An ultra-centrifuge for the measurement of adhesion and tensile strength

AN ULTRA-CENTRIFUGE FOR THE MEASUREMENT OF ADHESION AND TENSILE STRENGTH

RICHARD ROY HILL

AN ULTRA-CENTRIFUGE FOR THE MEASUREMENT OF ADHESION AND TENSILE STRENGTH

BY

RICHARD ROY HILL

A THESIS

SUBMITTED TO THE GRADUATE SCHOOL

IN PARTIAL FULFILLMENT OF THE REQUIREMENTS

FOR THE DEGREE OF MASTER OF SCIENCES

DEPARTMENT OF PHYSICS

THE UNIVERSITY OF WICHITA

WICHITA, KANSAS

AUGUST, 1960

CONTENTS

ABSTRACT

Adhesion and tensile strength measurements may be made by spinning rotors plated with thin films until the centrifugal fields produced cause the films to fly off of the rotors. The construction of a vacuum ultra-centrifuge for producing high centrifugal fields and techniques of its use in the measurement of adhesion are discussed. Some preliminary results of measurements of adhesion between thin copper films and steel rotors are also included.

CHAPTER I

INTRODUCTION

The measurement of adhesion and tensile strength by spinning rotors plated with thin films to high rotational speeds was made possible with the development by J. W. Beams and others of a vacuum ultracentrifuge in which the rotor is magnetically supported.[1] At present much work is being done on the measurement of the tensile strength of very thin metal films. Using the high speed rotor technique, the tensile strength of thin silver films has been measured.[2,3] These measurements show an approximately constant value of about the bulk strength of silver for films thicker than 6×10^{-5} centimeters, and for films thinner than 2×10^{-5} centimeters, the tensile strength is increased by as much as tenfold or more. Beams has also shown this increase by another method.[4] However, for very thin films (less than 3×10^{-6} centimeters thick) the centrifuge method of measuring tensile strength has been rendered useless due to the inability to make the adhesion sufficiently small.

For very thin films the equation

$$\omega^2 R^2 d = T + A\frac{R}{h} \qquad \text{(See Appendix I)}$$

is approximately true where; ω is the angular velocity of the rotor, R is the radius of the rotor, d is the density of the film, h is the thickness of the film, T is the tensile stress in the film, and A is the adhesive force per unit area between the film and the rotor. It is obvious from this equation that for very small values of h, the term involving adhesion will be the predominate one unless the adhesion is very, very small. For instance, for films of thickness of the order of 10^{-6} centimeters which are plated upon a one-sixteenth inch diameter rotor, the adhesion would have to be reduced to around 10^5 dynes/square centimeter (about one pound/square inch) for reliable measurements of tensile strengths of the order of 10^{10} dynes/square centimeter to be made. Adding to the adhesion difficulties, for films less than 4×10^{-6} centimeters in thickness, the adhesion seems to increase as the film becomes thinner.[5] Thus, new methods of reducing adhesion need to be found.

To construct a vacuum ultra-centrifuge to be used to find new ways of reducing adhesion and ultimately, by means of these new methods, to measure the tensile strength of very thin metal films has been my purpose. The actual discovery of these methods is to be left to future investigators, although some preliminary work has been done with respect to the adhesion of copper films to steel rotors.

CHAPTER II

THE VACUUM ULTRA-CENTRIFUGE

The vacuum ultra-centrifuge produces high centrifugal fields by spinning small steel rotors to high angular velocities. Very high angular velocities are made possible by using a magnetic bearing to support the rotor and by reducing air drag through the use of a high vacuum rotor chamber.

The Method of Rotor Support

The mechanism which supports the rotor is magnet attraction. If the symmetric magnetic field axis of a solenoid is designated as the z axis and a ferromagnetic object is placed on this axis, then the force acting on such an object is given by the volume integral,

$$F_z = \int_V M_z \frac{\partial H_z}{\partial z}\, dV,$$

where F_z is the z component of the force on the object, M_z is the z component of the magnetic moment of the object, H_z is the z component of the magnetic field of the solenoid, and V is the volume of the object. For the case where the object is small when compared with the solenoid, the force is approximately,

$$F_z = M_z \frac{\partial H_z}{\partial z} V.$$

The value of M is determined from the B-H curve for the ferromagnetic material and the two equations,

$$H_{internal} = H_o - H_{demagnetizing} \quad \text{and} \quad B = H_{internal} + 4\pi M$$

where $H_{demagnetizing}$ is proportional M and H_o is the applied field at the center of the object. If the object has high permeability and is well below its saturation region, $H_{internal}$ will be small and $H_o = H_{demagnetizing}$ (approximately). Now, $H_{demagnetizing} = KM$, where K depends upon the object's shape; then the force equation becomes

$$F_z = \frac{H_o}{K} \frac{\partial H_z}{\partial z} V.$$

Since both H_o and $\frac{\partial H_z}{\partial z}$ are proportional to the magnet current, the force may also be represented by $F_z = CI^2$, where C is a constant determined by the geometry of the whole system. With the z axis taken to be the vertical axis, the condition for support of the object is $F_z = mg$, where m is the mass of the object. Thus, the force on the supported object is proportional to the square of the current flowing through the solenoid and this force must be equal to the weight of the object for the object to remain stationary.

It should be noted however that the condition where the magnetic force is equal to the weight of the rotor is one of unstable equilibrium. If the rotor moves either up or down the forces will be unequal in such a way that the initial motion of the rotor is increased. In order to support the rotor in a stable manner it is necessary to detect the position of the rotor and vary the magnet current so that if the rotor moves upward the magnet current will decrease while if it moves downward the magnet current will increase. A servo system is provided to control the current in this manner. A block diagram of this servo system is shown in Fig. 1. This system consists of a sensor to locate the vertical position of the rotor, a servo amplifier to give correction signals, and a magnet driver to supply current to the solenoid.

