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The Zeeman Effect in Ammonia Microwave Spectra

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Synopsis

The microwave Zeeman effect of $N^{14}H_3$ inversion spectra was investigated at strong magnetic field to observe molecular g-factor in the decoupled state of spin-rotation interaction. The result on $J, K = 1,1$ line shows the transition from Zeeman effect to Paschen-Back effect near 10,000 oersteds. The obtained molecular g-factors for several different JK value give some information on g-factor of $N^{14}H_3$ both parallel and perpendicular to the molecular axis.

I. Introduction and Experimental Procedure

The microwave Zeeman effect in the inversion spectra of $N^{14}H_3$ has been investigated by several workers(1),(2),(3),(4),(5) at relatively low magnetic fields of several thousand oersteds. But at such fields the decoupling of the spin-rotation interaction is unsatisfactory, as suggested by C. K. Jen(3).

We attempted to study the Paschen-Back effect of $N^{14}H_3$ at a strong magnetic field in the complete decoupling state, in which the molecular g-factor of $N^{14}H_3$ should be observed, unaffected by nuclear g-factor of N^{14}.

In order to investigate the Zeeman effect of microwave spectrum at a strong magnetic field, a 10-ton electromagnet having ring-shaped pole pieces was constructed to be used in the present work. And for the absorption cell, a K-band waveguide of about 2 meter length was circularly bended at a diameter of 70 cm and placed between pole pieces described above in such a way that the narrow side of waveguide wall would be perpendicular to the static magnetic field. Thus the magnetic field up to 15,000 Oe, perpendicular to the r-f electric field, was easily applied to the full length of 2-meter absorption cell when the width of pole face was 4 cm and the gap between two pole pieces was 1.55 cm. The uniformity of magnetic field was within 0.4 percent along the longitudinal axis of the absorption cell.

The common sweep method was employed to observe the spectral line of $N^{14}H_3$;

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(1) D.K. Coles and W.E. Good, Phys. Rev., **70** (1946), 979.

(2) C.K. Jen, Phys. Rev., **74** (1948), 1396.

(3) C.K. Jen, Phys. Rev., **76** (1949), 1494.

(4) C.K. Jen, Phys. Rev., **81** (1951), 197.

(5) J.R. Eshbach and M.W.P. Strandberg, Phys. Rev., **85** (1952). 24.

the third harmonics from the output of a type 723 A/B klystron was generated by a crystal multiplier as the source of radiation.

In order to measure the frequency differences between separated Zeeman components, two methods were employed. The frequency modulation of the klystron source by a 5 Mc/sec quartz crystal oscillator produced images of the spectral lines spaced by the modulating frequency, namely, at the intervals of 5 Mc/sec, so the frequency differences between the separated Zeeman components could be measured from these 5 Mc/sec scales in oscilloscope photograph.

A variable frequency oscillator was used for another method, in which one could vary the modulating frequency so as to make images from adjacent lines coincide with one another, and could read the frequency differences directly from the frequency of oscillator calibrated by means of the standard frequency of 4Mc/sec of JJY. The results by both methods were in agreement within experimental errors. Whole setups of experimental apparatus are shown in Photo. 1.

