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By

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1. Introduction

In U. S. A., the field of microwave spectroscopy has grown with tremendous rapidity in post war years, but in our country, the researches in this field have been scarce so far.

In compliance with the physical importance of this field, we have designed and constructed the 1.5 cm wave-length spectroscope, whose general view is given in Fig. 1. With this set, we observed the absorption line of Methyl Bromide (CH_3Br). Our observed wave-length of maximum absorption coincides with the theoretical value within the

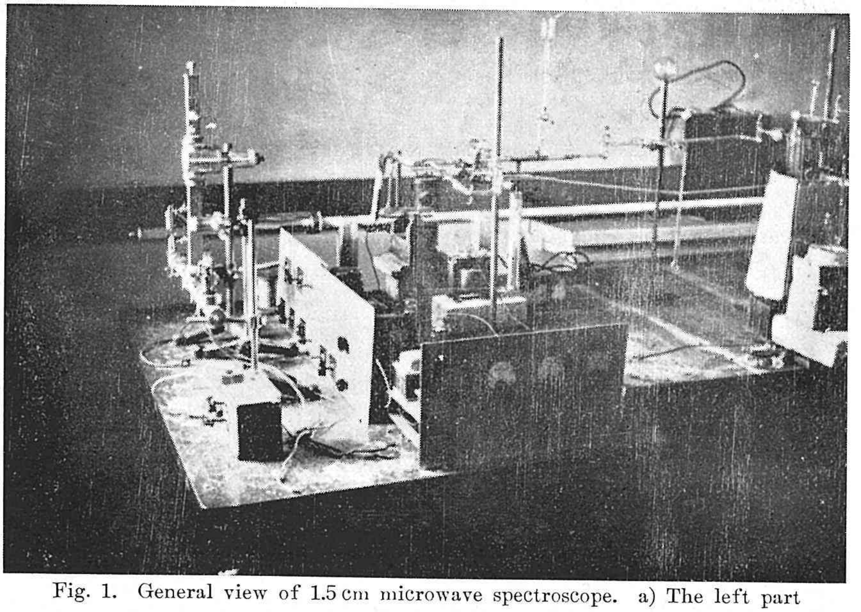

Fig. 1. General view of 1.5 cm microwave spectroscope. a) The left part

* Read on May 1, 1950, at the meeting of Microwave Physics Branch of the Physical Society of Japan.

error of our cavity wave meter and we confirmed the hyperfine structure of spectral line due to the presence of the quadrupole moment of Br nucleus.

Several accurate measurements about Methyl Bromide have already been reported by others (**11, 12, 13, 14, 15**). Methyl Bromide was used in our measurement too, since it is appropriate for checking our set and in our country no experimental report about centimeter wave spectrum has hitherto been given.**

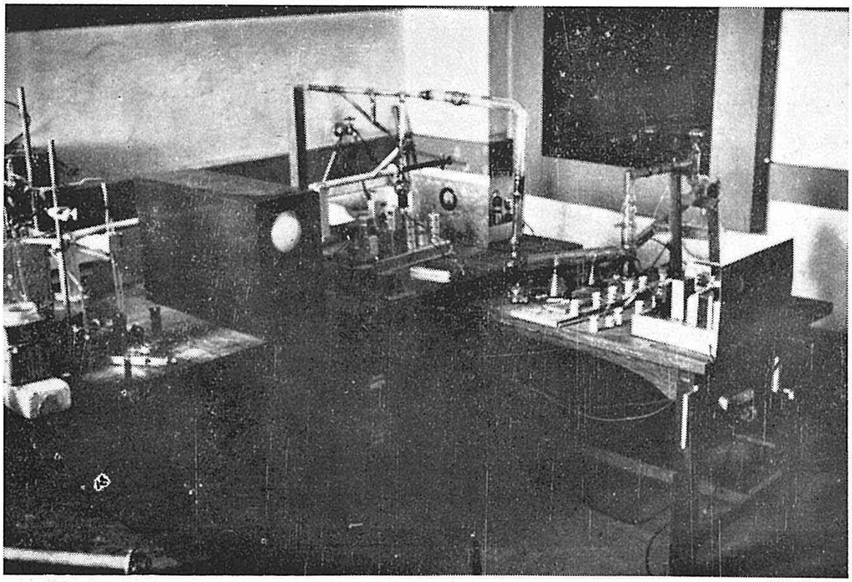

Fig. 1. General view of 1.5 cm microwave spectroscope. b) The right part

2. Description of the set and its components

The block diagram of our microwave spectroscope is shown in Fig. 2. A part of 3 cm microwave energy from the signal oscillator, passing through T-junction and attenuator A, is converted to 1.5 cm energy by the frequency converter. This converted energy is then led into the absorption cell formed by a circular cylinder and transparent mica windows at its both ends.