The position sensor is a coil of thirty turns of number 28 magnet wire wound with an average diameter of three-eights of an inch. This coil is connected to the grid circuit of a partially neutralized tuned-grid, tuned-plate oscillator operating in the 5 megacycle/second

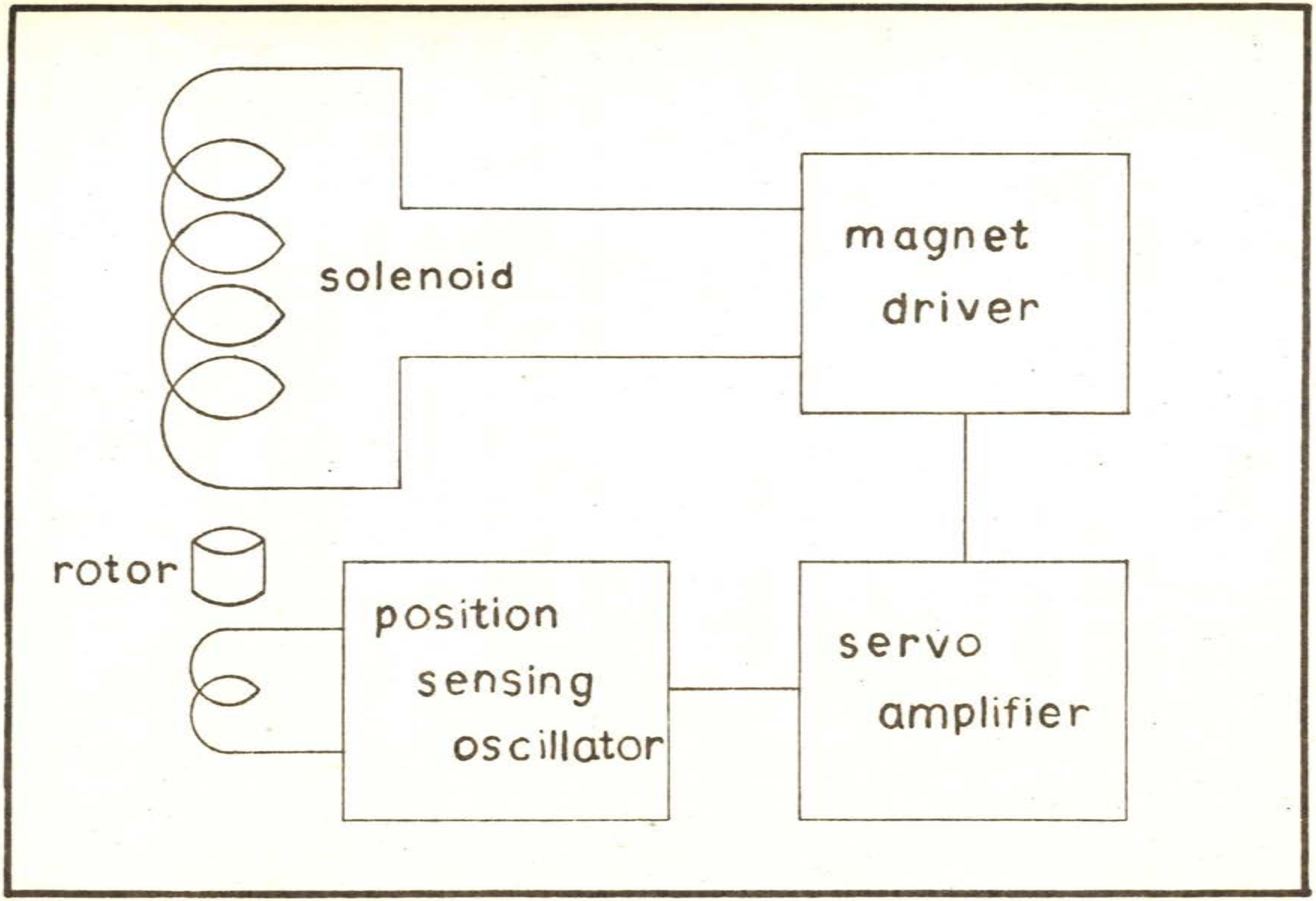


Fig. 1. A Block Diagram of the Support Servo System

range.[6] The output of this oscillator is detected by an infinite impedance detector to provide a direct current signal for the servo amplifier. The circuit is shown in Fig. 2.

The position of the rotor is sensed as follows. First, with the rotor at its closest proximity to the grid coil, the grid and plate tanks are tuned for maximum output with the neutralizing capacitor at minimum capacitance. When this condition exists, upward movement of the rotor increases the Q of the grid coil which results in an increase in the amplitude of the oscillations. When detected, this signal can be used to reduce the solenoid current and restore the rotor to its original position. The vertical position of the rotor is most easily adjusted by varying the neutralizing capacitor. As the neutralizing capacitance is increased, the amplitude of the oscillations is decreased as if the rotor had moved closer to the coil. This results in a correction which raises the rotor until the original amplitude of oscillation is restored. A great advantage of this type of position control is that if the rotor should escape laterally from the supporting field, the amplitude of the oscillations increases enough to cut off the solenoid current thereby protecting the solenoid from overloads.

The servo amplifier is shown in Fig. 3. It was originally designed by Breazeale for a photo-diode sensor, however it proved satisfactory when used with the oscillator sensor. The basic circuit is a cascade of three operational amplifiers of Philbrick type K2-X with their positive inputs grounded. When used in this manner, the operational amplifiers maintain their negative inputs at zero potential through feedback. Thus, to prevent overload of the first amplifier, a combination of fixed and variable bias is used to cancel most of the

