TABLE I. Microwave absorption spectrum of CH_2Br_2.

Frequency	Observed Intensity
25223 mc/sec.	*m*
25203	*m*
25170	*m*
25160	*w*
25152	*w*
25147	*w*
25128	*s*
25090	*m*
25072	*s*
25042	*s*
25013	*m*
25002	*w*
24982	*m*
24972	*w*
24943	*w*
24908	*m*

the observed lines are given in Table I. Previously a single line had been observed in this region by Turner,[1] who gave the frequency of this line as 25,056 mc/sec. We are extending our measurements to higher frequencies and are attempting an interpretation of the observed spectrum.

* The work described in this paper was carried out under contract between the Cambridge Field Station of the Air Materiel Command and the Ohio State University Research Foundation.

[1] T. E. Turner, Thesis, McGill University (1948).

Nuclear Spin and Quadrupole Moment of Cl^{36}*

C. H. TOWNES AND L. C. AAMODT
Pupin Physics Laboratories, Columbia University, New York, New York

July 18, 1949

EXAMINATION of the hyperfine structure of the $J=1\rightarrow 2$ rotational transition of the molecule $Cl^{36}C^{12}N^{14}$ allows determination of the Cl^{36} nuclear spin as 2 and its nuclear quadrupole moment as $-0.0172\pm 0.0004\times 10^{-24}$ cm^2.

The $J=1\rightarrow 2$ transition of $Cl^{36}CN$ occurs near 23635 mc/sec. The upper two curves of Fig. 1 show recordings of its hyperfine structure. The lower of these two curves represents the higher resolution, or the lower gas pressure. In the top curve resolution is somewhat poorer but random fluctuations are essentially eliminated. The other curves represent theoretical envelopes of the spectrum assuming different values of nuclear spin and resolutions approximately equal to that actually obtained. The upper curves for spin 2 and 3 and the curve for spin 1 correspond to line widths (total width at half-maximum) of 2 mc/sec. and the two lower curves for spins 2 and 3 are for line widths of 1 mc. Theoretical positions and relative intensities of each line is also shown. Magnitudes of quadrupole coupling constants for Cl^{36} are chosen for each value of spin to obtain the best fit of the experimental spectrum. The observed curve agrees very well with the theoretical curves assuming a spin of 2 but can be seen to disagree in a number of respects with those for spins of 1 and 3. Some small deviations in relative intensities of the components of the higher resolution experimental curve from those theoretically expected for a spin of two are caused by saturation effects, which depress the peaks of the stronger lines.

Measurements of relative intensities and separations of the three most evident components of the hyperfine pattern, labeled *a*, *b*, and *c* on Fig. 1, were made on approximately thirty recordings of this spectrum. Results are listed in Table I, which again shows very good agreement with theoretical expectations for a spin of 2. Intensity and spacing ratios for similar features of the theoretical spectrum assuming a spin of 1 are in clear disagreement with the results listed in Table I. Although the expected spectrum for a spin of 3 differs in general appearance from the observed spectrum, we have considered the outer bump on the right-hand side of the spin 3 spectrum as corresponding to com-

