A STARK MODULATED
MICROWAVE SPECTROGRAPH

Thesis for the Degree of M. S.
MICHIGAN STATE COLLEGE
Carroll Forester Augustine
1953

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Robert H. Noble
Major professor

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A STARK MODULATED MICROWAVE SPECTROGRAPH

by

Carroll Forester Augustine

A THESIS

Submitted to the School of Graduate Studies of Michigan
State College of Agriculture and Applied Science
in partial fulfillment of the requirements
for the degree of

MASTER OF SCIENCE

Department of Physics

1953

ACKNOWLEDGEMENT

I wish to express my sincere thanks to Dr. Robert H. Noble for suggesting this project and for his ever willing guidance during the stages of its completion. Gratitude is also expressed to Dr. Robert D. Spence for his helpful suggestions.

Carroll L. Augustine

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I. INTRODUCTION

Microwave spectroscopy is a comparatively new branch of spectroscopy utilizing radiation with wavelength between the region .20 to 15 centimeters. This region is similar to the infrared in that the absorption of radiation by material is detected more readily than the emission of radiation. The reduction of intensity of radiation of a known frequency on passage through the sample is measured.

In a gas at low pressure, absorption takes place at very sharply defined frequencies--the rotational frequencies of the molecules. These rotational frequencies can be calculated when the structural parameters of the individual molecules are known. Solids and liquids give absorptions which depend upon the interactions of the molecules as well as upon their structural parameters; therefore, these absorptions cannot be so readily calculated.

The field of microwave spectroscopy had its beginning with the pioneering experiment of Cleeton and Williams[1] at the University of Michigan in 1934. They used a combination of microwave and optical techniques to measure the inversion spectrum of ammonia in the wavelength region of 1.25 centimeters.

Cleeton and Williams published the only paper on microwave spectroscopy prior to World War II. In post-war years the field of spectroscopy in the microwave region has grown with tremendous rapidity. This growth has been stimulated chiefly by the great advances in microwave techniques during the last war and the fact that the method provides the molecular structure investigator with a new source of valuable information. Microwave spectroscopy permits precise evaluation of absorption frequencies (to about one part per million) and resolving powers thousands of times better than the best infrared spectrometers; thus contributing to a more complete knowledge of the spatial arrangement of atoms in the molecule, as well as a study of nuclear spins and quadrupole moments.

II. GENERAL PRINCIPLES

A. The Simplest Type of Spectrograph

In microwave spectroscopy the energy source is a reflex klystron. The klystron is a monochromatic source of high frequency electromagnetic radiation. The fact that the energy source is monochromatic accounts for the principle difference between the techniques of microwave and infrared spectroscopy. In infrared spectroscopy the source is not monochromatic. This necessitates the use of some method of dispersion, for example, a prism or grating. We may compare the two techniques in the following way. In infrared spectroscopy the energy source is not monochromatic but radiates instead a wide band of frequencies. After the energy passes through a sample, the frequencies are separated by the dispersion instrument and the amount of absorption at the various frequencies determined by an infrared sensitive detector. In microwave spectroscopy the detector is equally sensitive to all frequencies generated by the klystron and the frequency of the klystron is varied until the exact maximum of absorption for one spectral line is determined. Practical construction difficulties which limit the amount of dispersion and hence, the resolving power of the infrared spectrograph, are therefore not manifest in the microwave spectrograph.

One of the earliest high resolution spectrographs is the

direct absorption instrument outlined in Figure 1 and an explanation of its operation will aid in understanding the Stark modulated spectrograph.

If a D. C. voltage is applied to the repeller of a reflex klystron and varied over a range of a few hundred volts, there will be several regions in which the tube will oscillate and somewhat larger regions in which the tube is inoperative. The microwave frequency in an individual oscillating region, called a mode, changes with the voltage, being of the order of 50 megacycles lower on the low voltage end than at the high voltage end of the mode.

A saw-tooth voltage applied to the repeller along with a large D. C. voltage will thus sweep the tube over a small portion of the spectrum at a rate determined by the frequency of the saw-tooth voltage. A mode of the tube is the half circle displayed on the oscilloscope of Figure 1, but without the irregularities. The deflection above the horizontal line gives the intensity of radiation which reaches the crystal detector, whereas the position along the horizontal axis gives the frequency.

In this spectrograph, the amplitude of the saw-tooth voltage can be varied to change the magnitude of the spectral region viewed per sweep. To change the frequency limits of the mode there is a mechanical adjustment which changes the shape of a resonant cavity in the klystron. The usual reflex klystron can be tuned over several thousand megacycles.

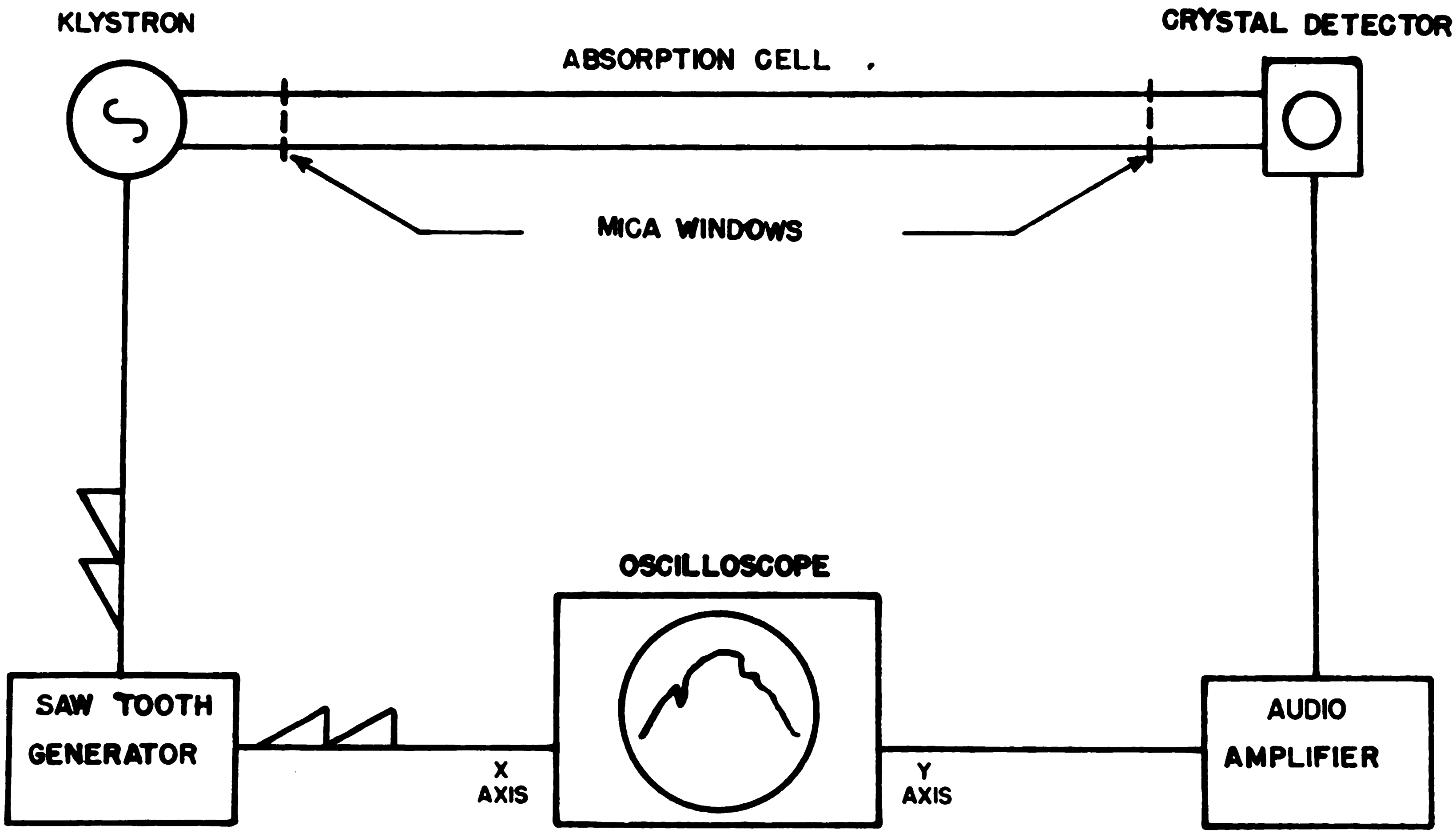


Figure 1. Direct Absorption Spectrograph

The radiation is propagated through rectangular copper waveguides. The absorption cell consists of a piece of waveguide made vacuum tight at the ends with mica windows. The rectified output from the crystal detector is amplified by a low frequency audio amplifier and displayed on the oscilloscope. At low pressures, around 10^{-1} to 10^{-3} millimeters of mercury, a strong absorption is displayed as a small dip in the mode, such as the one shown at the left of the center of the mode in Figure 1. The wider, smoother dip on the right side is one that might be caused by standing waves in the waveguide system.

The poor sensitivity of the direct absorption spectrograph just described limits its usefulness. Also, the dips in the mode caused by standing waves are troublesome since it is sometimes extremely difficult to tell whether a dip in a mode is due to molecular absorption or a standing wave. We are, therefore, presented with a dual problem of increasing sensitivity and decreasing spurious signals caused by standing waves. The Stark modulated spectrograph is one solution to the problems.

B. Stark Modulation

The Stark modulation scheme makes use of the change in the absorption frequency of a gas molecule when the molecular energy levels are perturbed by an external field, that is, the so-called Stark effect.[2] Referring to Figure 2, microwave energy from a source is passed through the gas to be studied

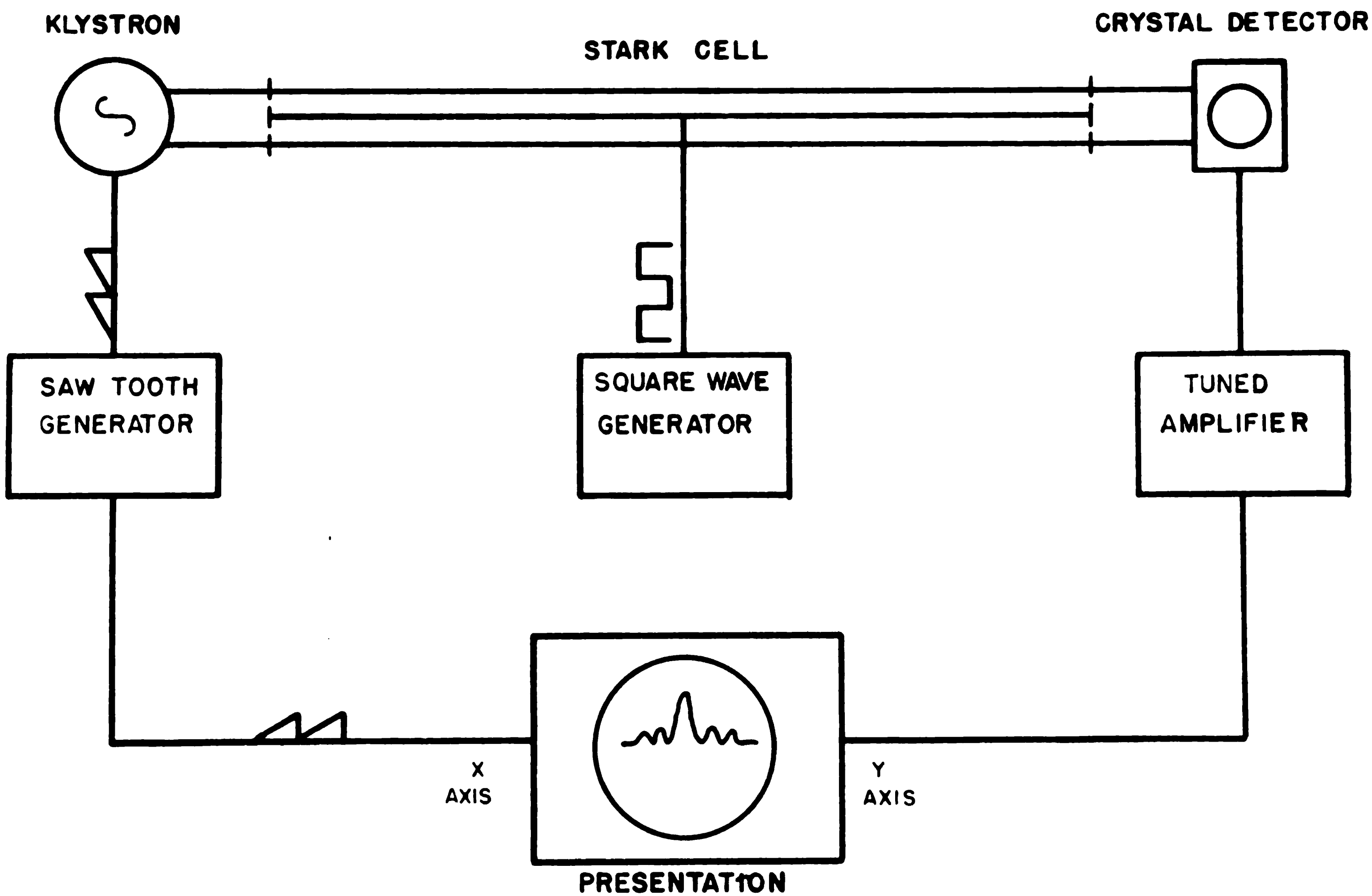


Figure 2. Stark Modulation Spectrograph

which is confined in a waveguide cell by transparent mica windows. If the source is tuned to an absorption frequency then a diminished amount of power will be received at the crystal detector. However, the absorption frequency of the gas is being alternately switched between two frequencies corresponding to the peaks and troughs of an externally applied high frequency square wave based on zero voltage. This field is applied between an insulated electrode strip running down the center of the waveguide and the waveguide itself. Hence, the signal reaching the crystal detector is amplitude modulated, or as we say, Stark modulated at a high frequency rate and may be amplified and placed on the vertical plates of an oscilloscope.