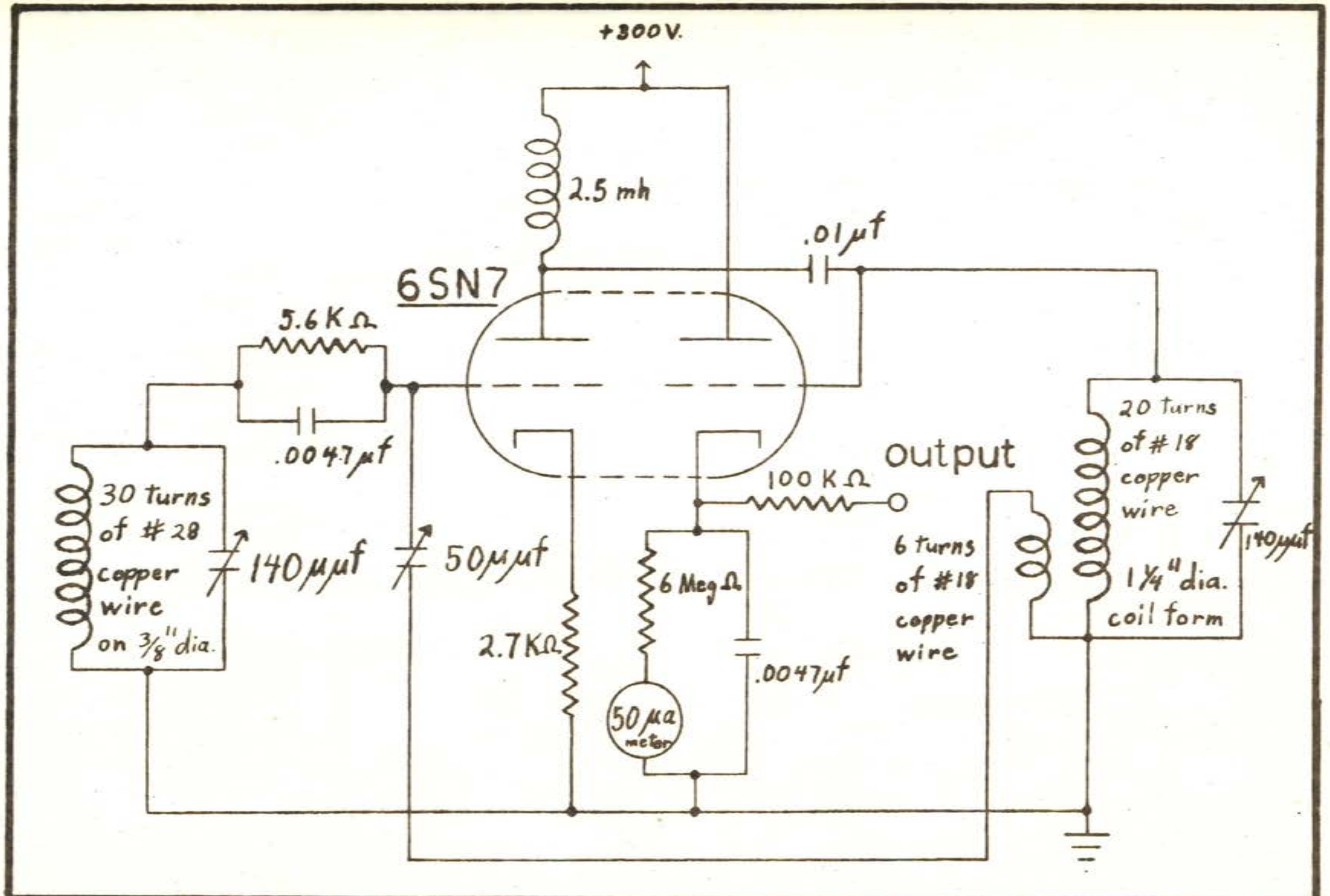


Fig. 2. The Position Sensing Oscillator Circuit

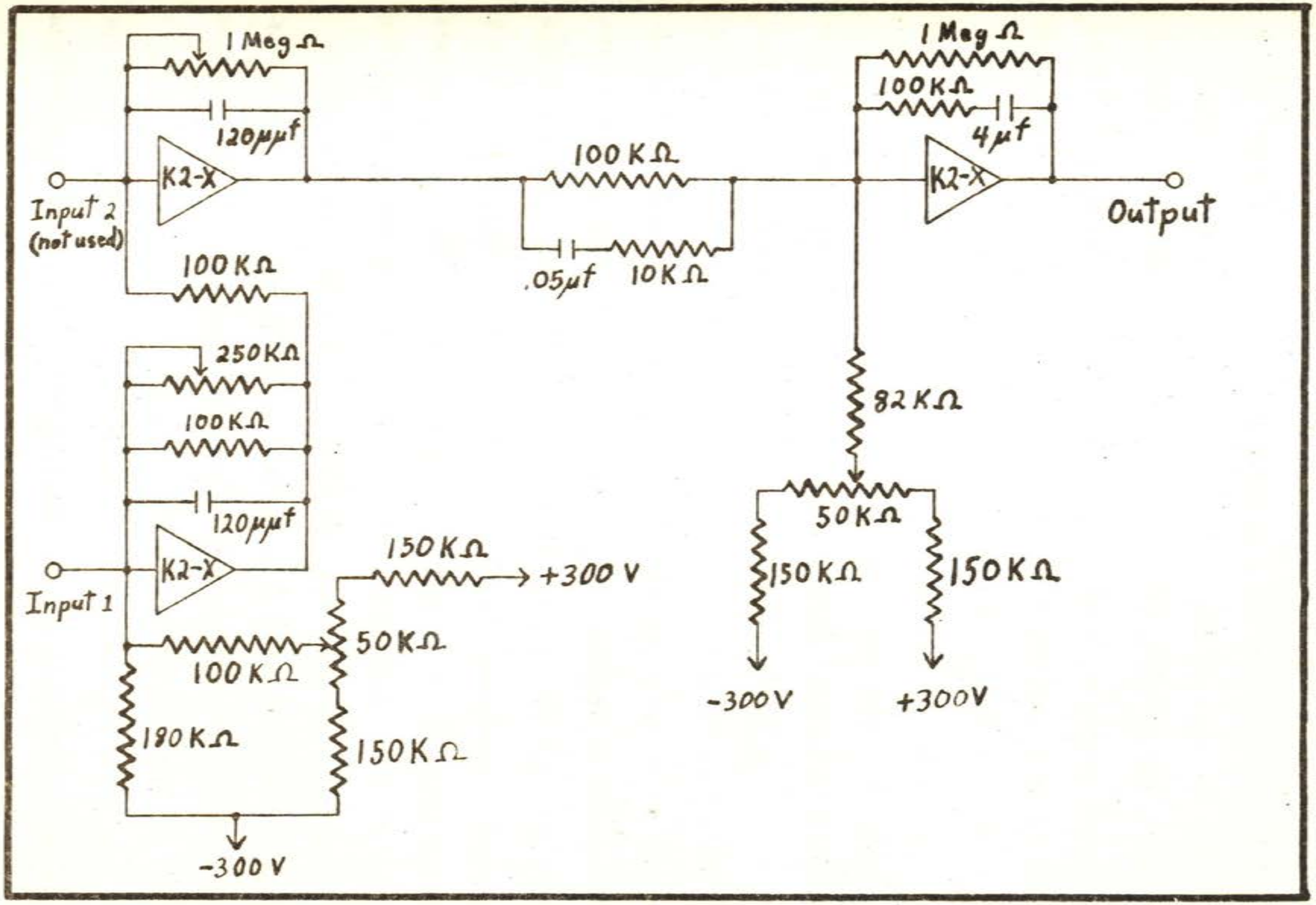


Fig. 3. The Servo Amplifier Circuit

signal coming from the sensor. The variable gain control shown is not used with the oscillator sensor and is set at maximum. The gain control on the second amplifier is increased to a value which is just below the point where over-correction begins. The variable bias for the third amplifier is set so that the solenoid current will be cut off when the sensor puts out its maximum signal. When these adjustments have been completed, no further change is required for the same type of rotor. Also, circuits are included which provide phase lead and damping to prevent high and low frequency vertical oscillation of the rotor.

The output of the servo amplifier feeds the magnet driver (see Fig. 4). The magnet driver uses an operational amplifier (Philbrick type K2-W) to control two 807 power pentodes in parallel. Since the necessary signal inversion was performed by the servo amplifier, the signal is applied at the non-inverting positive input of the operational amplifier. To increase