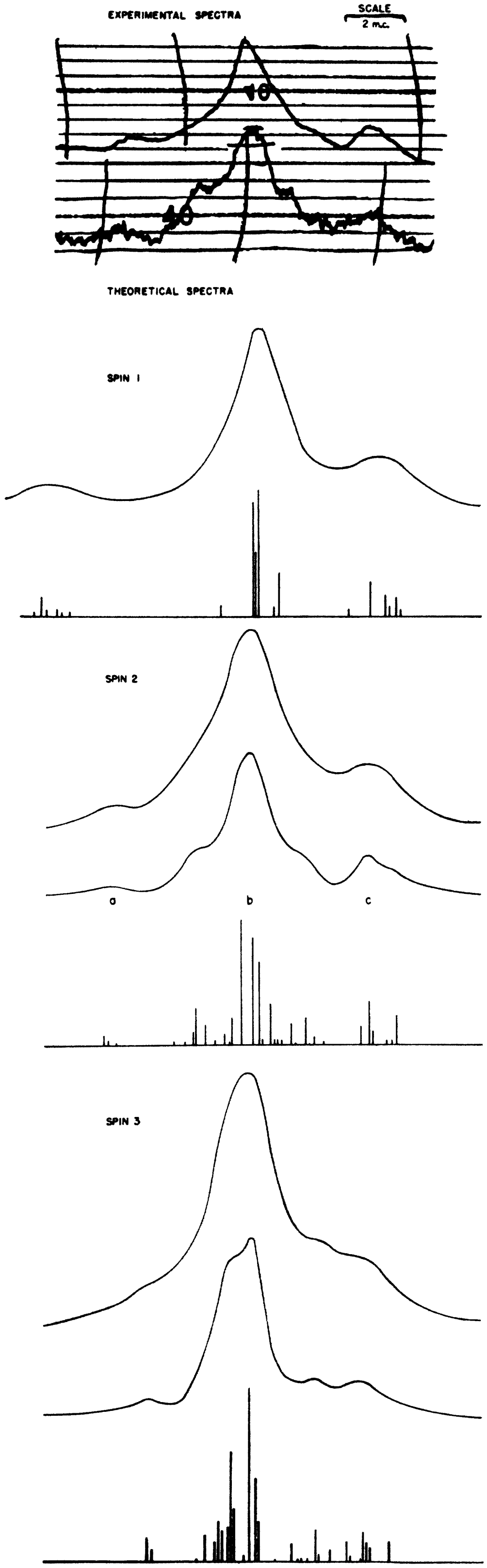

FIG. 1. Comparison of observed hyperfine structure of the $J=1\rightarrow 2$ transition of $Cl^{36}CN$ and theoretically expected structures for various assumed values of the Cl^{36} spin.

TABLE I. Intensity and spacing ratios. Designation of components as *a*, *b*, and *c* is shown in Fig. 1.

Intensity ratios	Measured	Theoretical for Spin 2	Theoretical for Spin 1	Theoretical for Spin 3
$\frac{a}{c}$	0.22 ± 0.03	0.20	0.43	0.50
$\frac{b}{c}$	4.05 ± 0.2	4.2	3.6	4.3
Spacing ratio				
$\frac{a-b}{b-c}$	1.17 ± 0.03	1.15	1.65	0.82

ponent *c* and the weak left-hand component to *a*, and given their theoretical relative intensities and positions for a comparison in Table I. Experimental measurements again clearly disagree with a spin of 3. Cl^{36} is, oddly, the first nucleus found to have a spin of 2.

N^{14} is present in the molecule ClCN and its quadrupole moment complicates the hyperfine pattern observed. However, the N^{14} quadrupole coupling constant in ClCN is accurately known[1] and its effect was included in the theoretical patterns. An exact calculation was made of energy levels and frequencies for the case of spin 2, although since the quadrupole coupling of Cl^{36} is five times larger than that of N^{14}, the exact frequencies differed only slightly from those obtained by a first-order calculation.[2] In the cases of spin 1 and 3, only a first-order calculation.[2] In the cases of spin 1 and 3, only a first-order calculation (assuming the N^{14} coupling small compared to the Cl^{37} coupling) has been made, since an exact computation would produce only minor changes in the theoretical curves.

The average frequency difference between the two weak components of the hyperfine pattern obtained from a number of spectral tracings is 9.08 mc/sec. This, combined with a knowledge of the Cl^{35} quadrupole moment[3] and its coupling constant[1,4] in the same molecule ClCN, allows a determination of the Cl^{36} nuclear quadrupole moment as $-0.0172\pm0.0004\ 10^{-24}\ cm^2$. This moment equals one-quarter of the average quadrupole moments of Cl^{35} and Cl^{37} within experimental accuracy. The ratio one-quarter would be expected if it is assumed that only the odd proton and neutron in *d* orbits[5] contribute to the nuclear moments and that Cl^{36} is in a $^3F_2$ state. This agreement may well be fortuitous, however, in view of the sizable quadrupole moment[6] of the neighboring odd neutron nucleus S^{33}.

The $Cl^{36}CN$ was prepared from a dilute solution of 70 mg of HCl furnished by the Oak Ridge National Laboratory. Isotopic abundance of Cl^{36} was approximately 0.8 percent. The HCl was titrated with NaOH and then evaporated to a volume of 5 cc. Two grams of MnO_2 were added, then concentrated H_2SO_4 introduced slowly and the evolved Cl_2 condensed in a dry ice bath. The mixture was heated to 80° to complete evolution of Cl_2. The resulting Cl_2 was mixed with an equal molal amount of HCN and a drop of water to obtain ClCN which, although not pure, was sufficiently concentrated to allow spectroscopic measurements.

The microwave spectrometer used was a Stark modulation instrument with a phase detecting amplifier and Esterline Angus recorder.[7]

The authors are indebted to Dr. B. P. Dailey for advice and Mr. A. F. Byas for assistance with chemical problems, and to Mrs. Zelda Droshnicop for the considerable amount of computation involved in obtaining the theoretical spectra.