As before in the direct absorption instrument, a frequency axis is provided on the oscilloscope by frequency modulating the source with a low frequency saw-tooth voltage and applying this same voltage to the horizontal deflection plates of the oscilloscope.

An increase in sensitivity over the direct absorption instrument is possible since by detecting and amplifying at a high frequency, one avoids the lower frequency region where there is high crystal and source noise.

Spurious signals caused by standing waves are completely eliminated since the amplifiers following the crystal detector may be sharply tuned to the Stark modulation frequency, thereby admitting only those signals which are due to molecular absorption.

Stark modulation frequencies may be as high as 100 kilocycles. It is convenient to operate in this region since narrow band communications receivers may be used as amplifiers. If frequencies in excess of 100 kilocycles are used, the true absorption line shape becomes distorted.[3,4]

The signal on the oscilloscope shown in Figure 2 illustrates the type of pattern we might expect to see. The large blip in the center is the absorption at the original line frequency. The two smaller blips on either side are Stark components.

C. Source Modulation

Gordy and Kessler,[5] and independently, Hershberger,[6] have shown that high frequency source modulation can be employed to achieve high sensitivity. Upon the slowly varying saw-tooth voltage used to sweep the klystron over its mode, there is superimposed a voltage wave of a much higher frequency (it may be as high as several megacycles) which causes the oscillator to make rapid excursions into and out of the regions of greater absorptions as the spectrum line is gradually passed over by the slow sweep. The spectrum line then acts as a discriminator to convert the frequency modulation into an intensity modulation of the same frequency. After detection by a crystal, the signal is then amplified by a narrow band radio frequency amplifier which is tuned to the modulation frequency. It is then passed through an audio amplifier with a rather sharp low frequency cut-off filter which discriminates against low frequencies generated as a result of reflections in the microwave line. Gordy[7] has suggested that as an alternative, one could use a low frequency Stark modulation in combination with a high frequency source modulation.

This is an excellent suggestion for the following reasons. As explained previously, for maximum sensitivity the optimum Stark modulation frequency should be in the region of 100 kilocycles. However, the construction of a square wave generator to work into the low impedance of the absorption cell at this frequency is difficult and expensive. Whereas, following the suggestion of Gordy, the Stark modulation frequency may be in the low audio range and yet sensitivity is not sacrificed since the source modulation may be at any frequency that can conveniently be amplified by an ordinary communications receiver. We have followed Gordy's suggestion in the design of this spectrograph.

D. Complete Block Diagram

A complete block diagram of the Stark modulated spectrograph which we have constructed in shown in Figure 3. The design of individual components will be explained in detail later--the intention now is to give a brief picture of the over-all operation.

The block diagram of the actual spectrograph depicted in Figure 3 is, of course, more detailed than the simple spectrograph previously explained and shown in Figure 2. However, with S_1 open and with S_2 in the left hand position, the over-all

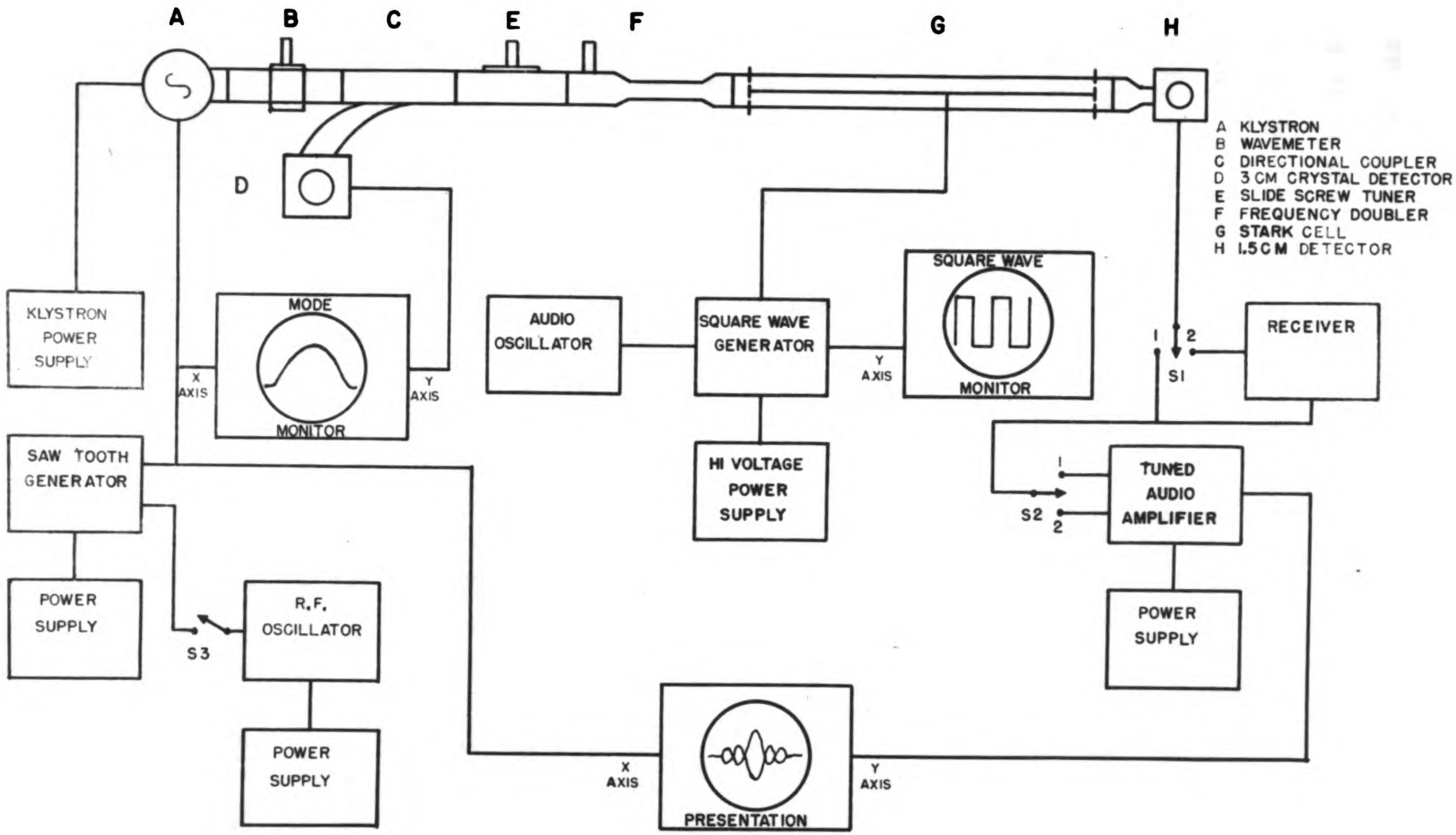


Figure 3. Complete Block Diagram

operation is exactly the same. Switches S_1, S_2, and S_3 do not exist as such but they will aid in this explanation.

The klystron (A) is a Varian, type X-13, and has a frequency range of 8,100 to 12,400 megacycles. The klystron is followed by a wavemeter (B) and a directional coupler (C). The directional coupler removes a very small fraction of the energy that would otherwise go straight to the absorption cell. This energy is detected by a three centimeter crystal detector (D), the output of which is connected to the vertical deflection amplifiers of a monitoring oscilloscope. In this way, the mode of the klystron may be constantly monitored while adjustments are being made.

The absorption type wavemeter will absorb a small amount of energy at a narrow band of frequencies determined by the mechanical tuning of the absorption cavity in the wavemeter. If the klystron is being electronically swept through a range of frequencies which include the absorption frequency of the wavemeter, a narrow dip will be observed in the klystron mode. The dip may be observed on the monitoring oscilloscope, but since, as explained earlier, the tuned amplifiers following detector (H) are designed to pass only those signals due to molecular absorption, the dip will not appear on the presentation oscilloscope. In order to determine the frequency of a particular spectral line, it is necessary to first make the X axis of the monitoring and presentation oscilloscopes the same length. The wavemeter dip is then moved to a position along the X axis of the monitoring oscilloscope corresponding to the position of the spectral line of the presentation oscilloscope.

The only klystron available for this work was one which generated energy having a wavelength of approximately three centimeters. Most of the simpler molecules exhibit strong absorption lines in the one and one-half centimeter region.[8] Therefore, in order to examine this higher frequency portion of the spectrum, it was necessary to construct the crystal doubler (F) which doubles the frequency of some of the energy generated by the klystron, and allows only this higher frequency to be propagated to the absorption cell (G).

Crystal detector (H) is a one and one-half centimeter detector and the signal from this detector may be channeled one of two ways as indicated by S_2.

Now, there are essentially four different ways in which the electronic components may be arranged to form four different types of spectrographs. These are: 1. straight absorption 2. Stark modulated 3. source modulation and 4. a combination of Stark and source modulation.

For straight absorption the square wave generator is left inoperative, S_2 is on the left contact and S_3 is in the straight amplifier position. With S_3 in this position, the amplifier is not tuned to any specific frequency but provisions are made within the amplifier to put a high pass filter in series with the signal thus eliminating the low frequency contour of the mode and many of the indentations in the mode caused by reflections and standing waves. This makes it possible to use a higher gain setting on the Y axis amplifier of the presentation oscilloscope and amplify the higher frequency components, that is, the sharper indentations in the mode caused by molecular absorption.

For Stark modulation, S_1 is open, S_2 is on the left contact, and S_3 is in the tuned amplifier position. A square wave of the desired frequency and amplitude is applied to the center electrode of the Stark cell.

For source modulation, S_1 is closed, S_2 is on the right contact, and S_3 is set for straight amplification which must include the high pass filter. The radio frequency generator and communications receiver are, of course, tuned to the same frequency.

For a combination of source and Stark modulation, S_1 is closed, S_2 is on the right contact, and S_3 is in the tuned amplifier position. Operating under these circumstances, the picture on the presentation oscilloscope is just as it would be with Stark modulation alone. The only difference to be expected is an increase in sensitivity.

It would be well to point out here that although we have followed the previously mentioned design suggested by Gordy, we have not departed greatly from the conventional design of a Stark modulated spectrograph. In order to obtain a combination of Stark and source modulation, we have added only two principle components--the communications receiver and the radio frequency generator. For the sake of economy we have constructed a low frequency 3000 cycle square wave generator instead of the higher frequency generators ordinarily used. There is, therefore, a sacrifice in sensitivity due to amplification at this low frequency when straight Stark modulation is used.

Although the Stark plus source modulation scheme is more sensitive, it is also much more difficult to tune and to keep in adjustment. For nearly all ordinary investigations, it is safe to say that the sensitivity obtainable from straight Stark modulation is adequate.

Now, some of the individual components will be explained in more detail.

III. EQUIPMENT DESIGN

A. Vacuum System

The vacuum system is designed to evacuate the absorption cell and provide a means of injecting a gas or vapor sample into the cell. A diagram of the system is shown in Figure 4 and a photograph is shown in Figure 5.

Two vacuum pumps are used; a mechanical fore pump and a two stage water cooled mercury diffusion pump. The diffusion pump is very effective when it is operating against the low back pressure provided by the fore pump. Lower pressures are arrived at much more quickly when the two pumps are operating together than when the fore pump is working alone.

To prevent mercury vapor from entering the manifold and damaging the pressure gauges, a vapor trap is placed between the diffusion pump and manifold. The trap is intended to be immersed in a slush composed of acetone and dry ice. The low temperature provided in this way will freeze out any mercury vapor that may enter the trap.