stability, negative feedback is incorporated by applying part of the voltage drop across the cathode resistor of the 807's to the negative input of the operational amplifier. The plate current of the 807's drives the solenoid and is normally about 100 milliamperes when a quarter inch rotor is supported.

The magnet itself is a solenoid of twenty thousand turns of number 28 formvar insulated magnet wire wound on a spool five inches in length and one inch in diameter. The ends of the spool are one-half inch thick bakelite, four and one-half inches in diameter, and the central section of the coil form is an aluminum tube, slotted along its length and cemented with epoxy resin to eliminate the transformer effect. Inside this aluminum tube, a five inch length of three quarter inch diameter iron rod with one end rounded to form a magnetic pole is

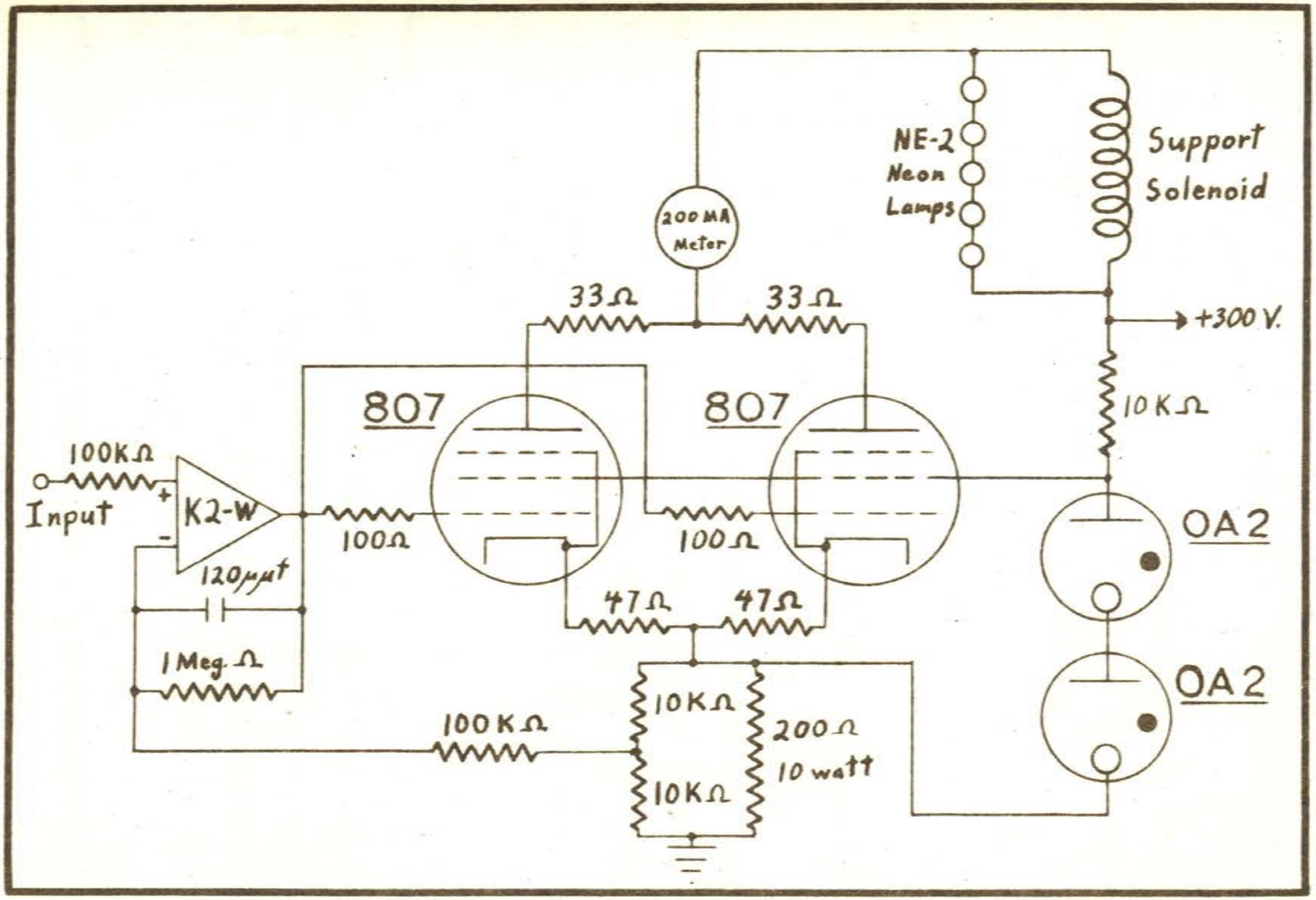


Fig. 4. The Magnet Driver Circuit

mounted rigidly to form the core of the solenoid. The completed solenoid and stand can be seen in Fig. 5. The stand is made from small brass stock and holds the magnet by screws which can adjust the position of the magnet.

