directly in front of the photographic plate permitted a complete set of calibration marks for each band.[14] The density marks on the plates were 0.5 mm wide and in order to compensate for any developer effects a slit 0.5 mm wide was used for the deuterium spectrum. These plates are referred to as plates 19 and 20. Under these conditions some of the lines were not resolved but these were not used in the intensity measurements as explained below. Two exposures (plates 21 and 22) were also made using a narrower slit (0.05 mm) for the molecular spectrum. With the wide slit, the time of exposure for the density marks and the spectrum was 2 minutes each. With the narrow slit, the exposure time for the molecular lines had to be increased to 20 minutes.

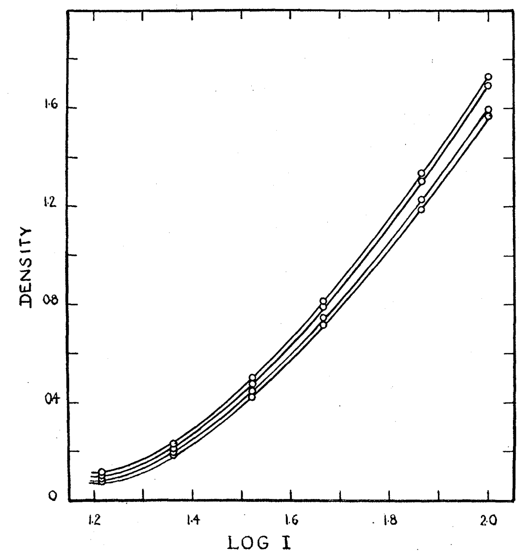

Fig. 1. Characteristic curves for plate 19.

The plates were carefully developed with Rodinal using all the necessary precautions for intensity measurements. Microphotometer curves were made with the Moll microphotometer and density curves plotted for each set of calibration marks. These curves were of the usual form and nearly parallel to each other as shown in Fig. 1. A typical microphotometer curve is given in Fig. 2. From the curves, log I for each line was determined by using a separate curve for each branch. The results for a typical plate are given in part in Table I.

Table I. *Logarithm of intensities of lines of the 2,2 band of deuterium.*

Branch	J	cm^{-1}	log I	Branch	J	cm^{-1}	log I
R	0	16,296.58	1.715	Q	4	16,232.64	1.892
	1	16,320.97	1.455		5	16,214.80	1.421
	2	16,341.73	1.718		6	16,193.59	1.483
	3	16,358.65	1.320	P	2	16,202.64	1.447
	4	16,371.71	1.432		3	16,164.94	1.373
	5	16,379.20	0.838		4	16,124.18	1.654
	6	16,384.58	1.006		5	16,080.43	1.359
Q	1	16,265.06	1.632		6	16,033.88	1.467
	2	16,257.80	1.997		7	15,984.70	0.952
	3	16,246.98	1.690		8	15,932.99	1.128

[12] Harrison, J.O.S.A. and R.S.I. **18**, 492 (1929).
[13] Murphy and Urey, Phys. Rev. **41**, 141 (1932).
[14] Joffe, Phys. Rev. **45**, 468 (1934).

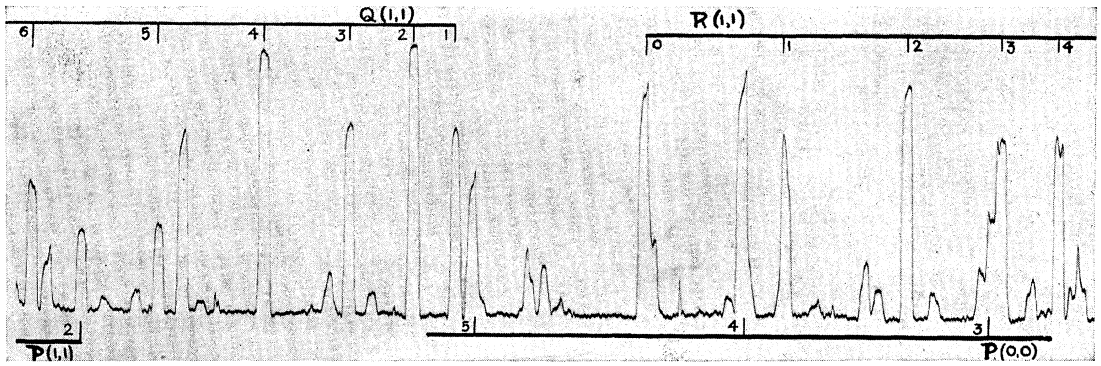

FIG. 2. Microphotometer curve showing portions of (0,0) and (1,1) bands from plate 19.

