MILLIMETER WAVE SPECTROSCOPIC COMPONENTS

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The very rapid development of the field of microwave electronics from the late 1930's to the present has opened up the region of the electromagnetic spectrum lying between the "radio" region and the infrared region. It is not generally realized, however, that these recent developments were by no means the earliest work in the field of centimeter and millimeter waves --- for example, Heinrich Hertz[1] published in 1889 a description of the apparatus with which he generated and detected radiation of approximately 50 cm wavelength. In 1893-94 the Italian scientist, Righi,[2] conducted experiments at even shorter wavelengths, and in 1923 the Americans, Nichols and Tear,[3] published results of experiments covering the entire region from 50 mm to 0.22 mm --- from 5 kMc to 1300 kMc! Using small tungsten spark gaps, paraffin lenses, an eschellete grating, and a thermal-type detector they were able to prove experimentally the complete equivalence of "radio" and "infrared" radiation, thus closing the gap which had existed between the two.

The new short waves were not applied to molecular spectroscopic research until ten years after the work of Nichols and Tear. In 1933 Cleeton and Williams[4] measured the wavelength of the very strong ammonia inversion spectrum occurring at 1.2 cm to an accuracy of about one percent. It is interesting to note that their work preceeded the use of waveguide as a carrier of microwave energy. Hence, they used reflectors to direct the output of their home-made magnetrons through a large rubberized bag of ammonia.

Eleven years passed before the next successful spectroscopic measurement in the microwave region was made. In 1944 R. Beringer[5] working at the MIT Radiation Laboratory, measured the absorption of oxygen and oxygen-nitrogen mixtures at 5 mm wavelength. He used a K-band reflex klystron to drive a crystal harmonic generator, and passed the energy thus obtained through a waveguide absorption cell --- an ordinary section of waveguide with vacuum-tight mica windows at each end and provision for admitting and pumping out the gas sample to be studied.

After the war, when microwave tubes and components became generally available to research workers, developments in spectroscopy came very rapidly. Hershberger at RCA, Bleaney and Penrose at The Clarendon Laboratory, and Good at Pittsburgh made early measurements and developed at K-band the techniques which are still in use. This work was done in the region covered by the 2K50 and 2K33 klystrons, but as soon as Raytheon made available a line of tubes extending the frequency coverage to 40 kMc spectroscopes for higher frequencies came into service.

The need for higher frequencies still existed, however, and Beringer's early work with crystal harmonic generators indicated a way to attack the problem. In 1949 Smith, Gordy, Simmons and Smith[6] reported work in the three-to-five millimeter region utilizing second harmonics of both K and K_A band klystrons, and by 1950 Simmons and Anderson[7] had made spectral measurements up to 96 kMc using third harmonic energy of a K-band fundamental. In 1951 Gordy, C. M. Johnson and co-workers[8] extended this work to 130 kMc by using fifth harmonic radiation.

A different approach to the problem was made at Columbia, where Loubser, Townes and others utilized magnetron harmonics at frequencies as high as 120 kMc for spectroscopic work. This is discussed by A. H. Nethercot in a succeeding paper.

The harmonic generators used at second through fifth harmonics in the work at Duke University were basically similar in design, and made use of conventional 1N26 crystal cartridges. Fig. 1 shows a generator of this type. The larger guide conducts the fundamental power to a probe; the probe is common to both the larger and the smaller guide, and to the center conductor of the coaxial crystal. As might be expected, crystals manufactured and tested for use as K-band superheterodyne mixers did not always work well as harmonic generators. The problem of tuning up a system involving an harmonic generator, a detector, and a signal which, when peaked, was only about 15 db above noise was a most formidable one and limited to a great degree the work which could be done.

The need for better harmonic generators and detectors prompted the development by Gordy and King[9] of the unit shown in Fig. 2. Energy coming down the larger guide is picked up by the probe and presented to the silicon crystal by the 2 mil tungsten cat-whisker. The internal dimensions of the small guide are .075 x .034 inches, the aperture between the two crossed waveguides is .016 inches in diameter, and the crystal chip itself is cylindrical in shape, .040 inches in diameter, fitting into a .042 hole in the waveguide wall. Whisker length is adjusted so that the face of the crystal chip is very nearly co-planar with the upper wall of the small guide when contact is made.