As can be seen in Fig. 4 a string of NE-2 neon lamps is placed across the solenoid. These lamps are necessary to prevent high voltage transients from occurring across the coil. Whenever the voltage across the coil reaches a certain value, the lamps conduct and behave like voltage regulator tubes. This starting voltage is several volts higher than the plate supply voltage of 300 volts.

When all controls are properly adjusted, the servo-system will support the rotor several millimeters above the sensing coil and the stability is such that no vertical motion can be observed with the unaided eye. However, though the rotor is at lowest potential when centered laterally in the field, if the rotor is set in motion laterally, the rotor will behave like a pendulum and will oscillate for long periods of time when in a vacuum.

To prevent this oscillation, a damping system is introduced by means of a small steel needle moving in oil (see Fig. 5). The needle is about two inches long and is anchored by a short chain and weight to the bottom of an oil filled beaker. A small cork was slipped on the needle to increase the damping action. The needle is pulled tight on its chain by the support solenoid and due to magnetic coupling, it follows the lateral motion of the rotor. When the coupling between needle and rotor is great enough, the damping is quite effective.

The Method of Accelerating the Rotor

The rotor reaches high angular velocities by being accelerated as

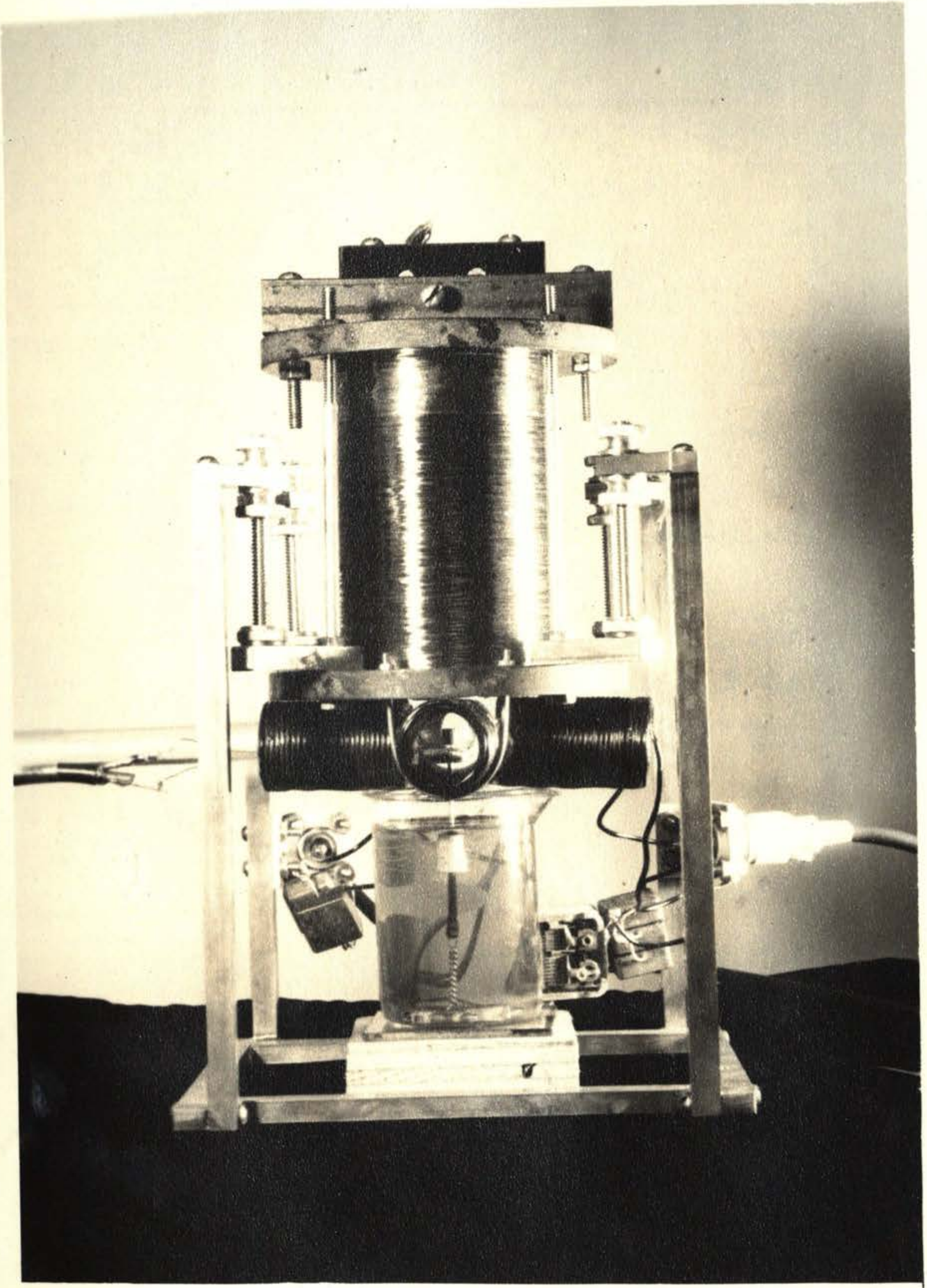


Fig. 5. The Ultra-Centrifuge in Operation

the rotor of a two phase induction motor. The two drive coils (see Fig. 5) of the motor are made of 120 turns of number 14 enameled copper wire wound two layers deep on a one inch diameter. Each coil has a one inch space at its center with half of its turns wound on either side and is made self-supporting by cementing the turns together. The two coils are joined perpendicularly at their centers with their axes in the same plane and are mounted below the solenoid with the open space below the pole of the magnet. Suitable capacitance is placed across each coil to make the resulting tanks resonant at the same frequency of around 130 kilocycles/second.

