We note that the last term on the right is a divergence, but the others are not unless

$$G^{B\alpha}{}_{A\iota}=F^{B\alpha}{}_{A\iota}. \tag{5.5}$$

In that case, we can form the sixteen functions

$$T^{\rho}{}_{\iota}=t^{\rho}{}_{\iota}+F^{B\rho}{}_{A\iota}L^{A}y_{B}, \tag{5.6}$$

whose divergence is identically zero whether the L^A vanish or are equal to P_A. This expression is identical with (I-3.11).

The author would like to express his appreciation to Dr. Peter G. Bergmann for the suggestion of and the helpful discussions in connection with this problem.

PHYSICAL REVIEW VOLUME 81, NUMBER 6 MARCH 15, 1951

The Mass of S^{35} from Microwave Spectroscopy*

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(Received November 30, 1950)

The $J=1\rightarrow2$ rotational absorption transition in OCS has been observed for the molecules containing S^{35} and S^{34}. From the frequencies, the frequency differences, and the previously known frequencies of S^{34}, S^{33}, and S^{32} we have evaluated the mass difference ratios $(S^{35}-S^{32})/(S^{34}-S^{32})$ and $(S^{35}-S^{32})/(S^{33}-S^{32})$. From these values and values of the stable S masses two independent values of the $(S^{35}-S^{32})$ mass differences are calculated to be 2.99844±0.00042 and 2.99770±0.00048, respectively.

I. INTRODUCTION

WE have remeasured[1] the frequencies of the $J=1\rightarrow2$ molecular rotational absorption transitions of OCS containing S^{32}, S^{33}, S^{34}, and S^{35} for the purpose of determining the mass of S^{35} and of evaluating the nuclear quadrupole interaction to a higher accuracy.

II. METHOD

Our apparatus, somewhat similar to various other K-band spectroscopes described in the literature,[2,3] is illustrated schematically in Fig. 1. It utilizes 100-kc/sec Stark effect modulation and for maximum sensitivity a phase-sensitive detector at the output. Our frequencies were measured by means of variable microwave frequency markers obtained from a frequency standard somewhat similar to those used in other laboratories.[4] Figure 2 shows a schematic arrangement of the system. The basis of our measurements is a General Radio 100-kc/sec crystal-controlled secondary frequency standard calibrated against Radio Station WWV.

As shown below in Eq. (3) the mass of S^{35} can be expressed in terms of frequency differences and ratios. In determining such differences small systematic errors, such as are caused by delays in the spectroscope amplifier, tend to cancel out of differences and ratios of nearly equal frequencies.

Because of the electric quadrupole moment of the odd S isotopes, the $J=1\rightarrow2$ transition is split into several components which are only partially resolved.[1] From the shape of the pattern one can infer the nuclear spin, while for the magnitude of the separations one may evaluate the quadrupole coupling constant.[5]

The significant spectral frequency referred to in the Eqs. (2) and (3) are those of the center of gravity of the $J=1\rightarrow2$ group of lines for one isotopic molecule. The displacement of the strong central line can be evaluated from the quadrupole coupling constant.[5]

Table I contains a summary of our data on S^{35} along with comparable results for the stable isotopes as measured by Geschwind and Gunther-Mohr.[6]

By taking the ratio of the intervals between the upper and lower minor components and the central one for S^{35} one gets a value of 1.48, which is in excellent agreement with the theoretical ratio of 1.46 for a nuclear spin of $\frac{3}{2}$. Clearly, the sign of the quadrupole moment of S^{35} is opposite that of S^{33}, since the patterns are inverted with respect to each other.[7] The value of the quadrupole constant is 20.5±0.2 Mc/sec. Townes and Dailey[8] have made a rough calculation of the molecular electric field gradient in OCS to evaluate the electric quadrupole moment of S^{33}. Using their figures, we get a value of 0.06×10^{-24} cm^2 for the electric quadrupole moment of S^{35}, which is considered to be good to within a factor of 2.