The vapor trap is connected to the manifold through a four millimeter stopcock A, which provides a way of isolating the manifold from the pumping system. Two two millimeter stopcocks D and E are connected to the manifold and terminated with eight millimeter tapered joints. It is through these stopcocks that samples of vapor or gas may be injected into

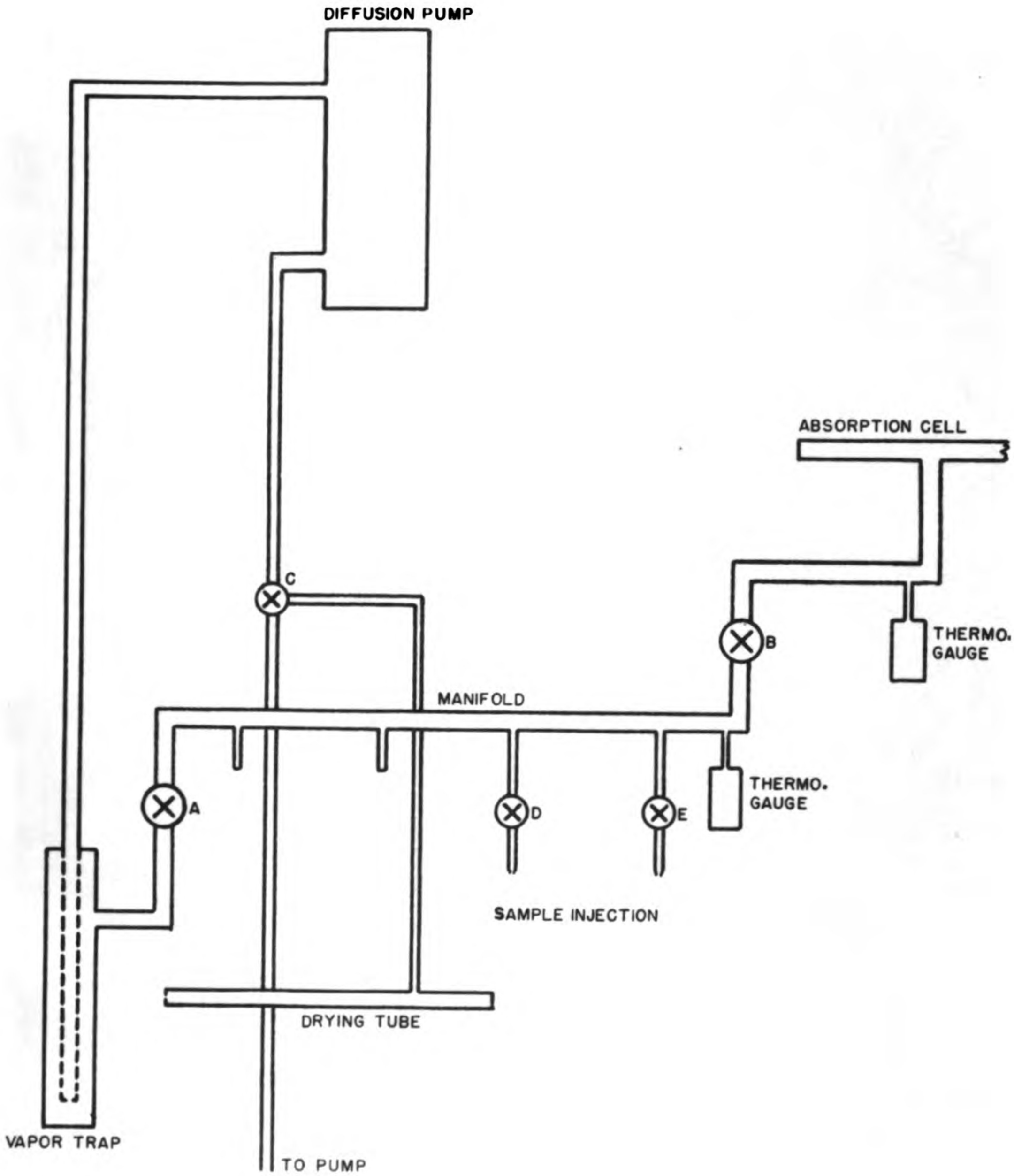


Figure 4. Diagram of Vacuum System

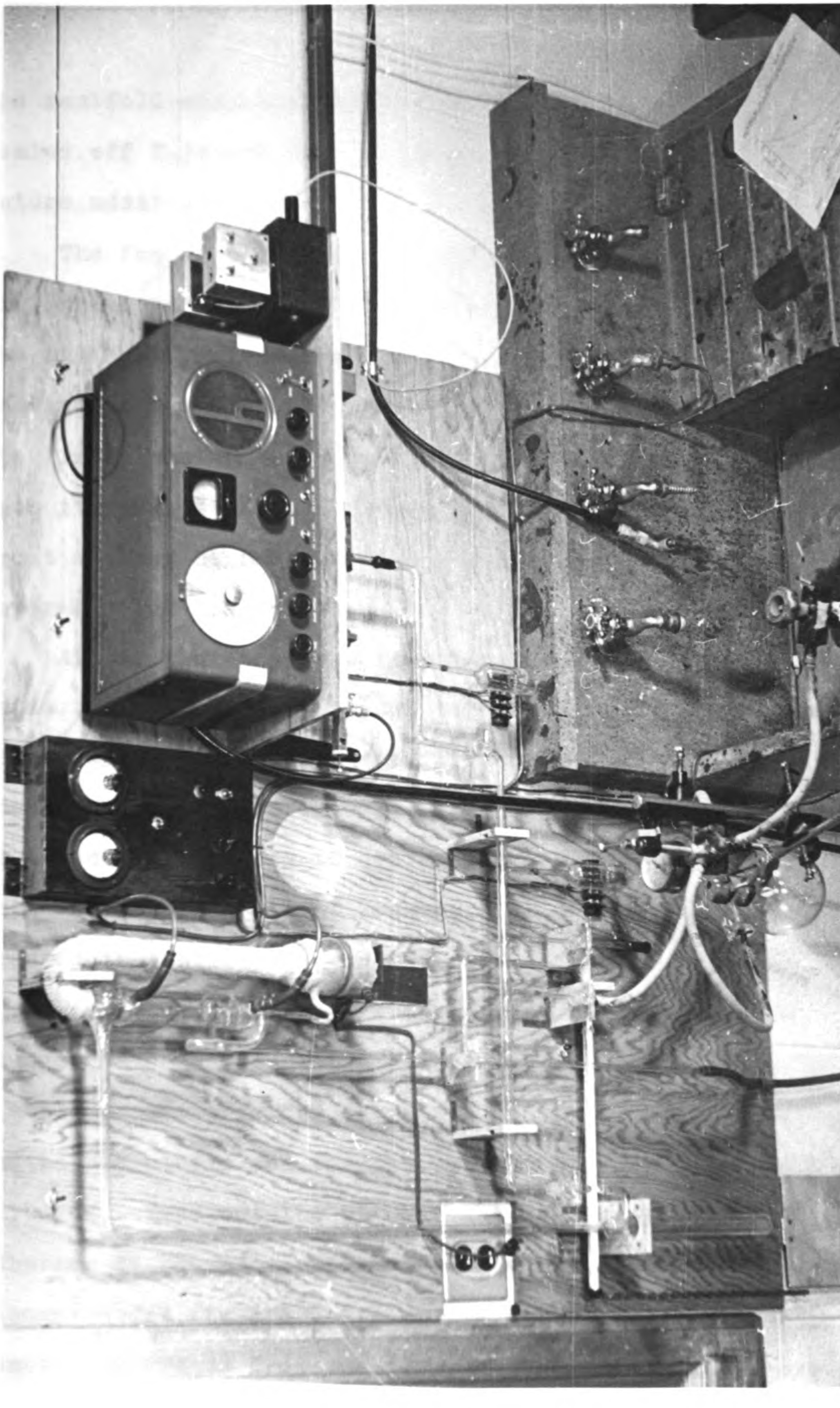


Figure 5. Vacuum Sytem

the manifold and utlimately into the absorption cell. Two sealed off T joints leading from the manifold allow for future additions.

The function of the three way stopcock C is to let air, which enters through a small hole in the far end of the drying tube, into the fore pump line or into the line leading to the diffusion pump. All three, or any two of the lines leading to stopcock C may be connected, depending upon its position. The drying tube is filled with a drying agent and may be removed from the system for cleaning at a tapered joint connection.

All of the stopcocks have been hand ground with cerium oxide to insure a more vacuum tight fit. The plungers and accompanying receptacles are file marked and the plungers should never be interchanged. Experience has shown that there is a general tendency to overgrease a vacuum stopcock, and this results in a slight leakage. If a stopcock is to be greased the following procedure is suggested.

First, clean the old grease from the receptacle and plunger using carbon tetrachloride and a clean cloth. Put a small amount of stopcock grease on the plunger, insert it in the receptacle and turn it until the grease is evenly distributed. Take out the plunger and remove all of the grease adhering to the plunger. Reassemble the stopcock, turn the plunger until the grease is evenly distributed, and again remove the grease from the plunger. Repeat this process several times until the grease remaining in the stopcock is an extremely thin, well distributed layer.

There are two Central Scientific Company type 94180 thermocouple vacuum gauges included in the system. The reader should refer to literature provided by the Central Scientific Company for information concerning the theory of operation, current ratings, etc. of this type gauge. One gauge is connected to the manifold and the other is connected directly to the line leading to the absorption cell. When the absorption cell is isolated from the manifold by closing stopcock B, the two pressures may be determined independently.

A wiring diagram for the gauges is shown in Figure 6. The heater sections of the thermocouple junction are connected in series. The rough and fine adjustments control the heater current which is indicated by the milliammeter. A double throw double pole switch, S_1, places the microammeter in series with the current generating arms of either of the two thermocouples. Curves supplied by the manufacturer relate pressure with heater and generated current.

If the absorption cell has contained a gas at low pressures for any length of time, sufficient pumping should be done to completely outgas the system before another sample is placed in the cell. With constant pumping, it may take as long as seven days to completely outgas the cell. The following procedure has proven helpful in determining when outgassing is completed and whether or not the system is leaking.

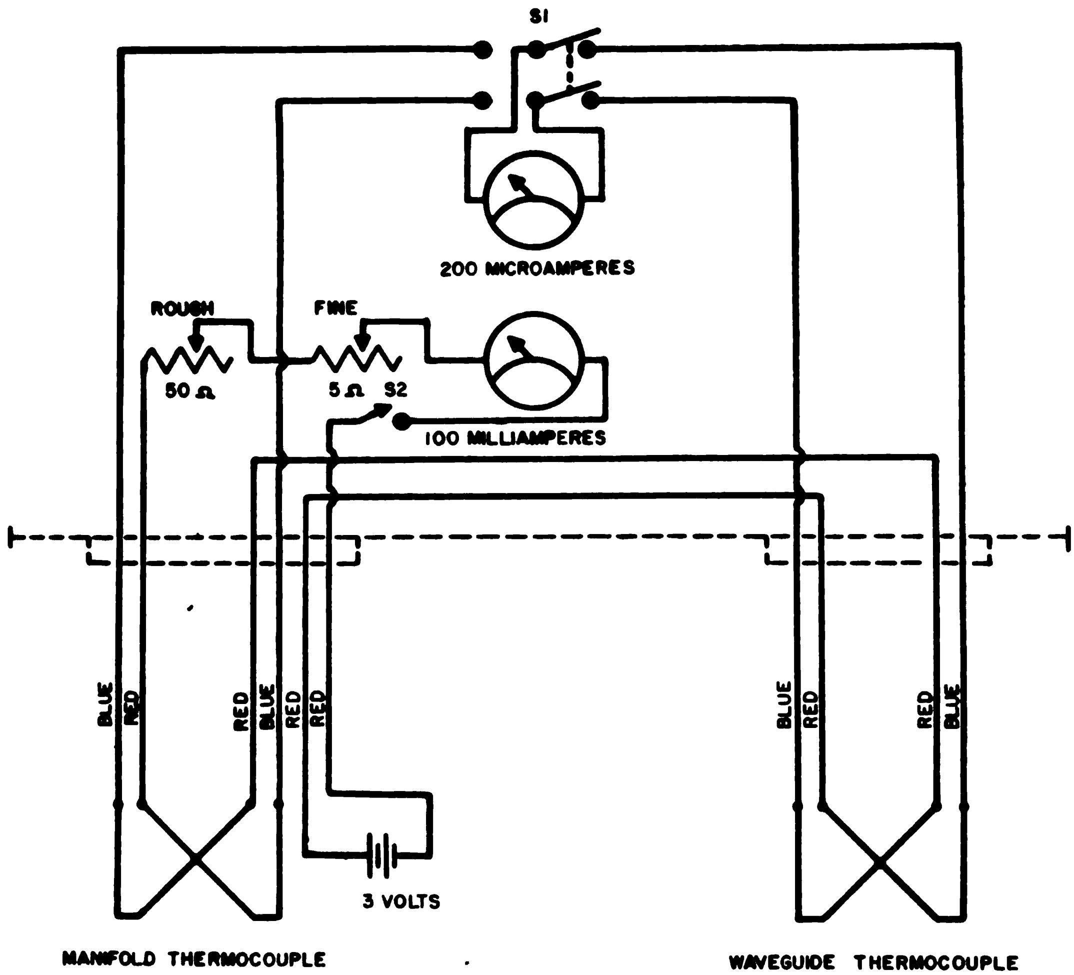


Figure 6. Vacuum Gauge Wiring

Start the pumps and record the time. After twenty-four hours of pumping, close stopcock B and keeping the heater current constant, make a plot of waveguide thermocouple current versus time. A one hour record taken at five minute intervals is sufficient. Open stopcock B and resume pumping. Repeat the process once a day, for several days. If there are no leaks and as the outgassing continues, the curves will approach a straight line. If the system is leaking, successive plots will become identical.

When outgassing is complete, the sample is let into the manifold through D or E with A and B closed. B is then slowly opened and the sample allowed to gradually pass into the cell until the desired pressure is reached. Any sudden increase in cell pressure may rupture the mica windows. The following is a list of noteworthy precautions.

1. Always keep the line between the fore pump and stopcock C at about atmospheric pressure when the fore pump is not running. Oil may be drawn into the system if the line is left evacuated.
2. Don't attempt to run the diffusion pump against large back pressures. The mercury may erupt violently if it has been heated under a high back pressure and this pressure is suddenly decreased by a large amount.
3. Do not change pressures between parts of the system rapidly. This may result in mercury being blown

from the trap into the manifold.