The two phase current source required to drive this motor is constructed in two sections; the oscillator and phase splitter, and the power amplifier.

A Colpitts variable oscillator (see Fig. 6) operating at a frequency variable about 130 kilocycles/second is used to allow precise tuning to the resonant frequency of the rotor driving tanks. The output is taken from the cathode of the oscillator by a variable potentiometer which feeds the phase splitting bridge. When the resistances of the bridge are made equal to the reactance of the capacitors, the two signals taken from the bridge will be exactly 90 degrees out of phase. This adjustment is made simpler by incorporating an indicating scope into the instrument which exhibits the Lissajous pattern formed by the two signals.

The power amplifier (see Fig. 7) amplifies these two signals separately by means of two separate amplifiers. Each amplifier consists of an 807 power pentode operating Class C which is driven by a 5763 power pentode operating Class A. The resulting two phase current is

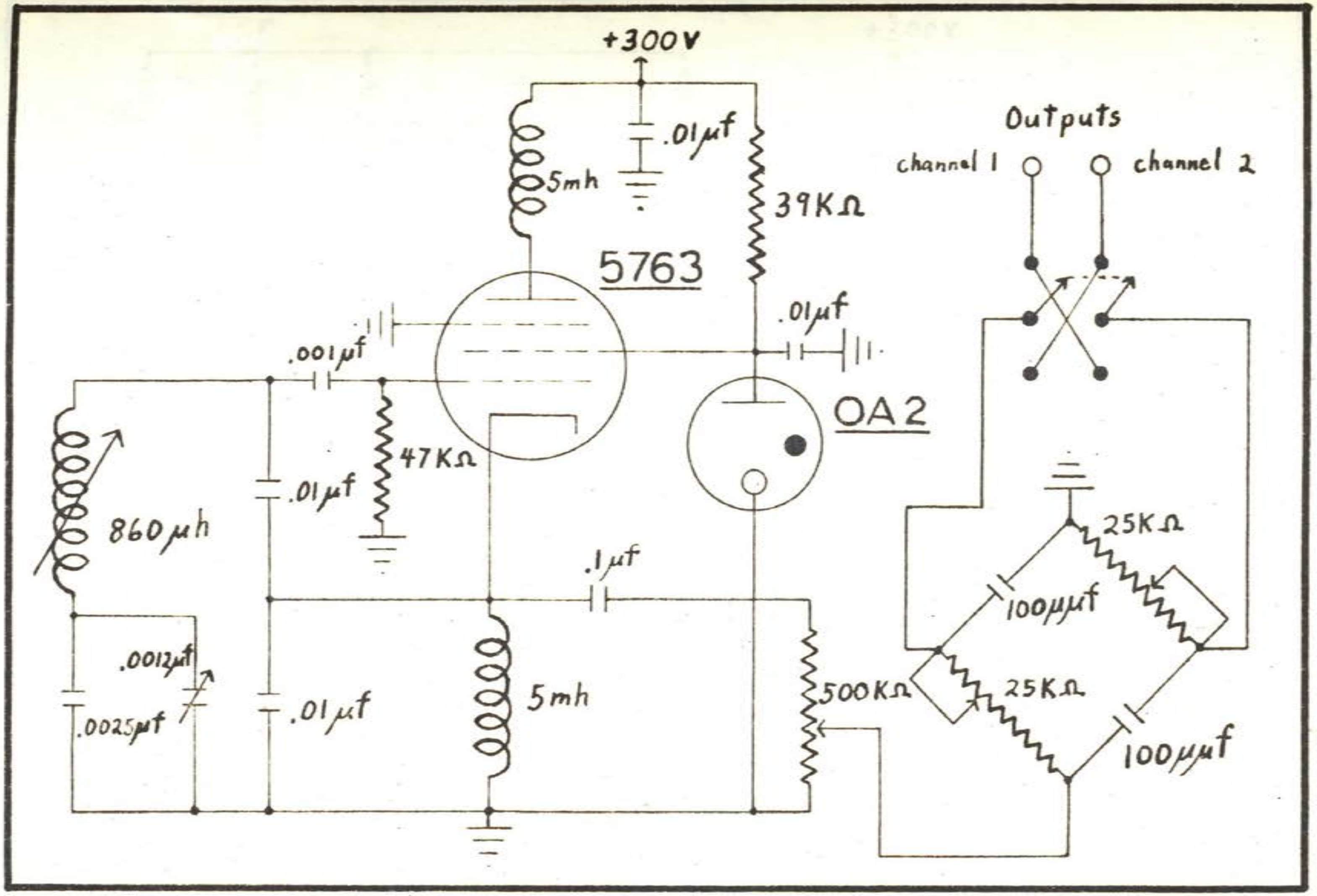


Fig. 6. The Oscillator and Phase Splitter of the Two Phase Current Source

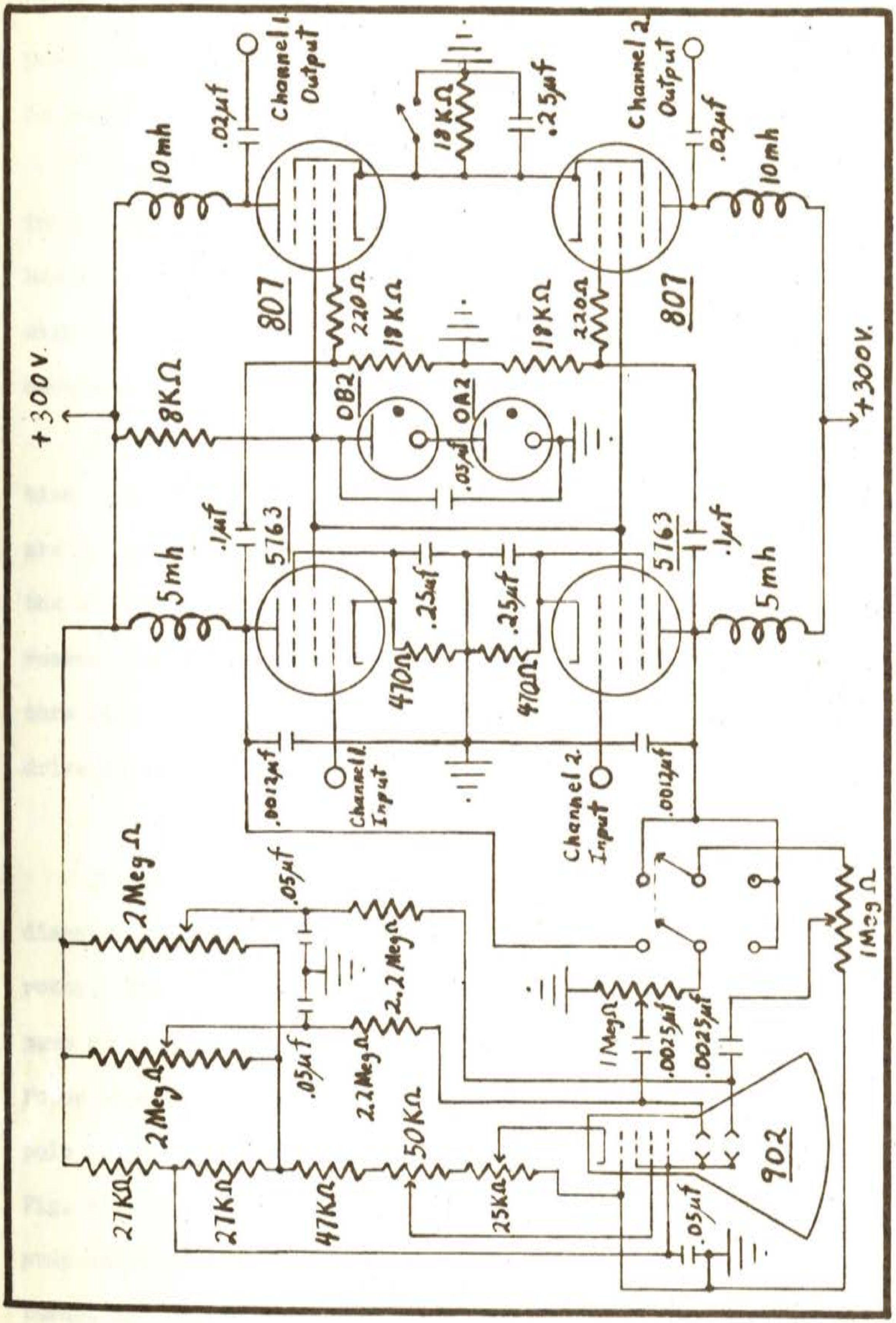


Fig. 7. The Power Amplifier of the Two Phase Current Source

connected to the motor coils. The 5763 also provides the signal to operate the phase relation indicating scope. When operated with a power supply rated 300 volts at 200 milliamperes, each amplifier channel is capable of about 12 watts output.

After the rotor has been accelerated to a certain angular velocity in a vacuum, it will continue to spin for days after the driving field has been removed. For this reason, a switch is used to change over the signals feeding the two amplifying channels so that the rotor can be decelerated in the same manner in which is was accelerated.

The heating of the rotor caused by eddy currents during acceleration is appreciable. After rotors have been driven for over an hour and are brought to rest, opening the vacuum chamber to air pressure causes the surface of the hot rotor to oxidize to a blue color. For this reason, induction driving of the rotor is useful only where a temperature rise can be tolerated. If the acceleration is not too rapid this drive is satisfactory for adhesion experiments.

The Vacuum System