In this unit the pressure of cat-whisker against crystal can be adjusted by advancing or retracting the crystal chip. Despite the softening effect of the hair-pin spring in the cat-whisker, this is a most critical adjustment, and is made by means of a differential nut mechanism having an effective pitch of 364 turns per inch.

For operation at the shorter millimeter wavelengths the point of the whisker is very critical. It is necessary that the point be very sharp; i.e., with a radius of curvature at the tip of the order of one ten-thousandths of an inch or less. Such points are readily produced by immersing the end of the tungsten wire in a ten-normal solution of potassium hydroxide and passing 6 volt ac current through it until electrolytic action ceases. (This method of pointing whiskers was used by Beringer in 1944, and it is described in detail in Torey and Whitmer.[10]) We have found that for work in the 2 to 3 millimeter region slightly more rounded points may be satisfactory, but for 2 millimeters and above the sharper points are preferable.

A video detector mount of similar design has been constructed and used in conjunction with the harmonic generator. As shown in Fig. 3, the internal dimensions are the same as for the multiplier unit. D.C. insulation and R.F. bypassing is provided by a small condenser consisting of a thin mica washer between the waveguide and the cat-whisker support. It is possible that R.F. choking might produce a gain in efficiency at a particular frequency, but at the cost of some reduction in broad-bandedness. A considerably neater mechanical arrangement designed by C. M. Johnson of such a detector is shown in Fig. 4. Here a differential screw mechanism, rather than a differential nut, has been used.

These units have been used in a wide-band video-type sweep spectroscope for the measurement of molecular absorptions up to 291 kMc (1.03 mm), and have been used by Burras and Gordy[11] in a narrow-band reflector-modulated video spectroscope at frequencies up to 391 kMc (.77 mm, or in infrared terminology, 770 microns wavelength).

The type of sweep spectroscope used by most workers is shown in Fig. 5. A saw-tooth sweep voltage is applied to the reflector of the klystron oscillator and to the horizontal deflecting plates of an oscilloscope, thus obtaining a panoramic display of frequency. The output of the klystron is fed to the harmonic generator, and its output in turn is carried through a vacuum-tight length of waveguide to a detector. A modified Type P amplifier (or other low noise amplifier) follows the detector, and its output is displayed on the oscilloscope. Because the molecular absorptions are narrow (Q's of the order of 100,000), while reflections and other resonances of the microwave circuits are relatively broad, a moderately high low-frequency cut-off in the amplifier tends to filter out the system resonances while passing the absorption characteristics. A bit of the low-frequency noise output of the video detector is eliminated by this filtering also.

Microwave frequencies can be measured approximately with a cavity wavemeter by superimposing the wavemeter pip and the absorption line on the oscilloscope, or can be measured to an accuracy of one or two parts per million by comparison with the multiplied output of a low-frequency crystal oscillator calibrated against WWV.

The decrement of power due to molecular absorption shows up on the scope as a sharp discontinuity. At centimeter wavelengths and gas pressures of about .01 mm of mercury such absorptions may decrease the power level by only a tenth or a hundredth of one percent; but since the absorption strength of the line varies with the cube of the frequency absorptions approaching 90% or so are found in the short millimeter wave region.

The resonant absorption frequencies of diatomic, linear, and symmetric top molecules in the ground vibrational state are almost but not quite harmonically related:

Diatomic and linear molecules:

$$f_{abs} = 2B(J+1) - 4D_J(J+1)^3$$

Symmetric top molecules:

$$f_{abs} = 2B(J+1) - 4D_J(J+1)^3 - 2D_{JK}(J+1)K^2$$

where $J = 0, 1, 2, \ldots$

$K = 0, 1, \ldots, J.$

The constant B is of the order of 10 kMc, while the constants D_J and D_{JK} are of the order of kilocycles. Hence, the frequency of an absorption is given to an accuracy of a few parts per hundred thousand by the first term alone, and to this accuracy the absorptions are harmonically related. However, the distortion terms (which are a measure of how the shape of the molecule changes as it rotates) represent a small, but measurable, displacement of the absorption from its undistorted frequency, and are <u>not</u> harmonically related.