As a result of the quadrupole interaction, the strong central line is shifted by 0.440±0.003 Mc/sec down in frequency. For the most reliable value of the OCS^{35} frequency we took the value of the OCS^{34} frequency

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* Research carried out under contract with the AEC.

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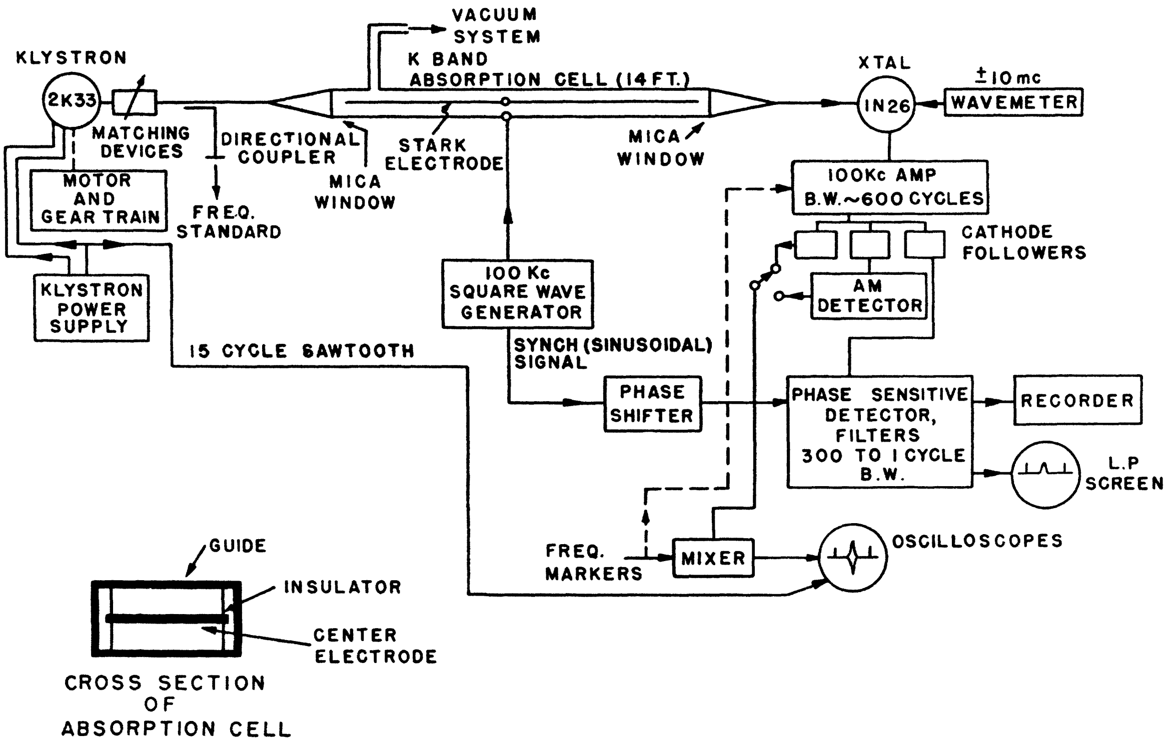

FIG. 1. Block diagram of microwave spectroscope.