* Work supported jointly by the Signal Corps and ONR.

[1] Townes, Holden, and Merritt, Phys. Rev. **74**, 1113 (1948).

[2] See J. Bardeen and C. H. Townes, Phys. Rev. **73**, 97 (1948).

[3] Davis, Feld, Zabel, and Zacharias, Phys. Rev. **73**, 525 (1948).

[4] Smith, Ring, Smith, and Gordy, Phys. Rev. **74**, 370 (1948).

[5] E. Feenberg, Phys. Rev. **75**, 320 (1949). L. Nordheim, to be submitted for publication in Phys. Rev.

[6] C. H. Townes and S. Geschwind, Phys. Rev. **74**, 626 (1948).

[7] W. Low and C. H. Townes, Phys. Rev. **75**, 529 (1949).

Radioactivity of Chlorine^{36}*

C. S. WU, C. H. TOWNES, AND L. FELDMAN
Pupin Physics Laboratories, Columbia University, New York, New York

July 18, 1949

THE half-life of the Cl^{36} activity has been estimated as $\sim 10^6$ yr.[1] and 2×10^6 yr.[3]. The maximum energy of the beta-radiation from absorption measurements was given as 0.64[2] to 0.66[1] Mev. Cl^{36} has also been reported to decay in the three parallel paths[2] of β^-, β^+ and *K*-capture. However, recent investigation[4] of the maximum positron to negatron ratio in the decay of Cl^{36} by gamma-ray absorption method puts an upper limit of the ratio of positron-to-negatron-emission at less than 3×10^{-4}. No nuclear gamma-rays have been reported. We have redetermined the half-life of Cl^{36} as $(0.44\pm0.05)\times10^6$ yr. by determining the specific disintegration rate of Cl^{36}. In addition, a search for positron and gamma-ray emission has been made with negative results.

The isotopic concentration of Cl^{36} was obtained from intensity measurements of the microwave absorption spectrum of $Cl^{36}CN$. Part of a dilute solution of HCl containing Cl^{36} was converted to $Cl^{36}CN$ as described in the preceding letter. A number of spectra of the $J=1\rightarrow2$ transitions of $Cl^{36}C^{12}N^{14}$ and of $Cl^{35}C^{13}N^{14}$ were taken on a recording spectrometer and the ratio of total area (integrated intensity) of the absorption lines due to the two transitions was measured from the recording. A variety of conditions of gas pressure and spectrometer adjustments were used, but for all intensity measurements the pressure was high enough so that hyperfine components of the $Cl^{36}C^{12}N^{14}$ line were not resolved, and only certain well-separated hyperfine components of the $Cl^{35}C^{13}N^{14}$ transition were resolved. Intensities and frequencies of the two absorption lines compared differed only slightly. Assuming the natural abundance of Cl^{35} to be 75.40 percent and C^{13} to be 1.12 percent, the abundance of Cl^{36} obtained for two different batches of HCl was 0.675 percent and 0.835 percent as shown in Table I. Accuracy of these determinations is thought to be ±5 percent.

The absolute beta-disintegration rate was measured on a mica window (5–6 mg/cm^2) G-M counter. The geometrical efficiency of the counter was calibrated with the help of several RaE standards from the Bureau of Standards. The procedure of preparing source and calculating the exact Cl^{36} content in the source is as follows. A careful titration of the HCl solution received from Oak Ridge with an NaOH solution of known normality gives the exact amount of chlorine contained in the solution. The neutralized solution is then slowly evaporated to dryness under an infra-red lamp. The total solid content of this neutralized solution is weighed on a microbalance. After weighing, the total solid is redissolved in a minimum amount of distilled water and several cc of pure alcohol or acetone are added to obtain a suspension. The particles in suspension are allowed to settle on a 1.24 cm^2 area of a standard silver disk which is the same as the RaE sources of the Bureau of Standards. After these sources are thoroughly dried, the weight is again determined on a microbalance. The Cl^{36} content in the source is then easily calculated.

The different amounts of backscatterings of the beta-radiations of Cl^{36} and RaE from the standard disks are determined experi-

TABLE I. Half-life determinations of Cl^{36}.

Group	Sample	Isotopic concentration	Sample weight	Half-life
1	*a*	0.675	0.836 mg	0.428×10^6 yr.
1	*b*	0.675	0.536 mg	0.394×10^6 yr.
2	*c*	0.835	0.431 mg	0.480×10^6 yr.
2	*d*	0.835	0.262 mg	0.494×10^6 yr.
2	*e*	0.835	2.236 mg	0.425×10^6 yr.
				Average $=0.444\times10^6$ yr.