The original 3 cm energy is frequency-modulated by the saw-tooth generator and consequently the 1.5 cm is so also. Thus, if the frequency of the converted energy is tuned to the absorption frequency of

** We are aware of only Prof. Kojima's spectroscopic researches with 12 cm wave-length microwave which are carried on at Tokyo Educational University. He also has read his paper on May 1, 1950.

the sample in the cell, the energy going out from the cell is much damped.

The output energy amplitude-modulated corresponding to the degree of absorption of the sample in the cell is detected by the crystal detector and the detected power which represents the absorption character of the sample is amplified and applied to the vertical axis of the oscillograph whose horizontal axis is swept by the voltage of the saw-tooth generator. Thus, the absorption character of the sample is visualized on the screen of the oscillograph.

In our set, the amplification of detected power is carried out in two ways. One way is the direct amplification by the video amplifier, and the other consists in modifying the signal generator with 463 kc/sec source modulator in addition to the saw-tooth generator and amplifying and detecting by means of 463 kc/sec narrow band receiver.

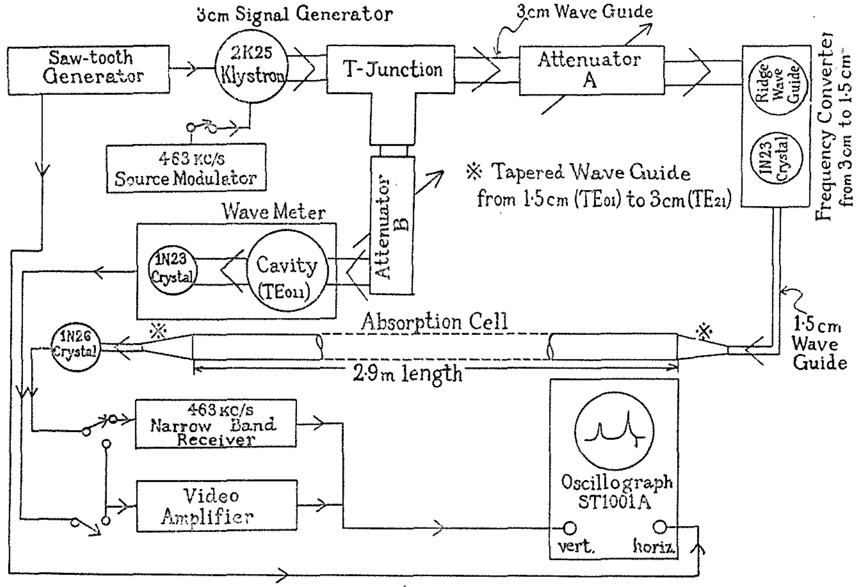

Fig. 2. Block diagram of 1.5 cm microwave spectroscope.

In the following, the main components of the set are described.
(1) *Signal Generator*: The well-known klystron tube 2K25 is used and its frequency range is from 8.5 to 9.8 kMc/sec.
(2) *Frequency Converter*: As shown in Fig. 3, it is composed of ridge type wave guide, coaxial line and silicon crystal 1N23. To design the ridge type wave guide, some papers (**2, 3**) were referred to, and this is the matching device between 50 ohm coaxial line and 3 cm wave guide.

To check its behavior, we measured the voltage standing wave ratio (V. S. W. R.) in 3 cm wave guide when the ridge type wave guide is inserted between 3 cm wave guide and 50 ohm coaxial line, and reasonable results were obtained as shown in Table I below.

TABLE I. Some results concerning ridge type wave guide.