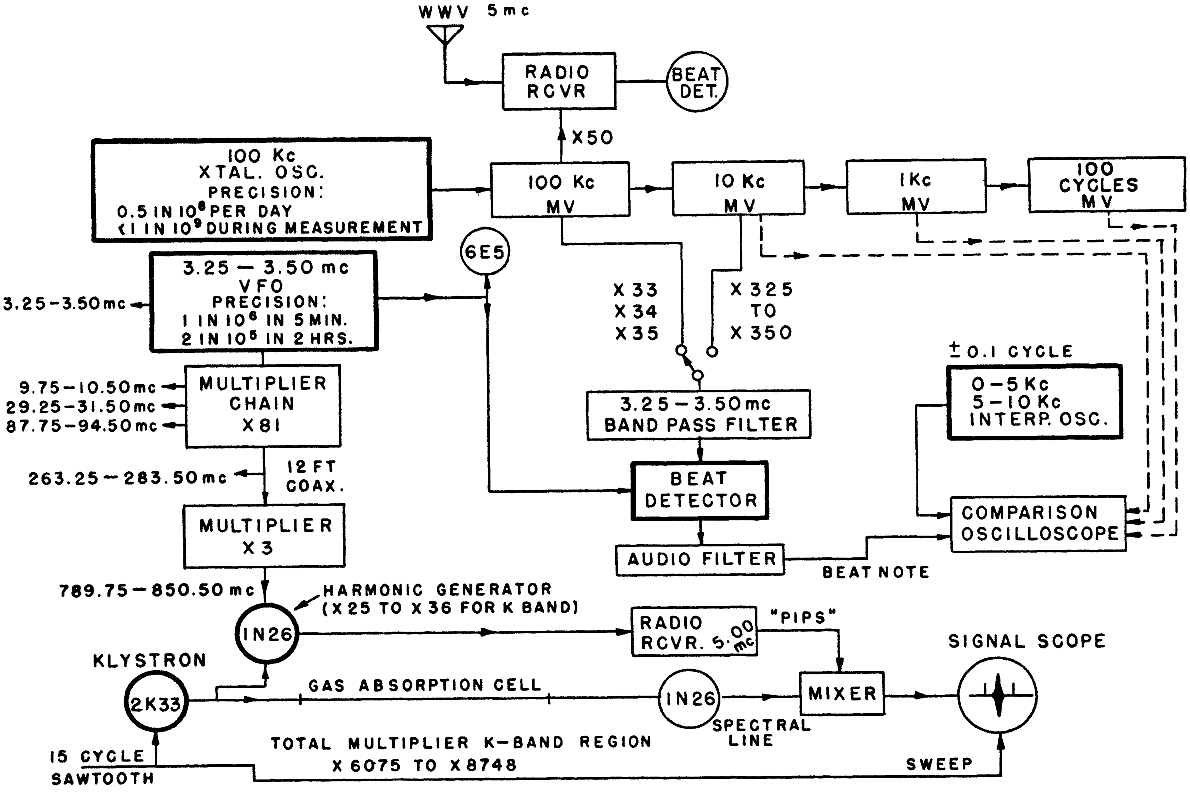

FIG. 2. Block diagram of the microwave frequency standard.

TABLE I.

Frequencies and frequency differences for isotopic OCS (Mc/sec)	OCS^{32} [a]	OCS^{33} [a]	OCS^{34} [a]	OCS^{35}
Center of gravity of pattern	24325.921 ±0.002	24019.641 ±0.004	23731.299 ±0.003	23457.403 ±0.011
$F=1/2\rightarrow 3/2$		24012.292		23462.343
$5/2\rightarrow 5/2$		24012.974		
$1/2\rightarrow 1/2$				
$3/2\rightarrow 5/2$		24019.641		23456.963 ±0.011
$5/2\rightarrow 7/2$		24020.264		
$3/2\rightarrow 3/2$		24025.467		23453.323
Frequency differences between centers of gravity (Mc/sec)				
$(\nu^{32}-\nu^{33})$ [a]		$(\nu^{32}-\nu^{34})$ [a]		$(\nu^{34}-\nu^{35})$
306.280±0.005		594.622±0.004		273.896±0.010

[a] Reference 6.

given in Table I and subtracted our value of the $S^{34}-S^{35}$ difference.