CALCULATION OF RESULTS

If the conditions of the discharge correspond to thermal equilibrium at an absolute temperature, T, the measured intensity of a line in emission is given by

$$I = Cgwe^{-E/kT}, \tag{1}$$

where E is the rotational energy of the molecule, w is the intensity factor and proportional to the statistical weight of the initial level neglecting any nuclear effects due to spin or statistics, k is Boltzmann's constant and g is the statistical weight due to nuclear spin. C is proportional to the fourth power of the wave number of the line and since the variation of the latter is small over the small range occupied by a band, C remains practically constant for each band.[15] The intensity factor, w has been computed by Hönl and London.[16] The hydrogen bands which were investigated represent a $^3\Pi \rightarrow {}^3\Sigma$ transition but since the spin separations in both states are so small, the bands may be treated as $^1\Pi \rightarrow {}^1\Sigma$ bands. Under these conditions the intensity factors are $J-1$, $2J+1$ and $J+2$ for P, Q and R branches, respectively, where J is the rotational quantum number for the initial level.

In the hydrogen molecule it is known that levels with even values of J are symmetric in the nuclei and those with odd J are antisymmetric.[17] The nuclear spin i is then given by

$$g_s/g_a = (i+1)/i \tag{2}$$

if the nucleus obeys Bose-Einstein statistics and

$$g_s/g_a = i/(i+1) \tag{3}$$

if it obeys Fermi-Dirac statistics.

If $E = BJ(J+1)$, it follows from Eq. (1) that

$$\ln (I/w) = \ln Cg - BJ(J+1)/kT \tag{4}$$

and if $\ln (I/w)$ is plotted against $J(J+1)$ for even and odd J, two parallel straight lines are obtained. The slope gives $-B/kT$ and the intercept on the $\ln (I/w)$ axis gives $\ln Cg$. This has been done for a typical case in Fig. 3. In this way we may determine g_s and g_a and hence the nuclear spin according to Eqs. (2) or (3).

The results for each branch have been plotted like Fig. 3 and in some cases it was found that certain lines were too intense and so did not fall on the parallel lines with the other points. In each case of this sort, it could be shown that these lines were blends due to the wide slit used and they were omitted in determining the nuclear spin as indicated in Table II. In obtaining

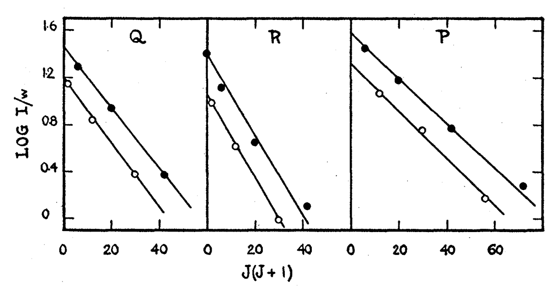

FIG. 3. Plot of log (I/w) against $J(J+1)$ for Q, P and R branches of the (2,2) band from plate 22. ○, odd values of J; ●, even values of J.

[15] Mulliken, Rev. Mod. Phys. **3**, 100 (1931).
[16] Hönl and London, Zeits. f. Physik **33**, 803 (1925).
[17] Heisenberg, Zeits. f. Physik **41**, 239 (1927); Hund, Zeits. f. Physik **42**, 93 (1927).

TABLE II. *Ratio of statistical weight factors calculated for various deuterium bands.*

Band	Branch	Plate 19 Lines used	Plate 19 g_s/g_a	Plate 20 Lines used	Plate 20 g_s/g_a	Plate 21 Lines used	Plate 21 g_s/g_a	Plate 22 Lines used	Plate 22 g_s/g_a
(0,0)	*R*	—	—	—	—	4	2.17	4	2.17
	Q	5	2.13	5	2.17	5	1.68	6	2.24
	P	4	1.93	—	—	4	2.47	4	1.61
(1,1)	*R*	4	1.85	5	1.73	4	2.27	5	2.16
	Q	5	1.90	5	1.91	6	2.13	6	1.81
	P	5	2.12	—	—	4	2.18	4	2.21
(2,2)	*R*	5	2.34	4	2.37	5	1.89	7	1.96
	Q	4	1.65	—	—	6	2.28	6	1.82
	P	5	2.01	4	1.72	6	1.51	7	1.57
(3,3)	*R*	4	1.52	4	1.71	6	1.98	6	2.29
	Q	4	1.93	5	1.90	6	2.22	6	1.89
	P	—	—	—	—	—	—	7	1.57
(4,4)	*R*	—	—	—	—	5	1.95	5	1.54
	Q	—	—	—	—	6	2.15	5	1.73
Average		1.94 ± 0.05		1.93 ± 0.06		2.07 ± 0.05		1.94 ± 0.05	

Weighted average of 4 plates $g_s/g_a = 1.97 \pm 0.03$.

actual values for g a least squares calculation has been made in each case. The ratio g_s/g_a for each band and branch is given in Table II.