The vacuum chamber for the rotor is a length of 14 millimeter diameter pyrex tubing with a small bubble blown on one end to hold the rotor. The other end is connected to a small silicone oil diffusion pump by means of an O-ring seal and a valve which can be closed if the rotor chamber should burst. The rotor chamber is inserted below the pole of the solenoid through the end of one of the driving coils (see Fig. 5). A thermocouple vacuum gage is mounted between the forepump and the diffusion pump to indicate when the diffusion pump may be turned on. There is, however, no gage on the high vacuum side of the diffusion pump since only a vacuum which allows the rotor to reach the desired angular velocity is necessary. To this extent, the spinning rotor is the vacuum gage for the rotor chamber.

The Construction of Rotors

The rotors were made from nominal quarter inch drill rod. To insure stable rotation about the axis of the rod, the ratio of the length to the radius of each rotor is made slightly less than the square root of three so that the moment of inertia will be greatest about this axis. The rotors are almost cut off in a lathe and left attached to a short length of drill rod. This short length of rod allows the rotor to be chucked in a drill press for polishing. Before polishing, the rotors used were heat treated to a Brinell hardness of 56. The rotors were then polished in a drill press with progressively finer abrasive cloth down to a final polish obtained with crocus cloth. Some of the rotors may be seen in Fig. 10. After polishing, the rotors are broken off from the rod and the end is ground flat.

CHAPTER III

THE EXPERIMENTAL METHOD

To obtain measurements of adhesion with a vacuum ultra-centrifuge, measurements of the various quantities in the equation on page 2 must be made. Also the film must be prepared on the surface of the rotor so that only adhesion is measured.

Measurements

The angular velocity of the rotor is found by reflecting a light off of the rotor and into a photo-multiplier tube (see Fig. 8). Small natural irregularities in the rotor cause a varying periodic signal at the output of the photo-multiplier tube. The period of this signal is the period of the spinning rotor; therefore, if this signal is compared with a signal from a signal generator by observing the Lissajous pattern formed on an oscilloscope, the angular velocity of the rotor may be determined.

The diameter of the rotor is measured with a micrometer caliper and the density of the film is assumed to be the handbook[7] value for the electroplated form of the metal used.