4. Remove all detector crystals before checking the glass for leaks with a tesla coil.
5. Keep the stopcocks tight. When turning the plunger, balance a steady inward force by a force applied to the body of the stopcock in the opposite direction.
6. Keep a constant flow of water through the diffusion pump and never run the diffusion pump without a dry ice and acetone slush surrounding the vapor trap.

B. Stark Cell

The Stark absorption cell consists of a twelve foot length of X band waveguide made vacuum tight at the ends by mica windows. A cross sectional diagram is shown in Figure 7. The flat copper strip which serves as the center electrode extends nearly the full length of the cell and is supported on the sides by grooves cut in six inch strips of insulating polystyrene.

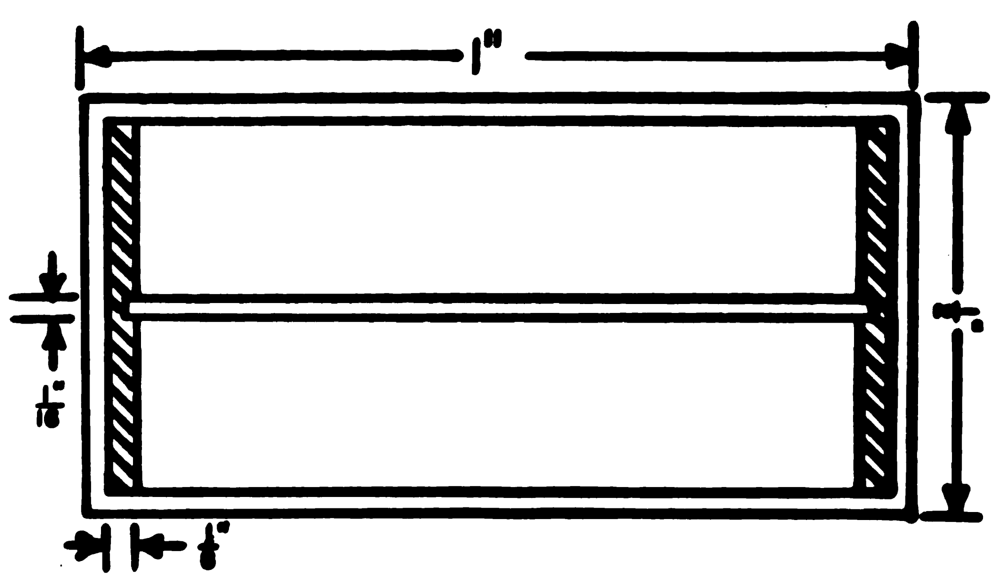


Figure 7. Stark Cell

The grooves were cut with a special jig on a milling machine to insure uniform depth and spacing. The jig is available as part of the equipment should additional strips be needed.

The cell was assembled by simultaneously feeding the electrode and strips into one end of the waveguide. It was found convenient to pass a wire down the guide and pull the electrode through rather than try to push it. Tolerances on the polystyrene strips were held as closely as possible to insure uniform spacing of the electrode, since this is the limiting factor in the precision with which the applied Stark field is known. An off center electrode will cause a doubling of the Stark pattern and make interpretation difficult.

The mica windows were sealed on with plycene. Each window was shaped to fit just inside of the flange of the choke couplings on the ends of the cell. Plycene was then placed around the edges of one of the windows and a temporary seal made over one end of the cell by simply heating the coupling to the melting point of plycene and holding the window in place until the coupling cooled and the plycene was firm. A permanent window on the opposite end of the cell was then made in a similar manner except that before the plycene had entirely hardened, the mechanical pump was turned on for a few seconds. The slight difference in cell and atmospheric pressures exerts a steady inward force on the window causing the window to warp slightly and distribute the plycene uniformly between it and

and the coupling. When the plycene was completely firm, the temporary window was removed and replaced by a permanent one made in an identical manner. Windows placed over the ends of the cell in this way were found to be much more leakproof than those placed on without any pressure difference.

Electrical connection to the electrode was made by running a wire through a small hole drilled through the short dimension of the waveguide. A polystyrene tube was placed through the hole to provide insulation. This feed-through was made vacuum tight by repeated applications of glyptol around the outside and inside of the polystyrene tube.

C. Crystal Doubler

As mentioned earlier, the only klystron available for this work was one which generated energy having a wavelength about three centimeters long. Only a few lines have been recorded at this low frequency but there are many strong absorption lines recorded in the one and one-half centimeter region. In order to perfect the equipment, it was necessary to operate in a region where strong, well-known lines were present. The crystal doubler changes some of the energy emanating from the klystron with a wavelength of three centimeters to energy having a wavelength of one and one-half centimeters. The klystron used was a Varian type X-13, having a frequency range from 8.100 to 12.400 kilomegacycles. Doubling the frequency yields a range of from 16.2 to 24.8 kilomegacycles. There are many strong absorption lines in this region.[8] A group of particularly strong lines are those due to the inversion spectrum of ammonia[9] distributed between the limits of 19.218 to 39.941 kilomegacycles.

A diagram of the crystal doubler is given in Figure 8 and it is shown in operating position in Figure 9. Energy emanating from the klystron enters the doubler from the left and a portion of this energy is rectified by a 1N23 type crystal fixed to the end of a plunger. This plunger may be raised or lowered perpendicular to, and in the center of, the long dimension of a two inch section of three centimeter waveguide. A gradual three and one-half inch taper follows this three centimeter section and is connected to a six inch piece of one and one-half centimeter waveguide. An identical taper at the far end brings the cross sectional dimensions back up to those of the three centimeter waveguide.

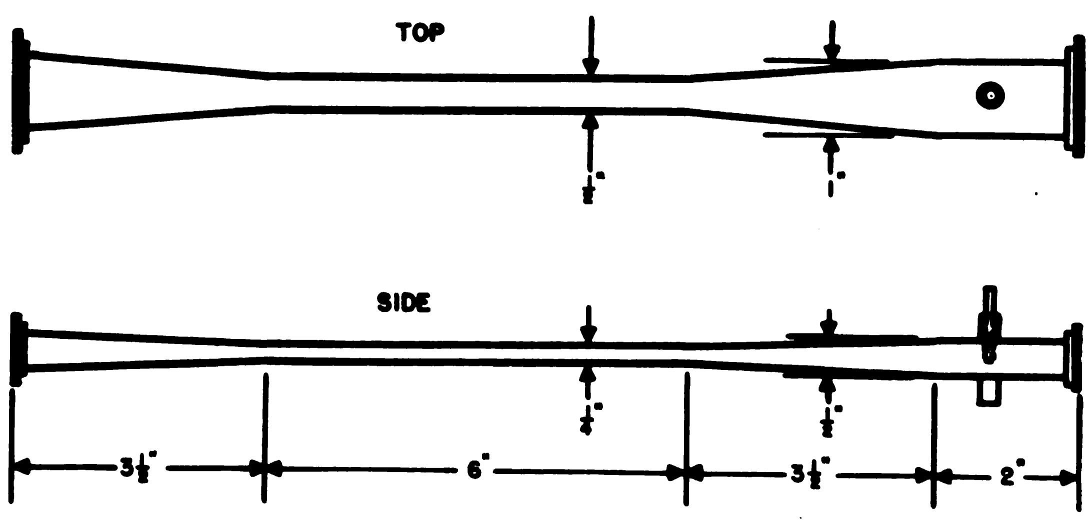


Figure 8. Crystal Doubler

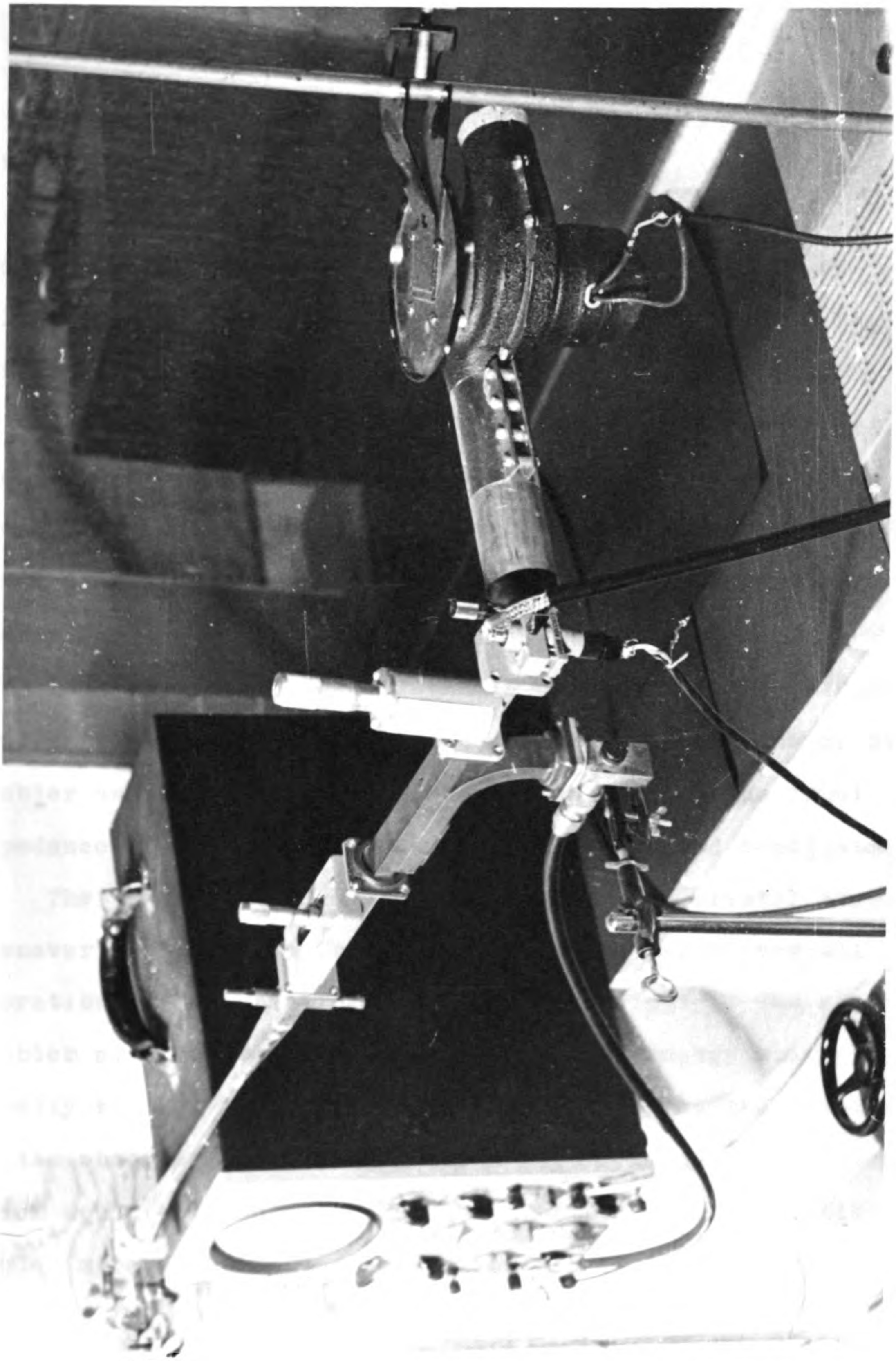


Figure 9. Wave Guide Components

The Fourier series representation for a rectified sine wave is: $$f(x) = \frac{1}{\pi} - \frac{2}{\pi}\sum_{1}^{\infty}\frac{\cos 2nx}{4n^2-1} + \frac{1}{2}\sin x$$

and the n=1 term is a strong second harmonic of the fundamental frequency. The magnitude of the higher order terms falls off rapidly. We wish to propagate the n=1 term corresponding to the one and one-half centimeter wavelength and eliminate the three centimeter energy. This is accomplished by the section of one and one-half centimeter waveguide which has transverse dimensions too small to propagate the three centimeter energy.

The position of the crystal is quite critical. As the center frequency of the klystron mode is changed, the crystal doubler and the slide screw tuner which matches the input impedance of the crystal doubler, will both need readjustment.

The necessity of carefully adjusting the crystal doubler whenever the klystron frequency is changed makes over-all operation of the spectrograph somewhat unwieldy. The crystal doubler also greatly reduces the amount of energy which finally reaches the absorption cell, and hence, the intensity of the observed spectra is low. A higher frequency klystron which would eliminate the necessity of the crystal doubler would improve the ease of operation and the results.

D. Sweep Generator

The sweep generator is designed to deliver a linear saw-tooth waveform of desired amplitude and frequency to the klystron repeller and to the horizontal amplifiers of the monitoring and presentation oscilloscopes. A schematic diagram is shown in Figure 10.

The design of the first stage is quite conventional. The rough frequency control condensers selected by S_1 are charged through the current path provided by the 6SK7 and discharged through the 884 thyratron. The constant plate current characteristics of the pentode are utilized in this way to improve the saw-tooth linearity. Control R_2 provides a fine frequency adjustment for each condenser selected by S_1. The range of frequencies available with each condenser are as follows:

2	microfarads	2	cycles/second	to	9	cycles/second	
.5	"	8	"	"	to 45	"	"
.1	"	40	"	"	to 193	"	"
.002	"	167	"	"	to 760	"	"

The 884 grid bias control should be adjusted so that the maximum frequency obtainable with the two microfarad condenser is nine cycles per second.