Thus, if the output of the harmonic generator contains several harmonics several transitions may be observed simultaneously. Fig. 6 shows an oscillogram of six transitions of the molecule OCS (carbonyl sulfide). These correspond to the 4th through the 9th harmonics of a K-band reflex klystron. The separation of the strongest and the weakest of these lines (referred to the fundamental oscillator frequency) is only 2.74 Mc, although the absorptions range in frequency from 97 to 219 kMc. Such absorptions give definite proof that energy of the corresponding order harmonic is present in the output of the harmonic generator.

At millimeter wavelengths the problem of using high-pass filters to reject lower order harmonics of K-band power sources becomes very difficult. Table I indicates the excessively tight tolerances which would have to be imposed on dimensions in order to use beyond-cutoff waveguide filters.

Another serious handicap in the use of such filters is that the desired harmonic will be severely attenuated since it is so very close to the cutoff of the guide. In practice this has rendered impractical the use of filters beyond 100 kMc; the multiplier and detector units described earlier have their cutoffs at 79 kMc.

TABLE I

High-Pass Filter Dimensions for Harmonics of a 25 kMc Fundamental

Harmonic	f kMc	λ mm	cutoff dimension (inches)	f/fc for next higher harmonic
4	100	3.00	.0591	1.25
6	150	2.00	.0394	1.17
8	200	1.50	.0295	1.12
10	250	1.20	.0236	1.10
12	300	1.00	.0197	1.08

Insertion of filters with cutoff frequencies higher than this usually affects adversely the power in the very high harmonics. For example, if harmonics four through eleven of a K-band fundamental are detectable in a spectroscope, insertion of a filter to suppress fourth and fifth harmonics will usually eliminate the tenth and eleventh as well.

The multiple presentation of absorption lines shown in Fig. 6 is a great help in tuning up a spectrometer to a particular frequency region. Invariably three or more lines (corresponding to fourth, fifth, and sixth harmonics) are seen immediately upon application of power, but it is a matter of some difficulty to obtain the 10th or 12th harmonic. Optimizing the setting of end-walls and other tuning adjustments on the various harmonics in a step-wise fashion offers one of the simplest, most rapid tuning procedures developed to date.

The techniques described have already been used at Duke, Columbia, and Johns Hopkins to measure the transition frequencies of a number of molecules. In particular, carbonyl sulfide has been studied at frequencies from 24 to 369 kMc. Of especial interest to those concerned with atmospheric characteristics, the resonant absorption frequency of water vapor at 183,311 Mc has been measured, and it is hoped that more experimental data on resonance absorptions of water near one mm wavelength will be forthcoming. Tables of atmospheric absorption calculated from microwave and infrared spectroscopic data on water and oxygen have been prepared at the Air Force Cambridge Research Center and should become available soon.[12]

Because of their high Q and the high accuracy with which the frequencies of these absorptions are known they provide excellent calibration points throughout the microwave region. For example, in conjunction with a spectrometer capable of producing fourth through eighth harmonic energy, the ground vibrational state of the molecule OCS alone provides twenty-four frequencies between twenty and thirty kilomegacycles which are known to within two parts per million or better. Furthermore, these frequency points are fixed; unlike a crystal oscillator they have absolutely no drift with aging, and cannot be altered by improper handling or misuse.

For these reasons the Bureau of Standards has developed the so-called "atomic clock", in reality a microwave oscillator frequency-stabilized against an absorption line. A recent development at Columbia indicates that Q's of the order of millions are achievable by means of forced emission, and it is quite possible that eventually the primary unit of time may be defined in terms of molecular characteristics, just as the unit of length has been defined in terms of the characteristic wavelength of light emitted by mercury. In the meantime, molecular absorptions furnish very good calibration points and high-Q discriminators for stabilization systems wherever greatest reproducibility is required.

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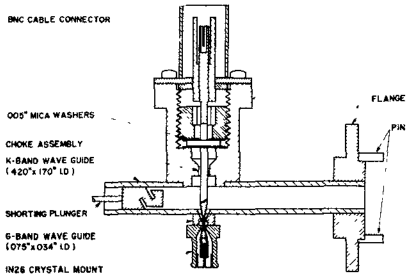

Fig. 1 - Crossed-waveguide harmonic generator for use with conventional IN26 coaxial cartridge crystals.