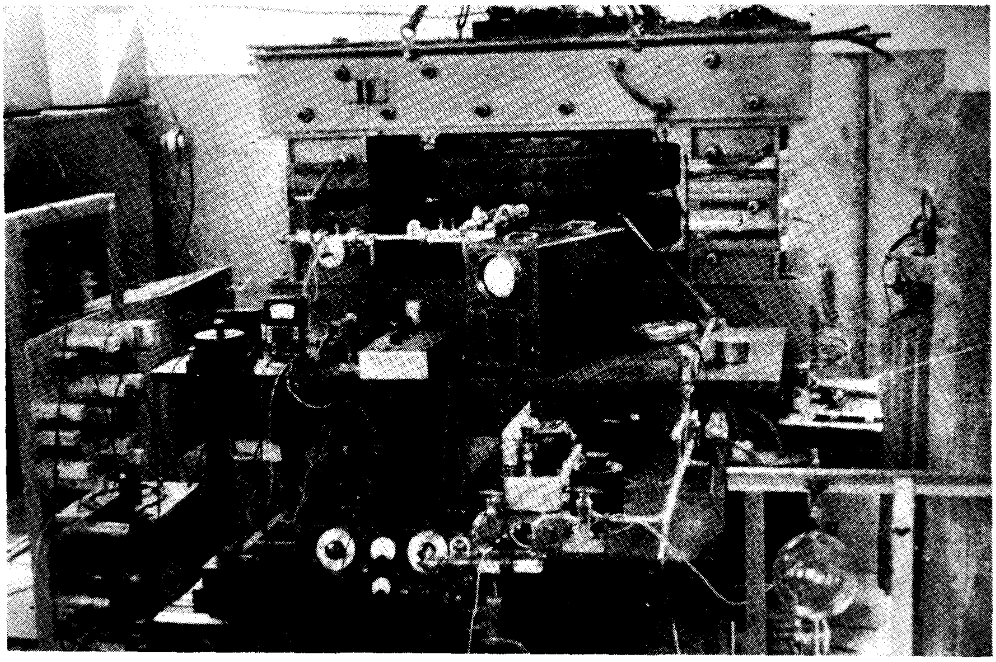

Photo. 1. The microwave spectroscope for measuring Zeeman and Paschen-Back effects.

The pressure of $N^{14}H_3$ gas in the absorption cell was kept at about 10^{-2}mmHg during the experiment in order to make observations under suitable conditions.

Theoretical Background and Experimental Results

The molecule in the $^1\Sigma$ ground state has very small magnetic moment due to molecular rotation. It is possible to detect small frequency splitting with microwave Zeeman effect as follows:

$$\Delta\nu = g(\mu_0/h)H \qquad (1)$$

where $\Delta\nu$ = difference between the frequency of a Zeeman component and that of the undisplaced line, μ_0 = nuclear magneton, g = gyromagnetic ratio.

On the other hand, nuclei have magnetic moment which is often strongly coupled with molecular axis.

In $N^{14}H_3$ there exists an interaction between nucleus and molecule having a molecular axis. In the absence of an external field spin-rotation coupling is still in effect.

The Zeeman effect on the resulting hyperfine pattern has been treated theoretically by Jen(2). When the the Zeeman energy is smaller than the spin rotation coupling, the unperturbed state is labeled by J, I, F, M, where J = angular momentum quantum number, $F=J+I$, $J+I-1$, ..., $J-I+1$, $J-I$ and $M=F$, $F-1$, ... ,$-F+1$, $-F$. The total hamiltonian $\mathcal{H}$, including the interaction with an external magnetic field, of a molecule having a molecular g-factor and single nucleus coupled with the molecular axis, is

$$\mathcal{H} = \mathcal{H}_0 - g_{mole}\,\mu_0\,\boldsymbol{J}\cdot\boldsymbol{H} - g_{nuc}\,\mu_0\,\boldsymbol{I}\cdot\boldsymbol{H}, \tag{2}$$

where g_{mole} = g-factor of the molecule along J, g_{nuc} = g-factor of the nucleus coupled with molecule, $\boldsymbol{J}, \boldsymbol{I}$ = vector operators of J, I.