A synchronizing output is provided and may be used to synchronize the sweep generators in the monitoring and presentation oscilloscopes should such an arrangement be desired. The synchronizing output is a sharp positive pulse having an amplitude of about 20 volts.

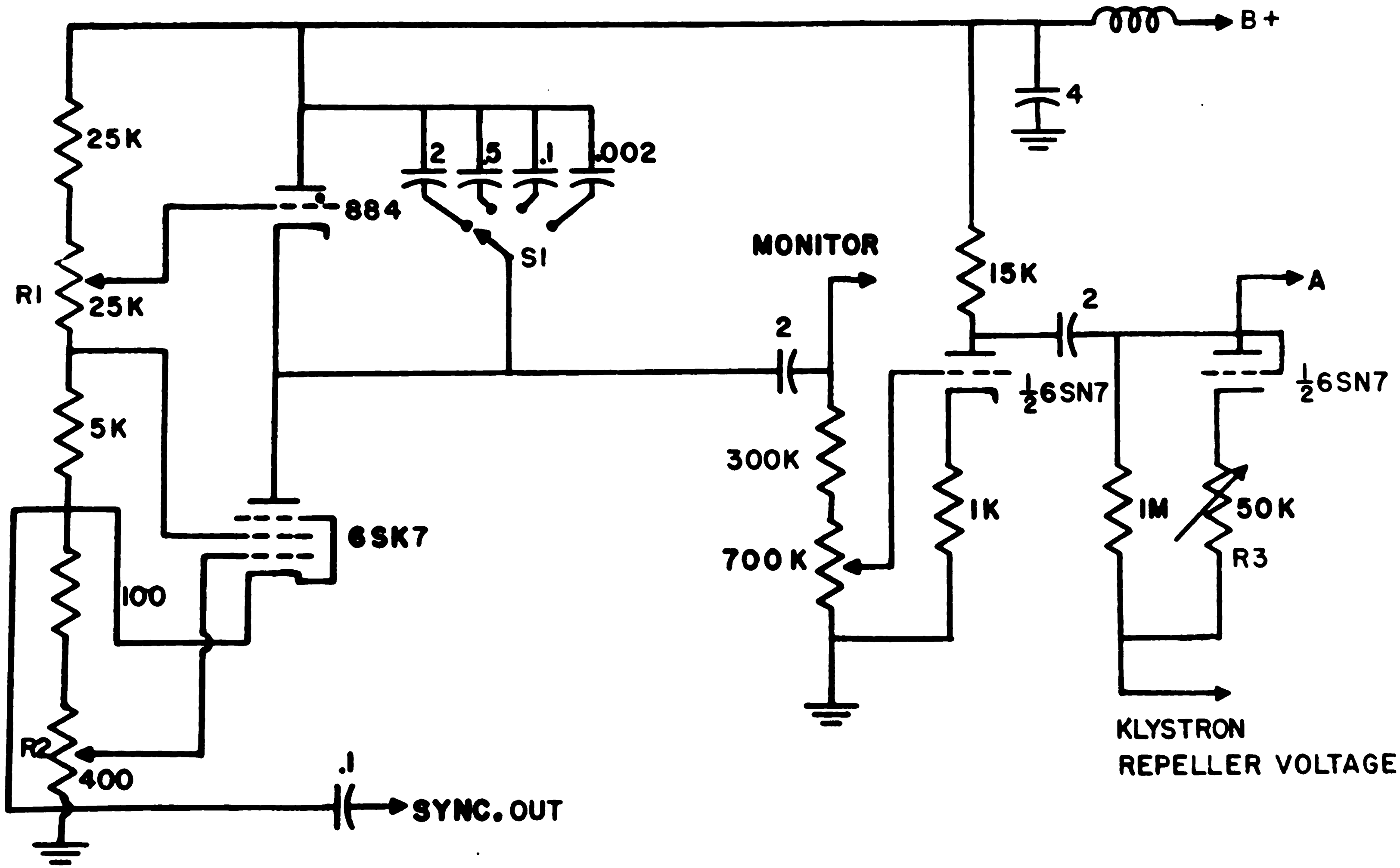


Figure 10. Sweep Generator

The saw-tooth waveform generated by the first stage is placed on the grid of one of the triode sections of a 6SN7. An output for the monitoring and presentation oscilloscopes is taken off following the coupling condenser. This triode is intended to be primarily an isolation rather than an amplification stage. A 700K potentiometer in the grid circuit controls the saw-tooth wave amplitude which will eventually appear on the klystron repeller. A maximum amplitude of 75 volts is possible.

The second triode section of the 6SN7 is connected as a diode and acts as a negative clamper. The clamping action provides a constant reference potential on the klystron repeller.

The same power supply serves both the sweep circuits and portions of the high voltage square wave generator. In order to eliminate interference between the two units, it was necessary to place an inductance capacitance filter in series with the B + supplying the sweep generator.

E. Square Wave Generator

The purpose of the square wave generator is to produce a well shaped, high voltage square wave based on zero voltage, which will charge and discharge the relatively large capacitance of the Stark cell. The capacitance of the Stark cell is .002 microfarads. An elementary calculation indicates that if we are to keep the charge and discharge times of the cell within appropriate limits, that is, if we intend to keep the edges of the square wave reasonably straight, we must be able to handle average charge and discharge currents on the order of .6 amperes. This figure was arrived at by assuming a square wave with an amplitude E of 1000 volts, a frequency f of 3000 cycles per second, and the fraction of the period allowable for charge or discharge equal to 1/100 of the period T. Hence, $I = \frac{Q}{Tk} = \frac{CEf}{k} = .6$ amperes. The best way that has been found to handle these large currents is to use one bank of high emission tubes to charge the cell and another bank to discharge it.

A schematic diagram of the square wave generator is shown in Figure 11. The input at J_1 to V1a is taken from a Hewlett Packard Model 200D audio signal generator. The V1a stage changes the shape of the input sine wave into a waveform more suitable for triggering a multivibrator. Values of cathode and plate resistance were selected so as to place V1a much nearer cut-off than saturation. Therefore, when a large sine wave voltage is placed on the grid, the output taken from the plate is a rectified sine wave. The abrupt change in the waveform when the clipping or rectifying action begins is sufficient to trigger the Eccles-Jordan multivibrator which follows as the next stage. Since the grid cut-off voltage is a constant for V1a, the point on the input sine wave where clipping action begins is a function of the sine wave amplitude. Therefore, the length of the straight line portion of the rectified sine

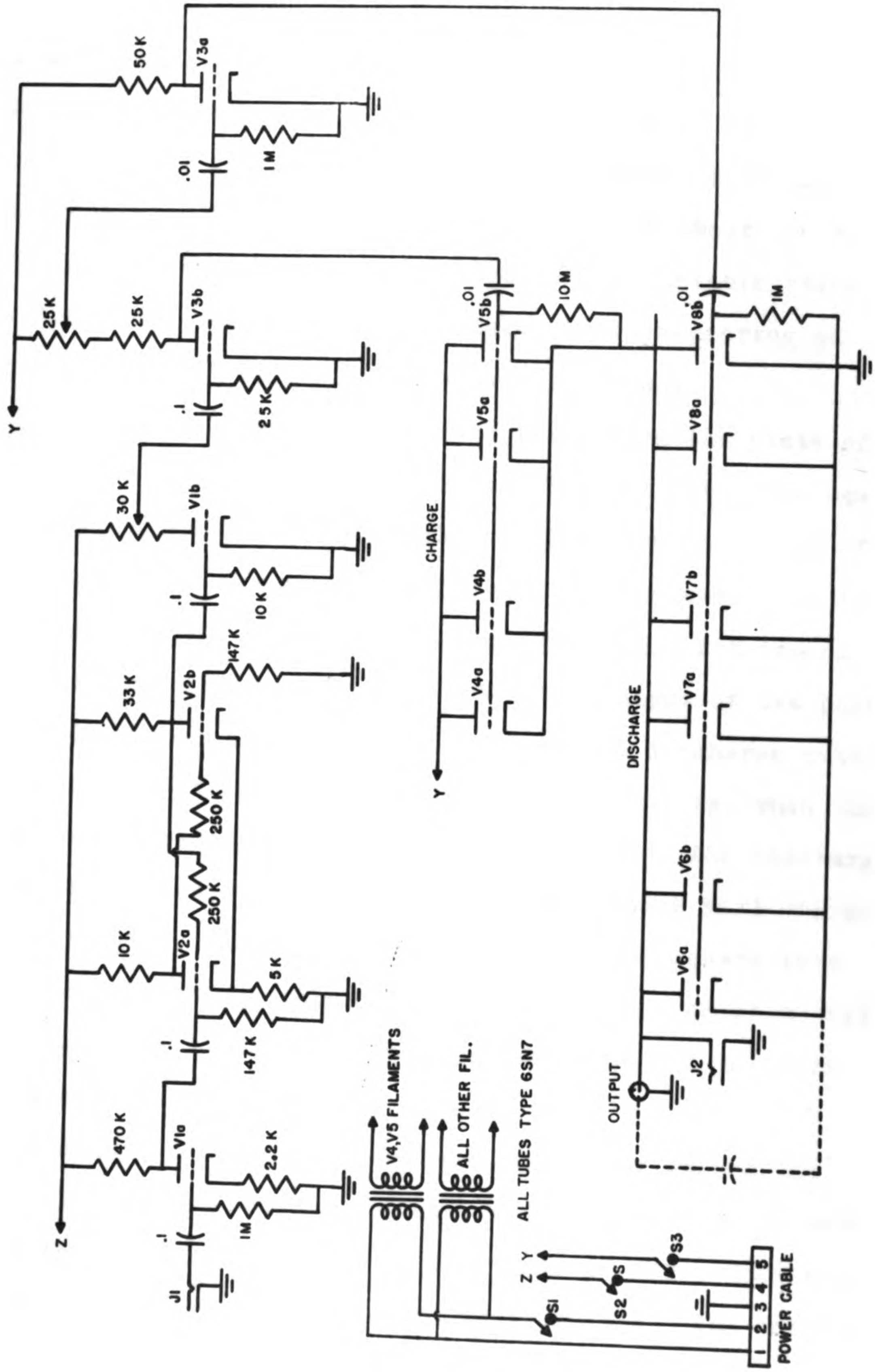


Figure 11. Square Wave Generator

wave, and hence, the length of time that the multivibrator spends in each stable state is also a function of the sine wave amplitude. The maximum output of the model 200D audio signal generator is 18 volts. An amplitude of about ten volts on the grid of V1a will cause the time of each stable state to be the same, and hence, the voltage pulses appearing on the plates of V2a and V2b will be evenly spaced.

The output of the multivibrator taken from the plate of V2b is fed through two shaping stages, V1b and V3b. The square wave appearing on the plate of V3b is placed on the grids of all of the charge tubes. A smaller amplitude square wave is fed to the grid of the phase inverter V3a from a 25K tapped resistor in the plate circuit of V3b. The output of the phase inverter is placed on the grids of all of the discharge tubes. Hence, when the charge tubes are turned on, that is, when the voltage is increasing in the positive direction, the discharge tubes will be turned off and the cell capacitance will charge to the voltage Y. During the other half of the square wave cycle, when the charge tubes are turned off by a negative cut-off voltage, the discharge tubes are turned on and the cell capacitance is discharged.

The magnitude of the square wave appearing across the Stark cell depends upon the voltage applied at Y and the setting of the 30K potentiometer in the plate of V3b. This setting should be made with 500 volts applied at Y. The control should be varied until a further increase in the square wave voltage appearing on the grid of V3b does not increase the magnitude of the square wave voltage appearing across the cell. Once the control is set in this way no further adjustment over the entire range of the voltage applied at Y is necessary.

The B + for the first three stages applied at Z emanates from a regulated power supply which is included as part of the spectrograph. The variable voltage applied at Y is taken from a high voltage power supply located in the generator room. The output was made variable by connecting the primary of the high voltage transformer to the output of a Variac. The magnitude of the voltage applied at Y should not exceed 1200 volts and should not be kept at this upper limit for long periods of time.

It is advisable to monitor the square wave appearing across the cell by connecting an oscilloscope at J_2. With high voltages and low cell pressures, arcing and corona effects sometimes occur within the cell. By monitoring the square wave the operator can tell when this is happening and reduce the magnitude of the square wave.

Good waveforms are produced over a frequency range of from 30 to 5000 cycles. Above 5000 cycles the waveform becomes distorted. The frequency selected for this work was 2900 cycles. One reason for this selection was that the twin T in the tuned amplifier can be very sharply tuned to this frequency.

F. Tuned Amplifier

The tuned amplifier is designed to amplify the portion of the energy which it receives from the detector crystal that is Stark modulated at one particular frequency and reject all other frequencies. A diagram of the amplifier is shown in Figure 13.

The detector crystal is connected directly between grid and ground at J_1 in the first stage. The first two stages are conventional R C coupled amplifiers, each having a gain of approximately 90. There is a gain control in the grid circuit of the 6SJ7 stage. The first two stages may be used independently by taking the output from J_2. Such an arrangement is necessary when the mode is being monitored during crystal doubler adjustments and more gain is needed than can be obtained from the oscilloscope amplifiers alone.

The third stage, a 6J6, is connected as a cathode follower, and its only function is to match the low input impedance of the high pass filter.