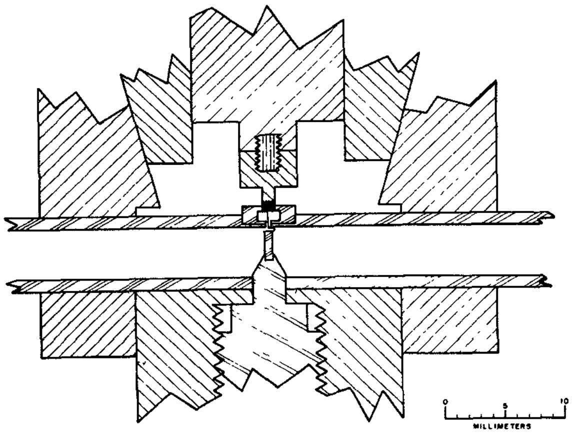

Fig. 2 - Crossed-waveguide harmonic generator utilizing built-in, adjustable crystal chip and finely pointed catwhisker.

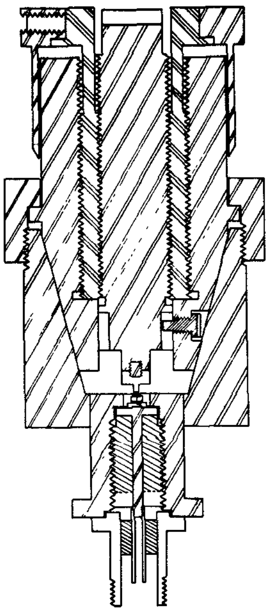

Fig. 3

Video detector for 79 kMc and above with crystal mounted in guide. Differential nut mechanism for adjusting crystal-catwhisker contact is identical with that used in harmonic generator.

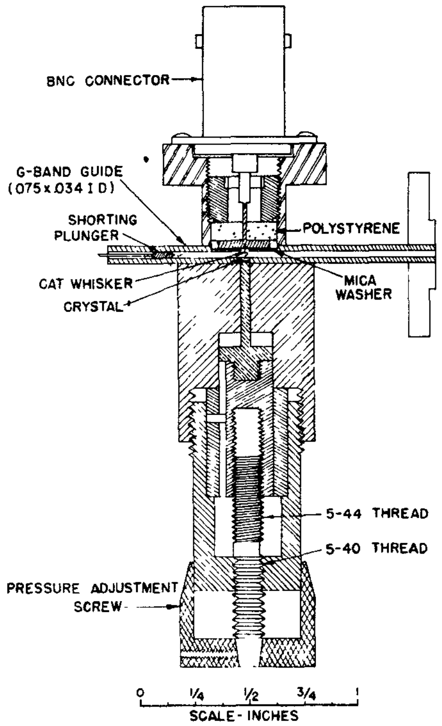

Fig. 4 - Video detector with differential screw mechanism for adjusting contact designed by C. M. Johnson.

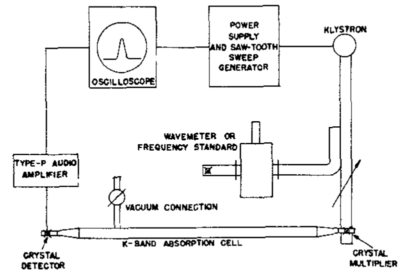

Fig. 5 - Block diagram of video sweep spectroscope.

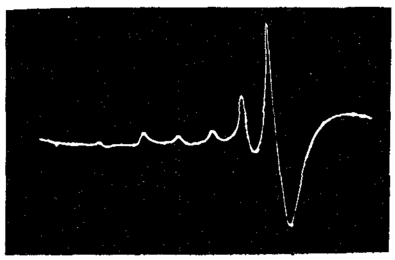

Fig. 6

Oscillogram of the gas OCS taken on sweep spectroscope at pressure of 0.01 mm Hg. From right to left, transition frequencies are 97.3, 121.6, 145.9, 170.3, 194.6, and 218.9 kMc.