Wave meter scale "L"	Converter crystal current	V.S.W.R. in 3 cm wave guide	Impedance calculated from V.S.W.R. "Z"
23.68 mm	2.50 mA	1.335	698 Ω
23.93	3.50	1.284	671
24.25	5.12	1.333	696
24.62	4.12	1.285	671

The characteristic impedance of 3 cm wave guide $z_0 = 523\ \Omega$

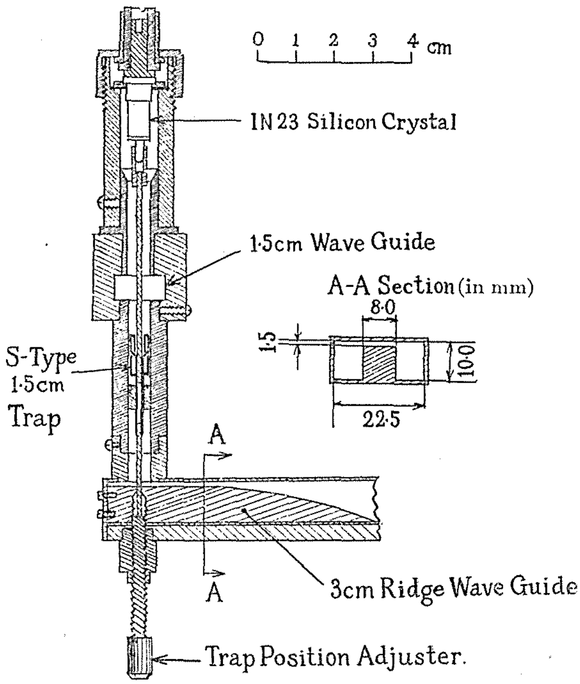

Fig. 3. Schematic sketch of frequency converter.

(3) *Attenuator*: Vane type variable attenuator is used. The vane is a thin bakelite plate coated with the mixture of graphite powder and binder consisting of polystyrene and benzol solvent. Maximum attenuation is about 20 db.

(4) *Wave meter*: A transmission type cavity wave meter utilizing

TE_{011} mode is used in 8.5—9.8 kMc/sec range. To suppress TM_{111} mode, behind its piston is inserted the link of thin nickel plate which is used because of unavailableness of polyiron. This wave meter is self-calibrating and can afford the frequency measurement to the accuracy of $\pm$ 5 Mc/sec.

The output from the crystal detector attached to the cavity is led through the video amplifier and used as a frequency marker pip on the oscillograph screen.

(5) *Crystals*: 1 N 23 silicon crystals are used for converter and 1 N 26 for detection of 1.5 cm wave. We remove 1 N 26 from the wave guide for its safety except in case of experiment being carried on.

(6) *Absoption cell*: The absorption cell is constructed from brass circular wave guide of 2.9 m length, 2.29 cm inner diameter and 2.59 cm outer diameter. Of course, we know that the rectangular wave guide is better for our purpose, but since it is not available for us under the present circumstances, we have employed the following procedure: The section of 1.5 cm wave rectangular guide is gradually changed from rectangle of 13.0 mm $\times$ 6.5 mm to the circle of 22.9 mm diameter through elliptic section tapered along 22 cm length. The surface finishing of this tapered part is most carefully done and so expected symmetry and inconsiderable reflection is attained.

The mica window of about 1/20 mm thickness is fixed at each end to seal the cell by means of red beeswax and brass ring to clamp and some cares are taken to minimize the reflection from these junction points. But we cannot yet succeed to suppress completely such reflection, as some ghosts have appeared which, however, can be clearly distinguished from absorption lines.

We suppose that the mode in the cell is mainly TE_{21} but some other modes can be anticipated to occur at the same time, which we are now planning to suppress.

(7) *Saw-tooth generator*: The saw-tooth generator of frequency range from 1.5 c/sec to 150 c/sec is used. Its circuit is similar to that of A. H. Sharbaugh (5). Low sweep rates serve to better resolution and to more accurate production of the absorption line shape. 50 volt sweep voltage applied to the reflector electrode of klystron suffices to sweep the 100 Mc/sec band width at 1.5 cm wave.

The sweep frequency is 60 c/sec when the video amplifier is used, and 2 c/sec when the narrow band receiver is.