The high pass filter has no attenuation at 3000 cycles, begins to attenuate at 2300 cycles, and increases rapidly to an attenuation of 10^5 at 100 cycles.[10] The contour of the klystron mode, as well as the Stark modulation components, have been amplified up to the input of the filter. The contour of the mode is composed primarily of lower frequency components and is therefore eliminated, whereas the Stark modulation components are not attenuated.

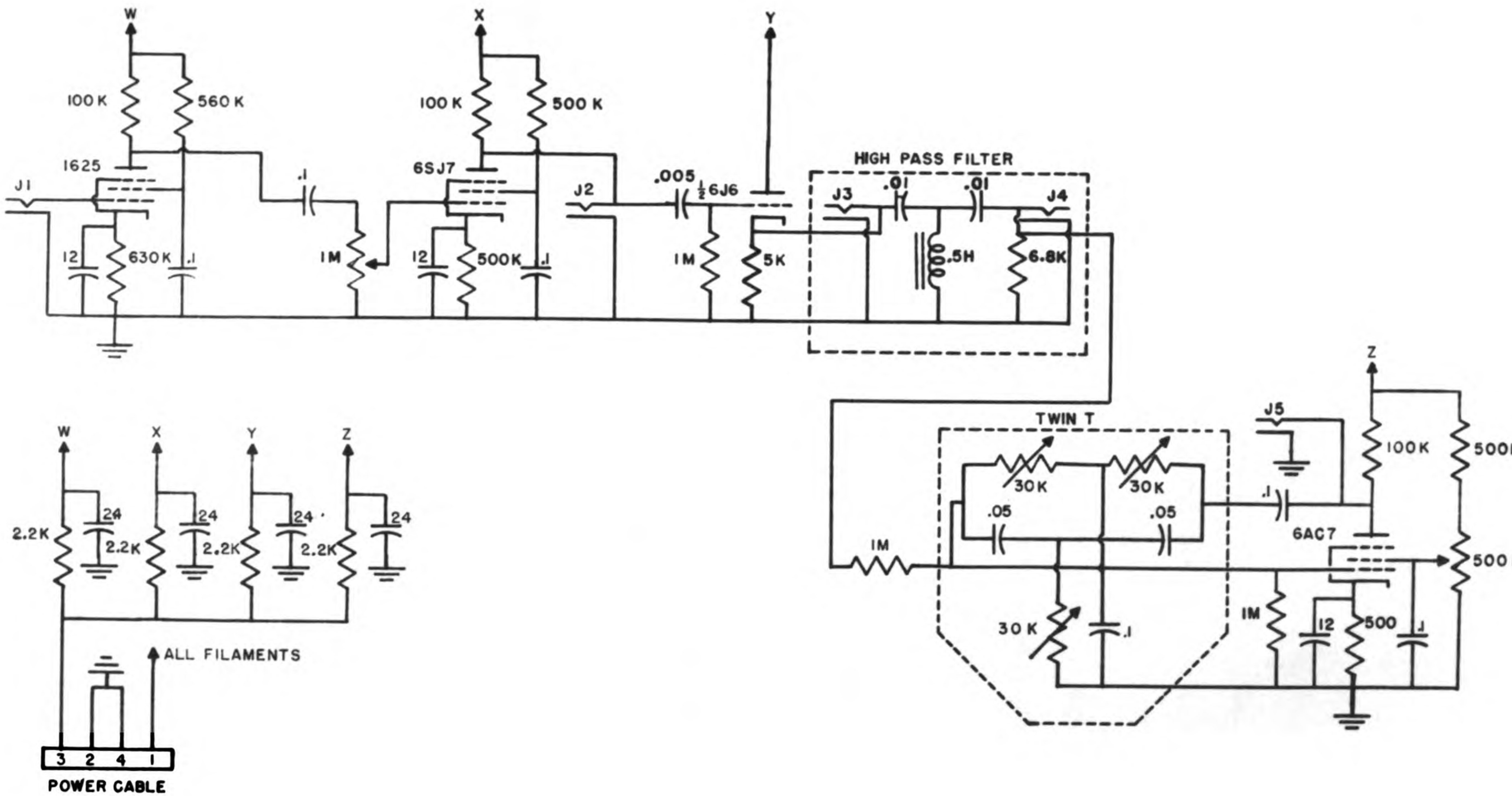


Figure 13. Tuned Amplifier

The jack J_4 allows the signal to be removed directly after it has passed through the high pass filter. This arrangement is desirable when source modulation or straight absorption methods are used. The sharp dip in the mode due to molecular absorption of energy in the straight absorption scheme carries higher frequency components than a dip caused by reflections and standing waves. Hence, the filter will attenuate the unwanted indentations more than those due to molecular absorption. The same situation holds true when the source modulation method is used.

The output of the high pass filter is placed on the grid of the last stage, a 6AC7. A twin T tuned filter is placed between the grid and plate of the 6AC7. The twin T is built in a separate plug-in unit and may be removed from the circuit. When it is not in the circuit, the 6AC7 acts as an ordinary amplifier. When it is in the circuit and tuned to one frequency, the 6AC7 stage is a very sharply tuned amplifier.

A twin T filter will attenuate any one particular frequency to which it is tuned. Since this filter is connected between the control grid and plate of the 6AC7 it will cause degeneration in the circuit as a whole, at all frequencies that it does not attenuate. Hence, the output voltage appearing on the plate will be greatest when the input frequency is the same as the attenuation frequency of the twin T.

There is a definite procedure which should be followed in tuning the twin T. First, the resistance of the arms are set roughly with an ohmmeter by using the equations[11] $\omega_o = \frac{1}{R_a C_a}$ where $R_a = R_b = 2R_c$ and $C_a = C_b = \frac{1}{2}C_c$. In this case, $C_a = C_b = .05$ microfarads and $C_c = .1$ microfarads. As was mentioned earlier, a frequency of 2900 cycles will give the best results. In the drawing R_a and R_b are the two horizontal variable resistances and R_c is the vertical arm.

After this first rough setting the attenuation frequency is checked by placing a signal between the control grid and ground of the 6AC7 from an audio signal generator. An audio voltmeter should be connected between the plate and ground. The resistance arms are now adjusted so that maximum attenuation occurs at about 2900 cycles. The vertical arm is the most critical. It should be varied first until the audio voltmeter reads a minimum voltage. The two horizontal arms are then set to further decrease the minimum, but changing the horizontal arms will disturb the setting of the vertical arm and it must be reset. This procedure is carried on until the audio voltmeter reads the lowest possible output voltage.

During the two previous steps in the adjustment all power to the amplifier has, of course, been turned off. As a final check the amplifier should be placed in operation and an audio signal applied between the output of the high pass filter and ground. It is important that this signal be applied so that the one megohm resistor in series with the output of the high pass filter and 6AC7 control grid is always between the control grid and the point where the test signal is applied. The circuit will not function as a tuned amplifier when a signal from a low impedance source is placed directly on the 6AC7 control grid. With the audio voltmeter again connected between plate and ground the test signal should be momentarily removed, and the screen grid bias control advanced until the circuit breaks into oscillation. The control should then be backed down to a point just before oscillation begins. This setting will yield the maximum amount of gain and selectivity. The test signal should now be applied and the signal generator varied over a range of a few hundred cycles to see at what frequency the maximum output occurs. It should occur within 100 cycles of the pre-set 2900 cycle frequency. Actually, it is not important that the circuit be tuned to one particular frequency. The important thing is that it be extremely selective and it has been found that the most selective region is near 2900 cycles. The signal generator should now be set to yield maximum output and the arms of the twin T again adjusted until the very maximum output is obtained at this one frequency. Figure 14 is a typical response curve, relating input frequency with output volts, for the 6AC7 stage.

The over-all gain from the input at J_1 to the output at J_5 is approximately 2000. There is no amplification in the cathode follower circuit and a slight loss in gain in the twin T circuit. However, the tuned amplifier gain plus the gain due to the vertical deflection amplifier of an oscilloscope yields an overall figure on the order of 10^6 and this is more than adequate.

The entire tuned amplifier is battery operated. This does away with the extremely bothersome ripple and line noise which are practically unavoidable when conventional power supplies are used to operate high gain amplifiers having very low level input signals. A six volt storage battery supplies the filament current and is connected between pins one and four on the power cable. A bank of B batteries supplies a 270 volt B+ voltage which is connected at pins two and three. The decoupling filters in series with the B+ connections to each stage help prevent interstage feedback and oscillation.

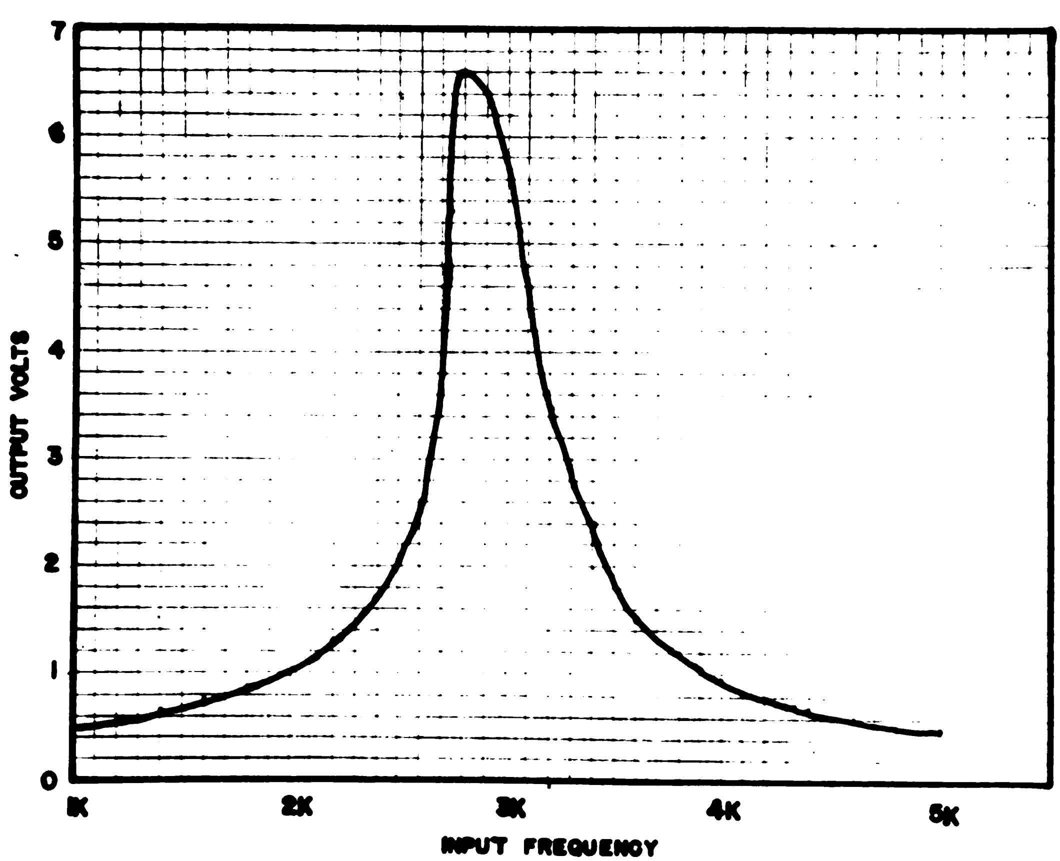


Figure 14. Tuned Amplifier Response

G. Radio Frequency Generator

When source modulation is desired the radio frequency generator is used to place a low amplitude high frequency sine wave on the klystron repeller. A schematic diagram is shown in Figure 15.

The oscillator is a conventional electron coupled type and has a frequency range of approximately eight to nineteen megacycles. A particular frequency is obtained by first tuning the control grid circuit and then tuning the plate circuit to resonance at this frequency. Resonance is indicated by a dip in the plate current meter M_1. Tuning the plate circuit will disturb the original frequency setting slightly and the two circuits must be tuned alternately by decreasing amounts in order to approach the desired frequency.

The oscillator output is coupled to the grid of a 6J5 which is connected as a cathode follower. Control R_4 determines the amplitude of the R. F. voltage on the klystron repeller.

Reference should be made to Figure 10 for the connection at A. The line from the output of the 6J5 to the klystron repeller may carry three different signal elements simultaneously; the constant D. C. repeller voltage, the saw-tooth sweep voltage, and the R. F. voltage. An R. F. choke in series with A isolates the R. F. voltage from the sweep circuits.