The moment of inertia I_0 of a triatomic linear molecule as a rigid rotor can be expressed as

$$I_0=[\Pi(m)/\Sigma(m)][(l_1^2/m_3)+(l_2^2/m_1)+(l_1+l_2)^2/m_2] \quad (1)$$

where for O, C, and S, m_1, m_2, and m_3 are the respective masses,

$l_1=\text{O}-\text{C}$ internuclear distance,
$l_2=\text{C}-\text{S}$ internuclear distance,
$\Pi(m)=m_1m_2m_3, \quad \Sigma(m)=m_1+m_2+m_3.$

The spectral frequency for the $J\rightarrow J+1$ transition will be given to a good approximation by[9]

$$\nu_0=h(J+1)/4\pi^2I_0. \quad (2)$$

From Eqs. (1) and (2) one can evaluate an expression involving mass differences between isotopes m_3', m_3'', and m_3''':

$$\frac{m_3'-m_3''}{m_3'-m_3'''}=\frac{(\nu''-\nu')}{(\nu'''-\nu')}\frac{\nu'''}{\nu''}\frac{(m_1+m_2+m_3'')}{(m_1+m_2+m_3')}, \quad (3)$$

where $\nu'=$ frequency for the molecule $m_1m_2m_3'$,
$\nu''=$ frequency for the molecule $m_1m_2m_3''$,
$\nu'''=$ frequency for the molecule $m_1m_2m_3'''$.

Equation (3), valid only for isotopic substitution of the same end atom, implies two approximations: (1) that the corresponding bond distances are the same in each isotopic molecule, (2) that the small vibration-rotation interaction terms which have not been evaluated completely are proportional to mass. The error thus introduced has been discussed at length by other authors[3, 7, 10–13] and has been shown to be of the order of one part in 10^4 in the mass difference ratio.

In our calculations we have taken the following mass values:

$$C^{12}=12.00386, \quad O^{16}=16.00000;$$
$$S^{32}=31.98199, \quad S^{33}=32.98168;$$
$$S^{34}=33.97890 \quad \text{and} \quad S^{35}=34.98006.$$

Since we have frequency data on four isotopic molecules, we can evaluate two mass difference ratios independently. We get

$$(m_{35}-m_{32})/(m_{34}-m_{32})=1.50155\pm0.00015,$$
$$(m_{35}-m_{32})/(m_{33}-m_{32})=2.99881\pm0.00030.$$

The principal contribution to the probable errors assigned is the estimated value of the error introduced by the neglect of the vibration-rotation interaction.

Evaluating the $m_{35}-m_{32}$ mass difference in terms of Davison's[14] mass differences

$$m_{34}-m_{32}=1.99691\pm0.00037$$

and

$$m_{33}-m_{32}=0.99963\pm0.00012,$$

we get for $m_{35}-m_{32}$, respectively 2.99844±0.00042 and 2.99770±0.00048. The agreement between these two values partially justifies the assumptions of Eq. (1). These values also check with the value 2.99818±0.00045 as given by Low and Townes computed from nuclear reaction data.[15] The absolute value of the S^{35} mass is dependent upon the absolute value of S^{32}. Since there has been some question concerning the accuracy of the published S^{32} masses, this will be the principal limitation on the S^{35} mass. Assuming the value of S^{32} mass of 31.98199±0.00021 as given by Penfold[16] we get the value for the S^{35} mass of 34.98006±0.00038.

We have calculated the OC—CS bond distances using Eq. (1) by taking pairs of moments of inertia and solving for l_1 and l_2. Since four isotopic molecules have been studied, six combinations are possible. One would expect that the differences in values would be chiefly due to the differences in the errors introduced by the neglect of the vibration effects. The results of these calculations are given in Table II. The variations are found to be greater than would be calculated only from experimental error. The consistency of the values given

TABLE II. Internuclear distances.

Isotopic combination used	C−O	C−S
32, 33	1.1634×10^{-8} cm	1.5584×10^{-8} cm
32, 35	1.1636	1.5583
33, 35	1.1638	1.5582
32, 34	1.1641	1.5579
33, 34	1.1648	1.5573
34, 35	1.1626	1.5591
Average	1.1637 mean deviation 0.0005	1.5582 mean deviation 0.0004

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serve as a very rough check that use of Eq. (1) is justified.