The interaction energy stated by Jen is

$$\Delta W = -M\mu_0 H(\alpha_J g_{mole} + \alpha_I g_{nuc}), \tag{3}$$

where

$$\alpha_J = [F(F+1) + J(J+1) - I(I+1)]/2F(F+1),$$
$$\alpha_I = [F(F+1) + I(I+1) - J(J+1)]/2F(F+1).$$

For the Zeeman splitting of $N^{14}H_3$ inversion spectrum, splitting of Zeeman component takes the following form for the central line,

$$\Delta\nu_0 = \pm\frac{\mu_0 H}{h}\left\{\frac{g_{mole}+g_{nuc}}{2} + \frac{J(J+1)-I(I+1)}{F(F+1)}\,\frac{(g_{mole}-g_{nuc})}{2}\right\}, \tag{4}$$

where selection rules for σ transition of $N^{14}H_3$ inversion spectrum are

$$\Delta J = 0, \quad \Delta M = \pm 1, \quad \Delta F = 0.$$

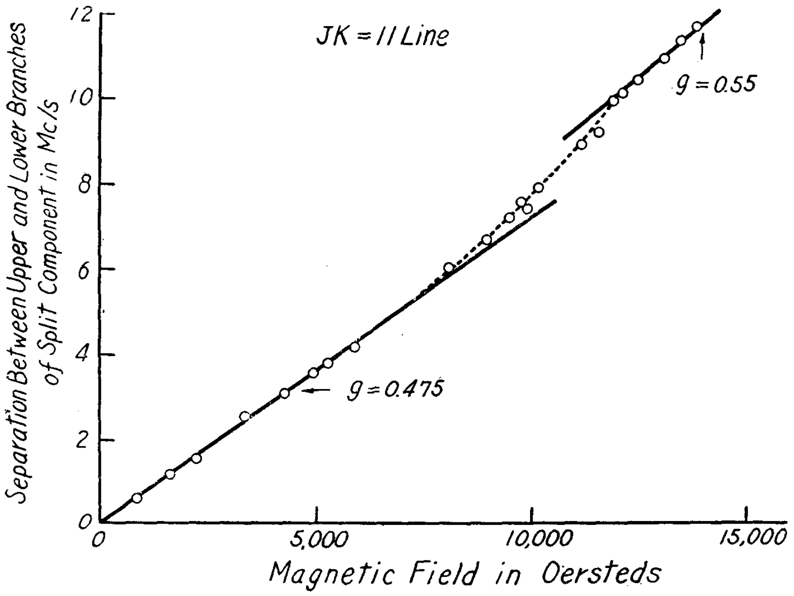

Fig. 1. Zeeman and Paschen-Back splitting for $JK=11$ line of $N^{14}H_3$ as a function of the magnetic field.

When the magnetic field is so strong that spin-rotation coupling breaks down, the splitting of the Zeeman effect changes to Paschen-Back effect.

Paschen-Back effect is observed by Jen(3) in $N^{14}N^{14}O$ but his result in $N^{14}H_3$ is not complete.

Fig. 1 shows the Zeeman effect of $N^{14}H_3$ with assigned rotational quantum number $J = 1$, $K = 1$. It is seen that a linear relation is maintained up to 8,000 oersteds which corresponds to g-factor being equal to 0.475. For still higher magnetic field, there is a slight non-linearity and gradual approach to a new straight line, of which the slope corresponds to g-factor of 0.550. The experimental errors are limited to the range of 1 percent which arise mainly from magnetic field measurement. The change in g-factor shows the transition from the Zeeman effect in which molecular g-factor was combined with nuclear g-factor, to Paschen-Back effect in the decoupled state.

The splitting of Paschen-Back effect in $N^{14}H_3$ is given as follows:

$$\Delta\nu = \pm\frac{2\mu_0 H}{h} g_{mole}. \tag{5}$$

That is, splitting is proportional to molecular g-factor. We obtained from the present results using equations (4) and (5),

$$g_{mole} = 0.550 \pm 0.005,$$
$$g_{nuc} = 0.400 \pm 0.004.$$

The value of nuclear g-factor of N^{14} determined from present study coincides with published nuclear resonance data, $g_{muc} = 0.403$ within the range of experimental errors.