When source modulation is used the output of the crystal detector is placed directly on the input of a communications

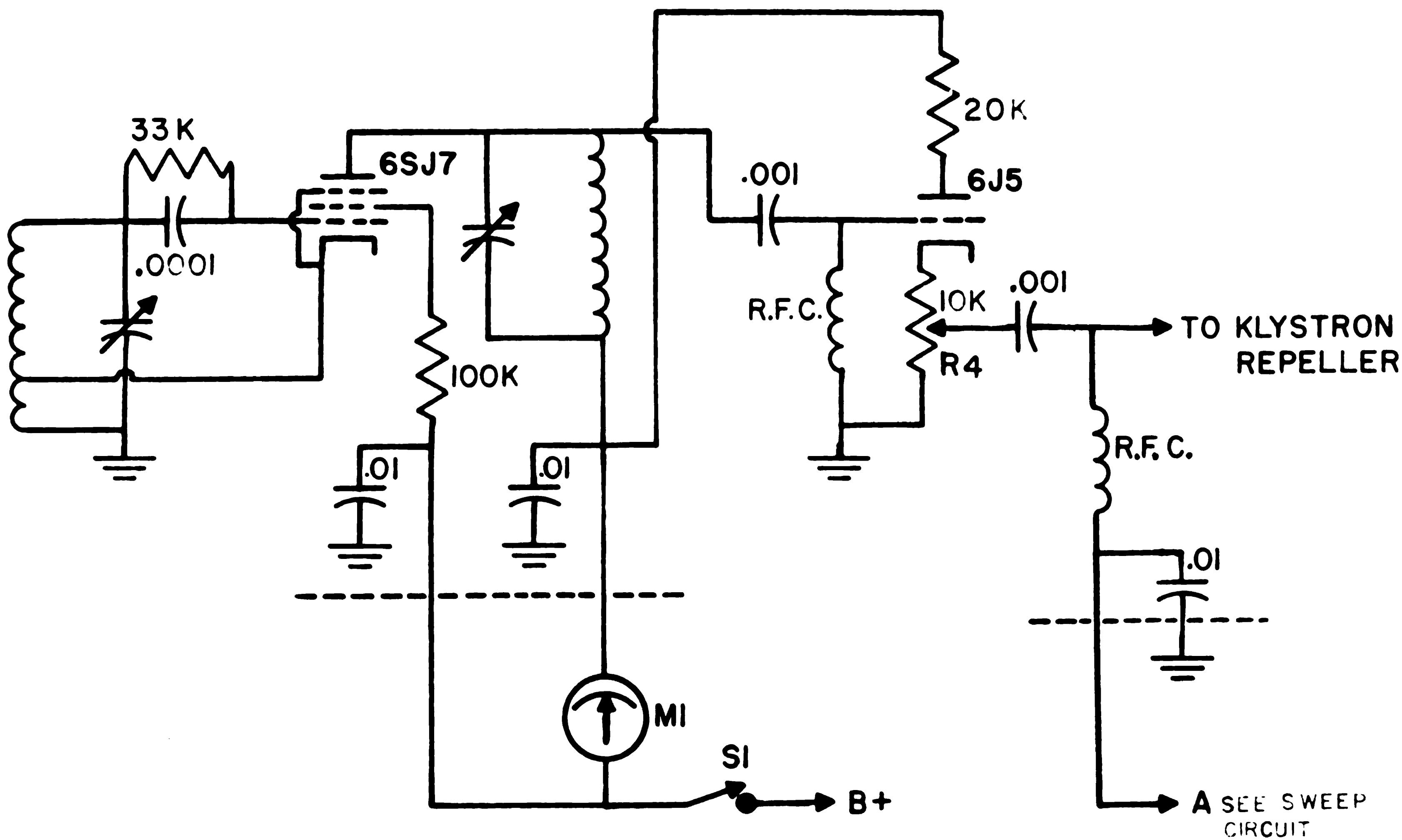


Figure 15. Radio Frequency Generator

receiver tuned to the frequency of the R. F. generator. It is very important that there be no leakage from the R. F. generator which might be picked up directly by the receiver. All possible precautions have been taken to avoid this by completely shielding the unit and all of the R. F. carrying lines.

H. Power Supplies

There are two regulated power supplies included as part of this equipment. The schematic diagrams are shown in Figure 16. Both regulators are the conventional gate type and provide good regulation when the prescribed current ratings are not exceeded.

Supply number one operates the sweep generator and the first few stages of the square wave generator. These circuits were designed to operate with a B + voltage of 340 volts and the 6SJ7 control grid bias should be adjusted so that M_1 reads this voltage. The resistances in series with both M_1 and M_2 are multipliers used to calibrate the meters to read 500 volts at full scale deflection.

In supply number two, two separate transformer, rectifier sections are connected in series and fed to the same regulator. These two transformers have smaller voltage, current ratings than the transformer used in supply number one. The R. F. generator which receives power from this supply was also intended to operate with a B + voltage of about 340 volts.

The two regulators are identical except that the supply

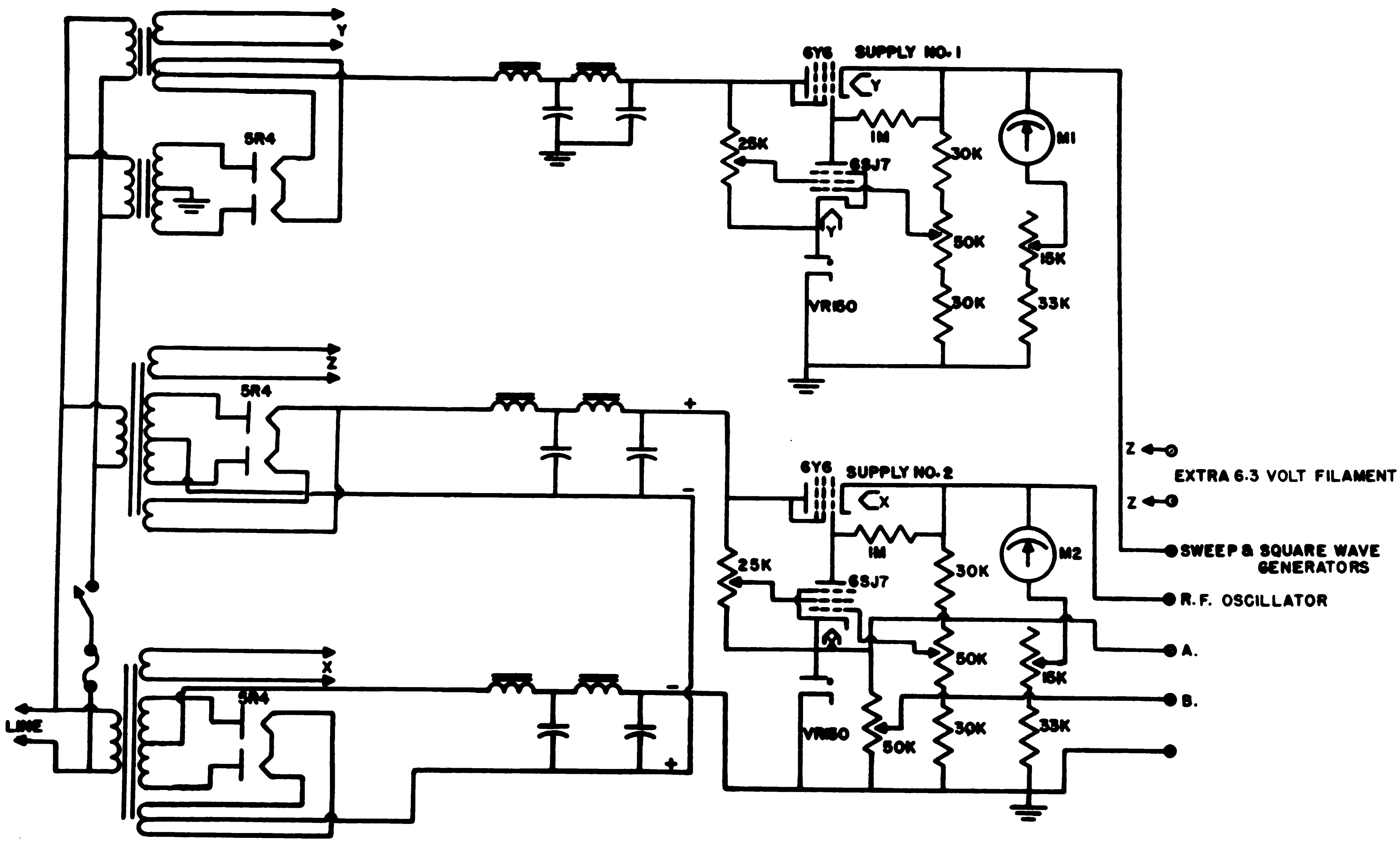


Figure 16. Power Supplies

number two regulator has two extra outputs at A and B taken across the VR150. This supply may be used to operate a Western Electric type 723A klystron or klystrons having similar voltage and current ratings. If such an arrangement is desired, the B- ground connection should be removed and the B + grounded to the chassis. The connection at A is made to the klystron cathode and the variable voltage at B placed on the klystron repeller.

The voltage and current ratings of the two power supplies are as follows:

Supply Number One:

at 400 volts do not exceed 30 milliamperes
at 300 volts " " " 70 "

Supply Number Two:

at 500 volts do not exceed 65 milliamperes
at 400 volts " " " 95 "

Sufficient accuracy may be obtained for intermediate values by interpolating between these end points.

IV. OPERATING PROCEDURE

The tuned amplifier and communications receiver may be seen in Figure 6. The rest of the electronic apparatus is shown in Figure 17. The rack and panel assembly on the far right contains the two regulated power supplies, the sweep circuits, and the R. F. generator. The other units from left to right are as follows: klystron power supply, square wave generator, a Variac to control the square wave amplitude, mode monitoring oscilloscope, audio signal generator, square wave monitoring oscilloscope, and presentation oscilloscope. There is a definite procedure which should be followed in order to place the equipment in operation.

After a sample has been injected into the absorption cell, the first step is to get the klystron and crystal doubler in operation over the desired frequency range. The klystron filaments are turned on but the operator should wait at least one minute before applying the high voltage to the klystron. It is very important that the anode voltage be set at its lowest point when the high voltage is applied, and then advanced to the desired operating point. This allows time for the two microfarad coupling condenser in the sweep circuit to charge slowly. If the high voltage is turned on when the anode voltage control is in its normal operating position, the sudden charging current of the condenser may damage the klystron.

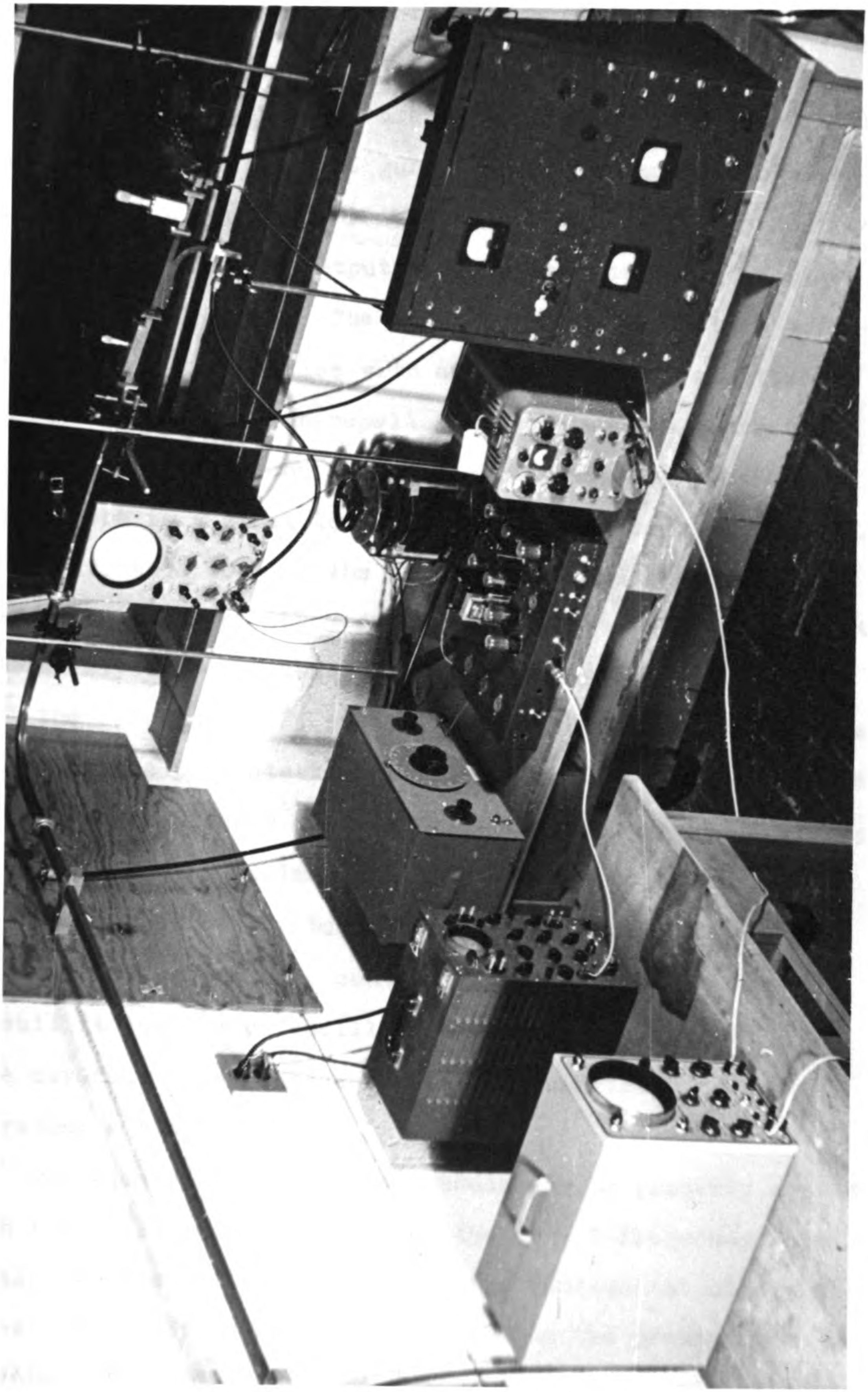


Figure 17. Electronic Components

All of the components are now placed in operation, with the exception of the R. F. generator and communications receiver which are used only when source modulation is desired. The meters indicating output voltages of the power supplies should read 340 volts. The sweep generator is set at a frequency of about 70 cycles with a maximum output amplitude. When the D. C. klystron repeller voltage is set correctly, the mode will appear on the mode monitoring oscilloscope. Since there is a great loss of power in the crystal doubler, it is advisable to pick the highest amplitude mode. The frequency limits of the mode can be checked with the wavemeter and the klystron frequency set accordingly.