(8) *Narrow band receiver*: A handmade single band receiver with crystal filter is used. The selection of frequency 463 kc/sec is made for

convenience only. The gain is about 140 db, the band width can be changed from 500 c/sec to 10 kc/sec on five steps, and for input stage, low noise tube 6AC7 is used.
(9) *Video amplifier*: The gain is about 100 db. The frequency response is sufficient for our purpose.
(10) *Oscillograph*: The type ST1001A of Matsuda Mfg. Co. is used, some additional measures to adapt for our set being taken.
(11) *Vacuum system*: The block diagram of vacuum system is shown in Fig. 4. Phillips gauge was used for measuring pressure of the gas, since it has fairly good sensitivity and moreover is not spoiled by gases. Its measuring limit is about 10^{-5} mmHg.

In order to obtain greater accuracy at lower pressure, however, a more sensitive ionization gauge with L. N. Ridenour stabilization circuit (6) was used. We have thus obtained the minimum pressure as low as 10^{-6} mmHg in wave guide cell.

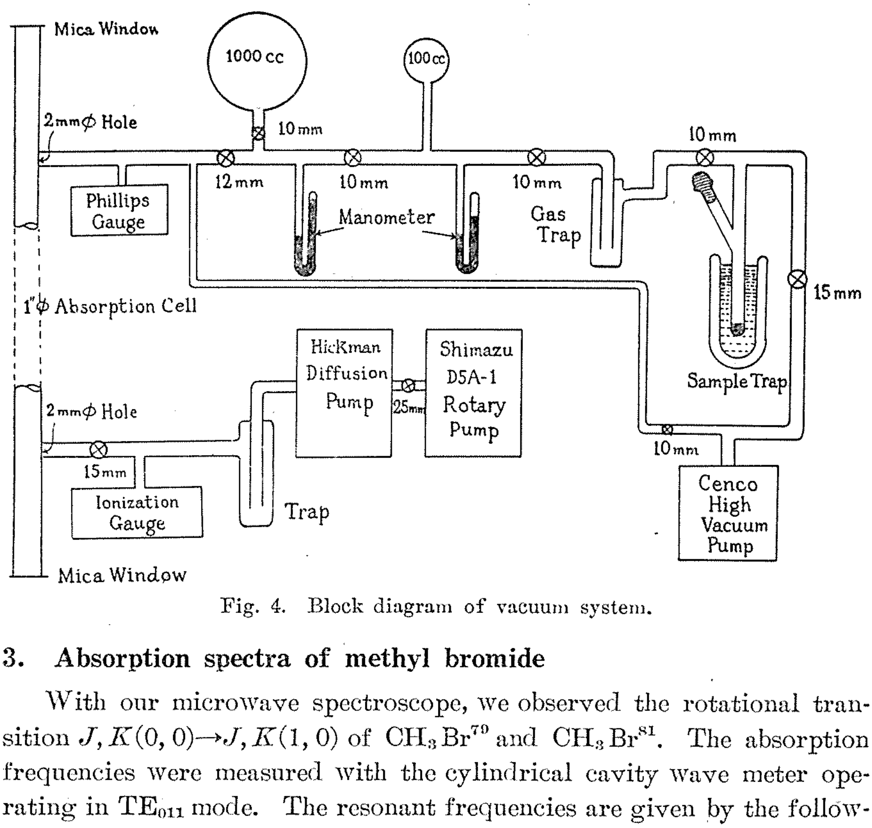

Fig. 4. Block diagram of vacuum system.

3. Absorption spectra of methyl bromide

With our microwave spectroscope, we observed the rotational transition $J, K(0, 0) \rightarrow J, K(1, 0)$ of CH_3Br^{79} and CH_3Br^{81}. The absorption frequencies were measured with the cylindrical cavity wave meter operating in TE_{011} mode. The resonant frequencies are given by the following formula: $f = (c/2)[(7.6634/\pi D)^2 + (1/L)^2]^{\frac{1}{2}}$, where c is the light velocity, and D and L are the diameter and length of the cylinder respectively.

Our measuring system can afford the accuracy of ± 5 Mc/sec at 3 cm wave. The satellite structure of rotational line of CH_3Br is caused by the interaction of nuclear quadrupole moment of Br with molecular field. The theoretical formula for the interaction (**7, 8, 9, 10**) is

$$E_0 = eQ\frac{\partial^2 V}{\partial z^2}\left(\frac{3K^2}{J(J+1)}-1\right)\frac{\frac{3}{4}C(C+1)-I(I+1)J(J+1)}{2(2J+3)(2J-1)I(2I-1)},$$

with

$$C = F(F+1) - I(I+1) - J(J+1),$$

$$F = J+I,\, J+I-1,\, \cdots\cdots,\, |J-I|,$$

where I is the spin of the interacting nucleus, e the electronic charge, Q the nuclear quadrupole moment, and $\partial^2 V/\partial z^2$ is the divergence along the molecular axis of the molecular field at the interacting nucleus.