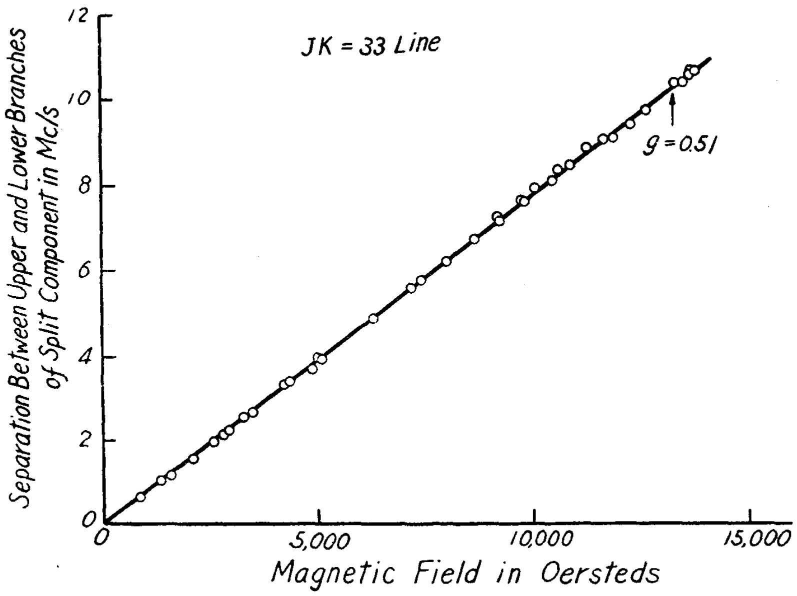

Fig. 2. Zeeman splitting for $JK = 33$ line of $N^{14}H_3$ as a function of the magnetic field.

Fig. 2 gives experimental results of 3-3 line in $N^{14}H_3$. The separation of the Zeeman component is linearly proportional to the applied magnetic field up to 14,000 oersteds. In 3-3 line we could not observe Paschen-Back effect explicitly, but from the separation at 14,000 Oe, we could estimate $g_{mole} = 0.510$ in 3–3 line. In the range of weak field, if we substitute $g_{mole} = 0.510$ and $g_{nuc} = 0.403$ in equation (4), g-factor at weak field is shown to consist of three factors as follows:

$$\begin{aligned} g &= 0.483 \quad \text{for} \quad F = 4, \\ g &= 0.501 \quad \text{for} \quad F = 3, \\ g &= 0.546 \quad \text{for} \quad F = 2. \end{aligned}$$

The observed g-factor should be some intermediate value of the above three, according to the transition probability of each component of fine structure. Therefore, the coincidence of the observed g-factor in 3–3 line at low and high field seems to be reasonable.

In 2–2 and 6–6 line of $N^{14}H_3$, the same field dependence but with a slope slight different from that in 3–3 line was observed.

Table 1. Molecular g-factors of $N^{14}H_3$ deduced from σ component Zeeman effect measurements at 14,000 oersteds.

J, K	1,1	2,2	3,3	6,6
$g(J, K)$	0.55	0.53	0.51	0.50

Table 1 shows rotational g-factor of $N^{14}H_3$ determined at 14,000 Oe for different JK value.

In the nonlinear molecule, molecular g-factor is a function of J and K. For the symmetric molecule, T. R. Eshbach and M. W. P. Strandberg have found the following equation:

$$g(J,K) = g_N + (g_K - g_N)\frac{K^2}{J(J+1)}, \tag{6}$$

$g_K g_N$ defines the component of magnetic moment $g_K \mu_0 K$ along the symmetry axis and the component of $g_N \mu_0 N$ (where $N = \sqrt{J(J+1) - K^2}$) perpendicular to this axis.

From our data, the extrapolated g_N and g_K are

$$\begin{aligned} g_N &= 0.62 \pm 0.02, \\ g_K &= 0.48 \pm 0.01. \end{aligned}$$

These results can be compared with Eshbach and Strandberg's in $N^{15}H_3$.

Acknowledgement

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