The S^{35} was furnished by the AEC Isotopes Branch in the form of $H_2S^{35}O_4$ with only about one-half percent of the S present being S^{35}. Considerable effort was spent in working out a chemical process for converting the small total amount of sulfur (approx. $\frac{3}{4}$ mg) into OCS without reducing the ratio of S^{35} to S^{32}. Our procedure was essentially as follows:

(1) precipitating the $SO_4^=$ with $Ba(OH)_2$ in a Pt test tube and removing the supernatant liquid by centrifuging and decantation;

(2) reducing the dried $BaSO_4$ to BaS at 900°C in an H_2 atmosphere;

(3) adding I_2 and HCl in an alcohol solution in excess to produce elementary S;

(4) carefully drying the S under vacuum and then distilling it;

(5) adding CO and baking for about 16 hr at 450°C.

The total gas pressure in the absorption cell of the spectroscope was about 120μ. In spite of the low S^{35} concentration, the central OCS^{35} line was distinctly visible in an oscilloscope presentation of the output of the 100-kc amplifier. The satellite lines were observed and measured by cooling the guide with dry ice, using a 1-cycle sweep, a phase-sensitive detector, and an oscilloscope with a long persistent screen. Graphical records were also obtained by using a gear-driven Klystron, a phase-sensitive detector, and an ink recorder.

We wish to express our appreciation to Mr. James Sharman for his valuable assistance with the electronic circuits and especially the frequency standard. We also wish to thank Miss Theresa Danielson for her assistance with some of the numerical calculations.

PHYSICAL REVIEW VOLUME 81, NUMBER 6 MARCH 15, 1951

Photo-Activation of Bismuth*

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(Received November 10, 1950)

Work is reported on nonfission activities produced in bismuth under irradiation with photons of 48- and 86-Mev maximum energies. The results are similar to those obtained by bombardment with high energy particles in that many particles are emitted.

THE activation of bismuth with high energy photons from the University of Chicago betatron has been reported to result in fission[1] as well as in the production of activities tentatively assigned to Bi^{204} and Bi^{206}. This paper reports some nonfission activities produced in bismuth under irradiation by photons of 48- and 86-Mev maximum energies. The results in general resemble those obtained by bombardment with high energy particles in that many particles (spallation) are also emitted when high energy photons are used.

Two procedures were followed. (1) Bismuth foils of one gram in weight and 2 cm^2 in area were irradiated for about 4 hr and counted directly on an Eck and Krebs counter of 30 mg/cm^2 wall thickness at a geometry factor of about 15 percent. (2) Aliquots containing 15 g of bismuth from a 500 ml solution, prepared by dissolving in conc. HNO_3 150 g of bismuth irradiated for about 4 hr, were subjected to analysis for lead and thallium. The isolated lead and thallium samples weighing about 15 mg and of 2-cm^2 area were counted on a thin end-window counter at a geometry factor of about 20 percent. The isolations of lead and thallium from the irradiated bismuth were made at various times after the end of the irradiation in order to distinguish between the activity of a lead or thallium species formed directly in the irradiation from that descended from radioactive parents.

The radioactive species observed in irradiated bismuth, their probable assignments, the nuclear reactions leading to their formation, and the activity level of each species appear in Table I. The 86-Mev irradiations were performed at 55 cm from the target and at a radiation intensity of about 2400 r/min; the 48-Mev irradiations were made at 55 cm from the target and at a radiation intensity of about 900 r/min.

The assignments given to the radioactivities in Table I were made on the basis of reported[2] radioactivities and the observed trend in the level of activity with the number of emitted particles. The reactions given for the individual activities of Table I are those consistent with the mass assignments and are written in the most elementary way for balance of mass and charge. In the cases of the isolated lead and thallium activities, the data were analyzed to give the activity at the end of irradiation of the parent as well as the daughter activity formed independently. Thus, for example, from the activity of 68-min Pb^{204m} formed independently, it was found that at 86 Mev the yield of the $(\gamma, 4np)$ reaction was about 1/50 that of the $(\gamma, 5n)$ reaction, as determined from the activity of 68-min Pb^{204m} formed by the $(\gamma, 5n)$ reaction followed by electron capture (designated e.c. in Table I).

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