The output from J_4 of the tuned amplifier should now be placed on the presentation oscilloscope. The crystal plunger and slide screw tuner must be adjusted until proper doubling is indicated by a maximum undistorted mode contour.

The output of the tuned amplifier is now changed to J_5 and the 6AC7 screen bias control advanced until the twin T circuit is just below oscillation. This setting should be made carefully and rechecked after the amplifier has been operating a short time.

The square wave generator should now be properly adjusted. With the audio oscillator set at the twin T frequency, the high voltage is slowly increased until the fundamental absorption lines and the Stark components appear on the presentation oscilloscope. The audio oscillator should be tuned through a small range of frequencies to make certain it is at a frequency which will yield a maximum amplitude of the spectral lines. The output of the audio oscillator must be set so that the square waves appearing on the square wave monitoring oscilloscope are evenly spaced.

It is advisable to view the spectrum using a very low sweep frequency on the order of two or three cycles per second. There are two reasons why this is necessary. First, as the sweep frequency is increased, the mode contour is composed of higher frequency Fourier components. At higher sweep frequencies there will be relatively high amplitude components which exceed the frequency at which the high pass filter in the tuned amplifier begins attenuation. They will appear on the oscilloscope along with the spectrum and make interpretation difficult. Secondly, the ratio $\frac{\text{square waves/second}}{\text{saw-tooth waves/second}}$ will be greater at lower frequencies. Since the spectrum as viewed on the oscilloscope is actually a modulation envelope, a more detailed outline is presented with greater ratios. A high persistancy screen in the presentation oscilloscope makes interpretation easier at these low sweep frequencies.

As the square wave amplitude is increased from zero volts, the first effect observed is a widening and smearing of the lines. At greater field intensities, the line starts to split up into its Stark components, and at sufficient field strength is entirely resolved. The various components, labeled with the allowed values of the quantum number M, arise because the molecule can assume different positions with respect to the applied field which have various discrete values of energy.

The deviation of a component from the original line frequency is proportional to EM for symmetric rotors and proportional to E^2M^2 for linear molecules, where E is the field in the cell. The intensities of the various components are not all the same, but depend upon transition probabilities as calculated from quantum mechanics. Separate components may be viewed in more detail by decreasing the sweep amplitude and adjusting the klystron repeller voltage until a very small, select range of frequencies containing the component is isolated.

V. TEST SPECTRA

Figures 18, 19, and 20 illustrate typical oscilloscope patterns of fundamental absorption lines and Stark components. All figures are of the $J = 7$, $K = 4$ line of the inversion spectrum of ammonia which has a frequency of 19,218.52 megacycles. The pressure is a constant at 50 microns and the only variables are the field intensity and the end points of the frequency range. The range of frequencies covered in the trace is approximately 40 megacycles.

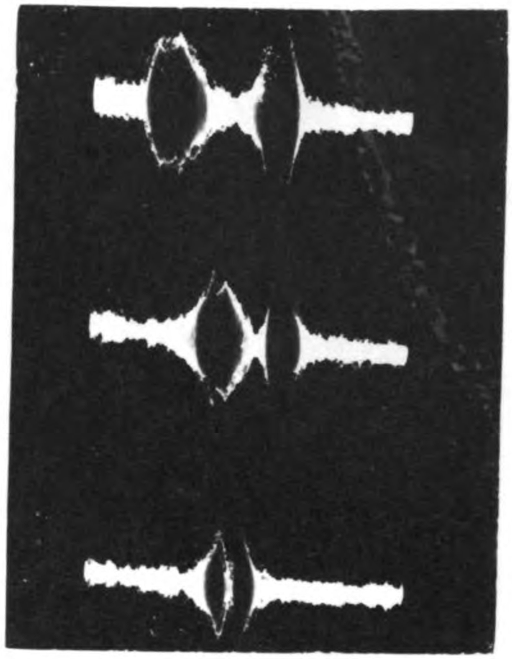
In Figure 18A, the square wave amplitude is 50 volts, and the line is just beginning to split. The contour on the right is the fundamental line, and the outline on the left is a contour of all of the Stark components. In 18B, the square wave amplitude has been increased to 100 volts and Stark components have moved to the left and isolated the stationary fundamental line. In 18C, the square wave amplitude is 150 volts, and the Stark components have moved still further to the left, but there is no clear defining line between any of the individual components.

In Figure 19A, the square wave amplitude is increased to 200 volts, and the Stark components are beginning to split up into two separate outlines. In 19B, with an amplitude of 250 volts, the left hand outline in 19A has moved off the frequency axis and the right hand outline has broken up into two separate envelopes. In 19C, with an amplitude of 300 volts, the Stark components are separated still further.

In Figures 20A, B, and C, the square wave amplitudes are 400, 500, and 600 volts respectively, and the Stark components are completely removed from the frequency axis. At these higher voltages, the individual components may become completely separated and the operator can shift the frequency axis and view them individually.

Figures 22 and 23 are of the same ammonia line but at a pressure of about 140 microns. The signal to noise ratio is greater at this lower pressure, but the line is much broader and it is difficult to distinguish between individual Stark components. In 23A, the square wave amplitude is 50 volts, and the line is just beginning to split. In 23B, the Stark voltage is 500 volts, and in 23C it has been increased to 800 volts. Figures 22A, B, and C show some of the Stark components that were off the frequency axis when 23B was taken.

Figures 21A and B are typical oscilloscope patterns when the source modulation method is used. Both are of the $J=7$, $K=4$ ammonia line, but Figure 21A was taken with a cell pressure of 140 microns while 21B was taken at a lower pressure of about 60 microns.

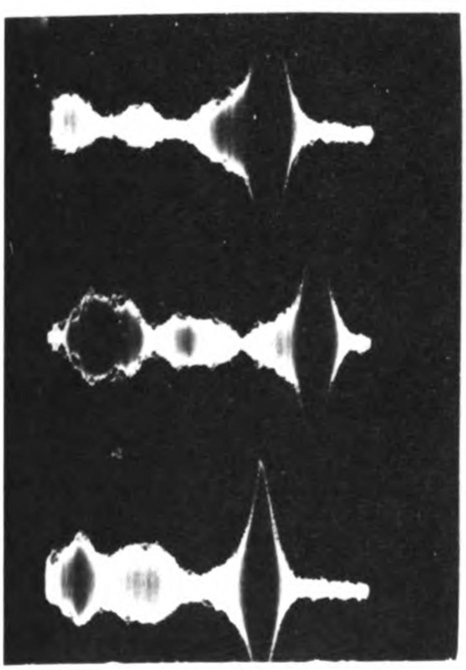


C

B

A

Figure 18

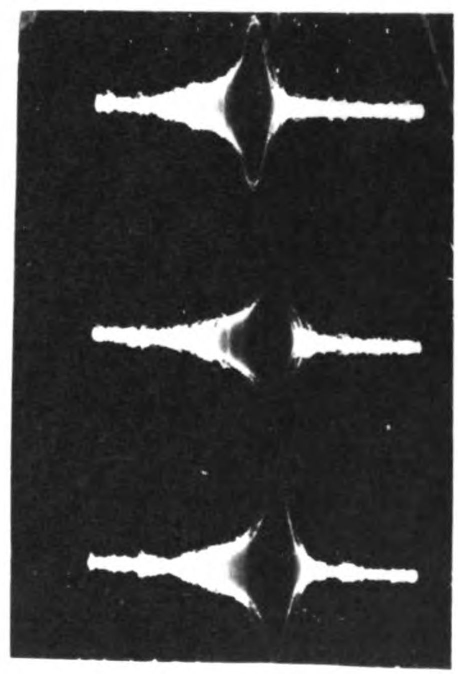


C

B

A

Figure 19



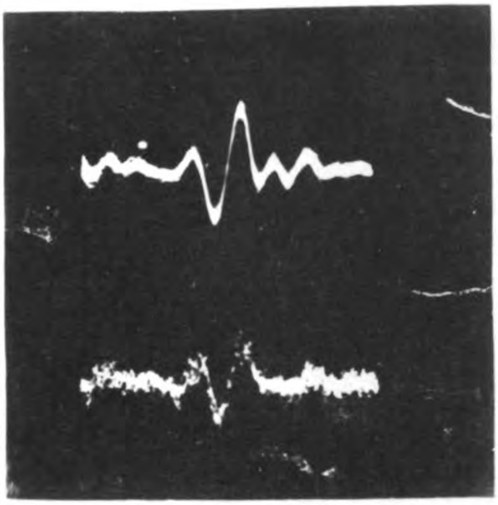
C

B

A

Figure 20

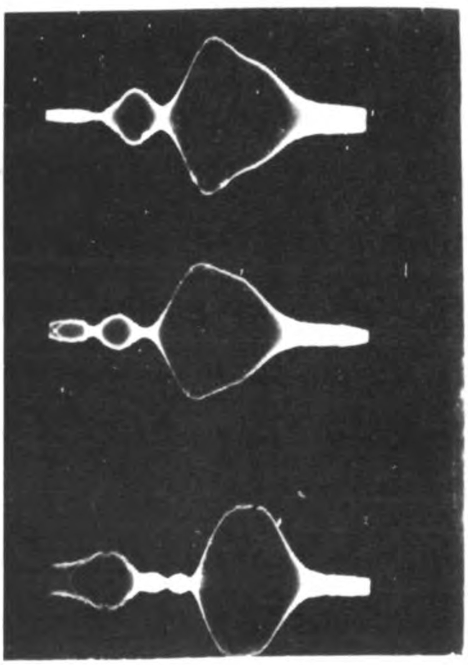
Test Spectra



A

B

Figure 21

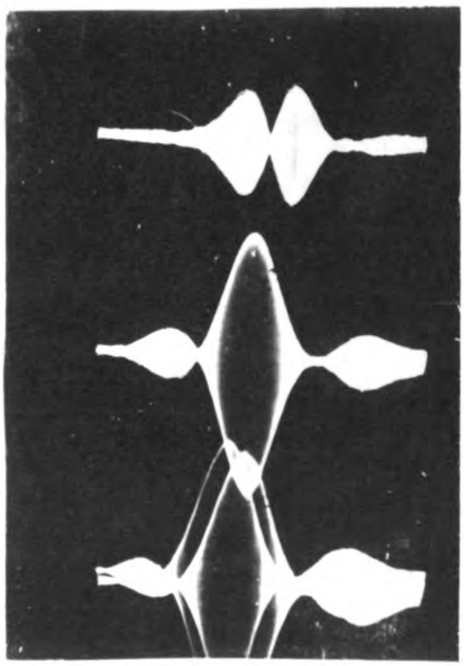


A

B

C

Figure 22



A

B

C

Figure 23

Test Spectra

SUMMARY

A Stark modulated microwave spectrograph has been designed and constructed. The specific units which were constructed are: a vacuum system, Stark absorption cell, crystal doubler, sweep generator, radio frequency generator, square wave generator, tuned amplifier, and two regulated power supplies.

The equipment is flexible in that the operator may choose either the direct absorption, Stark modulation, source modulation, or source plus Stark modulation method of viewing molecular spectra.

Each of these methods has its merits. The direct absorption spectrograph is very simple to operate but lacks sensitivity and provides no solution to the troublesome problem of standing wave interference. The source modulation spectrograph has greater sensitivity and much of the standing wave interference may be eliminated by a high pass filter. Source modulation is advantageous when a simple spectrum of only the fundamental absorption lines is desired. The Stark modulation method has good sensitivity and the interference problem is completely eliminated by employing a tuned amplifier. The additional information obtained from the Stark components of the spectra is an aid in computing structural paramaters of molecules. When source plus Stark modulation is used, a high degree of sensitivity is possible, but a larger number of

components must be kept in very good adjustment. For the great majority of applications, the Stark modulated spectrograph has adequate sensitivity and will yield the best overall results.

A comparison of test spectra taken of ammonia with similar Stark modulation spectra taken by other investigators indicates that the sensitivity and resolution of this spectrograph compares favorably with those generally in use.

BIBLIOGRAPHY

1. Cleeton, C. E. and Williams, N. H., Physical Review 45, 234 (1934)

2. White, H. E., "Introduction to Atomic Spectra" 401

3. Sharbaugh, A. H., Review of Scientific Instruments 21, 120 (1950)

4. Karplus, R., Physical Review 73, 1027 (1948)

5. Gordy, W. and Kessler, M., Physical Review 72, 644 (1947)

6. Hershberger, W. D., Journal of Applied Physics 19, 411 (1948)

7. Gordy, W., Reviews of Modern Physics 20, 668 (1948)

8. "Molecular Microwave Spectra Tables", Bureau of Standards Research paper, R P 2107, 44 (1950)

9. Good, W. F., Physical Review Nos. 3, 4, 213-218 (1946)

10. Terman, F. E. "Radio Engineers Handbook" 234

11. Zeluff, V. and Markus, J. "Electronics Manual for Radio Engineers" 197

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