For the case of CH_3Br, we performed calculations, by putting the following values (**15**)

$$eQ\,\partial^2 V/\partial z^2 = 577.3 \text{ Mc/sec for } CH_3Br^{79},$$

$$= 482.4 \text{ Mc/sec for } CH_3Br^{81},$$

$$I = 3/2 \text{ for both } Br^{79} \text{ and } Br^{81},$$

and using selection rules

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

Our observed values and calculated values are summarized in Table II.

TABLE II. Absorption frequencies of CH_3Br.

	Transition	Frequency (obs.)	Frequency(calc.)
CH_3Br^{79} $\nu_0 = 19197$ Mc/sec	$F=3/2 \rightarrow 1/2$	1899_1 Mc/sec	18992 Mc/sec
	$3/2 \rightarrow 3\ 2$	1926_2	19252
	$3/2 \rightarrow 5/2$	1910_8	19108
CH_3Br^{81} $\nu_0 = 19064$ Mc/sec	$3/2 \rightarrow 1/2$	1896_0	18943
	$3/2 \rightarrow 3/2$	1917_0	19160
	$3/2 \rightarrow 5/2$	1906_8	19040

(ν_0: frequency of the unsplit rotational line)

The calculated and observed values agree well with each other within the error of our present frequency measuring system, whose accuracy is ± 10 Mc/sec at 1.5 cm wave.

Several accurate measurements for CH_3Br have been already reported (**11, 12, 13, 14, 15**). Among them, however, 1.5 cm wave-length spectrum was observed only by A. H. Sharbaugh and J. Mattern (**15**).

Two absorption lines accompanied by one ghost are shown in Fig. 5.

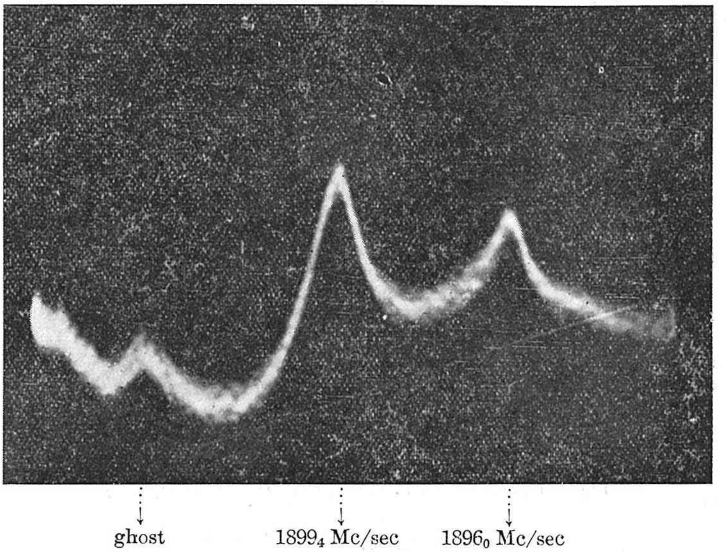

Fig. 5. The absorption lines of CH_3Br.

The intensity comparison among absorption lines was not made this time, because it was difficult to adjust all circuit components for each line into the same conditions, especially so for the lines lying in the different electronic tuning ranges of klystron, and further the output power of klystron could not be kept constant without any necessary procedure over the range of electronic tuning.

The more accurate frequency measuring system utilizing harmonics of 100 kc/sec crystal controlled oscillator which makes zero beat with the standard wave of the Standard Frequency Station JJY is now under construction by us.

4. Acknowledgements

The authors express their heartfelt thanks to Prof. R. Nozu, Dr. R. Goto, and Mr. H. Shakutsui for the preparation of the sample, and we owe deep thanks to Dr. O. Cary who helped us to get klystron and silicon crystals. Also, much credit is due to Messrs. H. Narumi and H. Takeyama and several members both in Arakatsu Laboratory and in our Laboratory. We thank further to Messrs. D. Nakayama and H. Iwata for their cooperation in constructing wave meter and other components used in our research.

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