As noted above, the slope of the parallel lines equals $-B/kT$. In this way one could determine the effective rotational temperature of the discharge tube. However, the branches are so short that considerable error is involved in obtaining the slope of the lines. A small error in the slope introduces a very large error in the temperature and it has been found impossible to determine the effective temperature from the experimental data.

Discussion of Results

There are several possible sources of systematic error. We have tried to reduce developer effects by careful brush development and in plates 19 and 20 by using spectral lines of the same width as our calibration marks. The plates were of very fine grain and since the lines were quite far above the background of the plate on the microphotometer curves the error due to grain size is negligible.

We have tried to account for variation in plate sensitivity with wave-length by using a separate set of density marks for each branch. Since we do not know the energy distribution of the lamp with wave-length, the lines in one branch are not directly comparable in intensity with lines in another branch, but since a branch extends only over about 100 cm^{-1}, we feel justified in assuming that the energy output of the lamp is constant over that region.

The error involved in measuring peaks of lines instead of areas may be considerable. Since we have used a very wide slit on plates 19 and 20, and a narrower slit on plates 21 and 22, any error due to this effect should be apparent. Since the variations in g in both these cases seems to be entirely random as may be seen from Table II, we believe this error to be small.

One further difficulty arises from the wide variation in intensity of the lines measured. For lines in a single branch, the strong lines may be as much as 35 percent more intense than the weaker lines. This means that the strong lines fall on the upper part of the density curves and the weak lines on the lower part, which fact introduces considerable inaccuracy. The ideal situation would require that all lines be near the middle or straight part of the density curves but since there is such a wide variation of intensities in the lines of a branch there is no simple way of fulfilling this requirement.

Conclusion

In spite of the fact that some systematic errors might be present the results from the analysis of 4 plates seem to indicate a random distribution of errors. Since the average of all the results gives $g_s/g_a = 1.97 \pm 0.03$ it immediately follows that the nuclear spin of deuterium is 1 and since the symmetric levels are more intense than the antisymmetric levels the nucleus obeys Bose-Einstein statistics. The value given for g_s/g_a is the weighted mean of the results for the 4 plates, and the deviation given is the probable error. With a nuclear spin of $\frac{1}{2}$ or $\frac{3}{2}$, the intensity alternation would be 3 : 1 or 1.66 : 1 and these figures are far outside of the deviation found even if the probable error had been much larger.

We take great pleasure in thanking Professor H. C. Urey for the heavy water used in these experiments. We also wish to thank Professor G. H. Dieke who provided us with the wave-lengths of the lines whose intensities we have measured and Dr. R. L. Garman of New York University for the microphotometer curves.

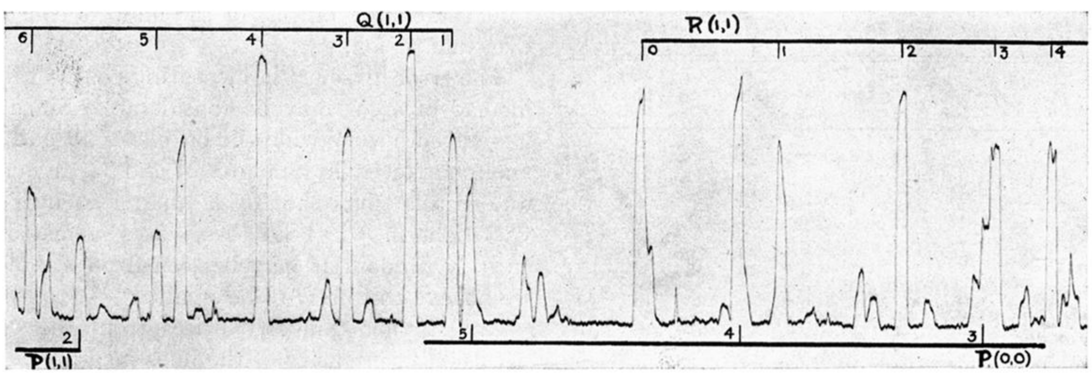

FIG. 2. Microphotometer curve showing portions of (0,0) and (1,1) bands from plate 19.