Originally the thickness of the plated film was to be determined by interferometer methods. However, the lack of such an instrument forced the use of a less accurate method in which the thickness is computed by measuring the total charge transferred during plating and relating this to the total amount of copper plated as determined by a sample weighing. By knowing the surface area of the rotor and the density of the plated metal, the thickness of the film can then be calculated. For this method to have reasonable accuracy, the plating bath temperature, the current density, and the plating time for all of the

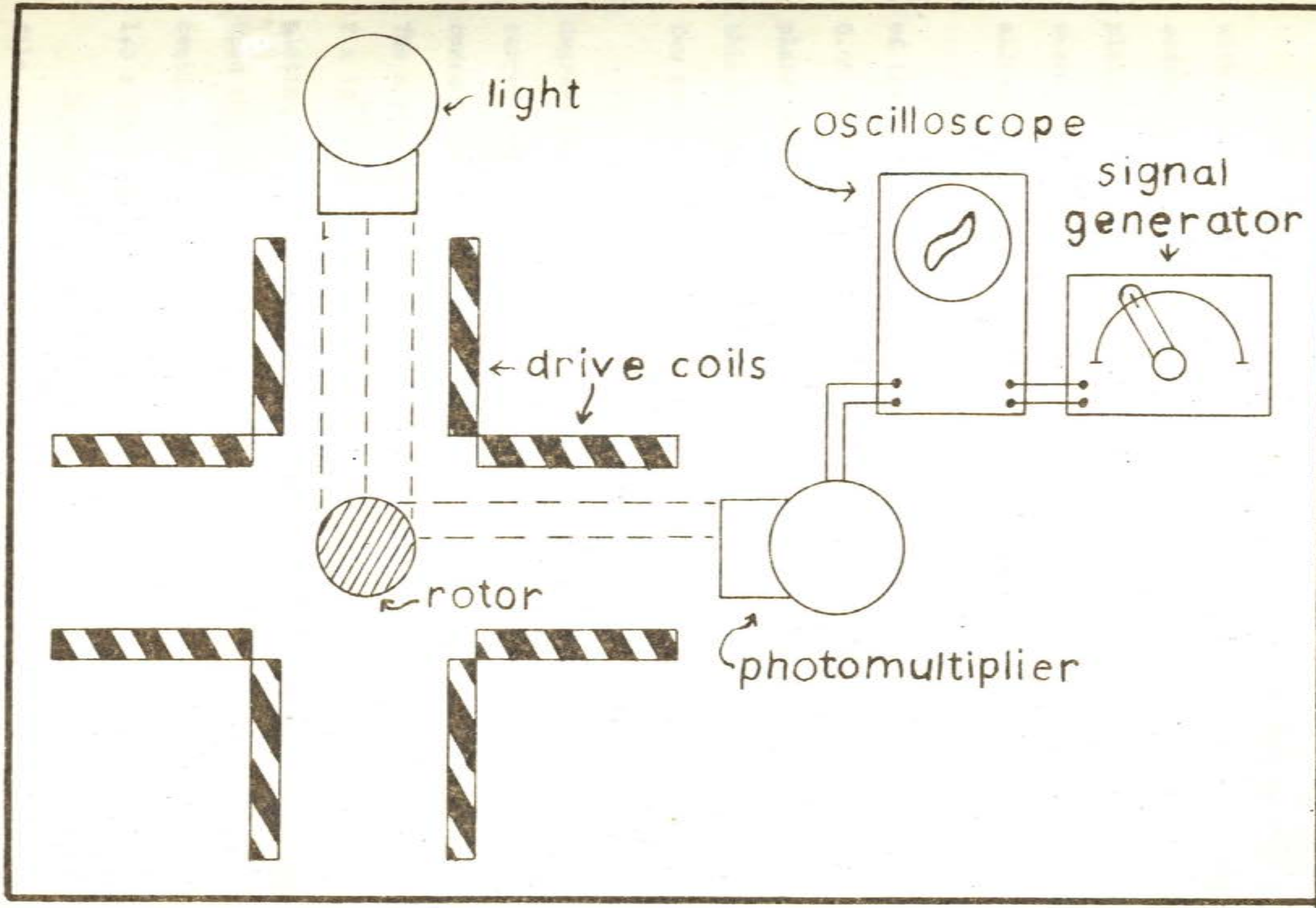


Fig. 8. A Diagram of the Equipment Used to Measure the Angular Velocity of the Rotor

rotors should be held as constant as possible.

Preparation of the Metal Films

Before plating the films onto the rotors, the rotors were washed with acetone, with liquid detergent, and with distilled water in that order. If the surface was to be contaminated to reduce adhesion before plating, the rotor was dipped in the contaminant and any excess was washed off with distilled water; if an oil was used the excess was allowed to drain off.

The rotors were then copper plated in a bath[8] containing chemicals of the following normalities: cuprous cyanide, 0.25 N; sodium cyanide, 0.65 N; and sodium carbonate, 0.30 N. The anode used was a thin copper plate formed into a cylinder. The rotor was supported in the center of this cylinder by a permanent magnet and was agitated by rotating at a few revolutions per minute.

For all films plated, the bath temperature was maintained at 37 degrees Centigrade and the plating time was 37.5 minutes. The plating current used was either 13 or 17 milliamperes which corresponds to a current density at the rotor of about 10 milliamperes/square centimeter. The approximate film thickness expected from charge transfer is about 2×10^{-3} centimeters. However, on the basis of actual weighing, the plating current was found to be 64% efficient. Therefore, depending upon the plating current, the film thickness was about 1.0×10^{-3} centimeters for a plating current of 13 milliamperes or about 1.3×10^{-3} centimeters for a plating current of 17 milliamperes.

In order to eliminate the hoop strength (tensile strength) the film was cut in four places parallel to the axis of the rotor.

Detection of the Film Leaving the Rotor

When a piece of film flies from the rotor, the rotor is left in a disturbed condition due to conservation of momentum and loss of weight. This disturbance can be detected in either of two ways. First, if the piece of film that breaks away is large enough, the oscilloscope will indicate the disturbance as a jumping of the scope pattern. Secondly, if a radio with a beat frequency oscillator is tuned to the position sensing oscillator frequency, the disturbance will be indicated by an abrupt change in the beat tone. The second method is most convenient since it eliminates eye strain.

The complete experimental set-up is shown in Fig. 9. The barricade shown around the rotor chamber and support solenoid is necessary to contain pieces of a rotor if one should explode.

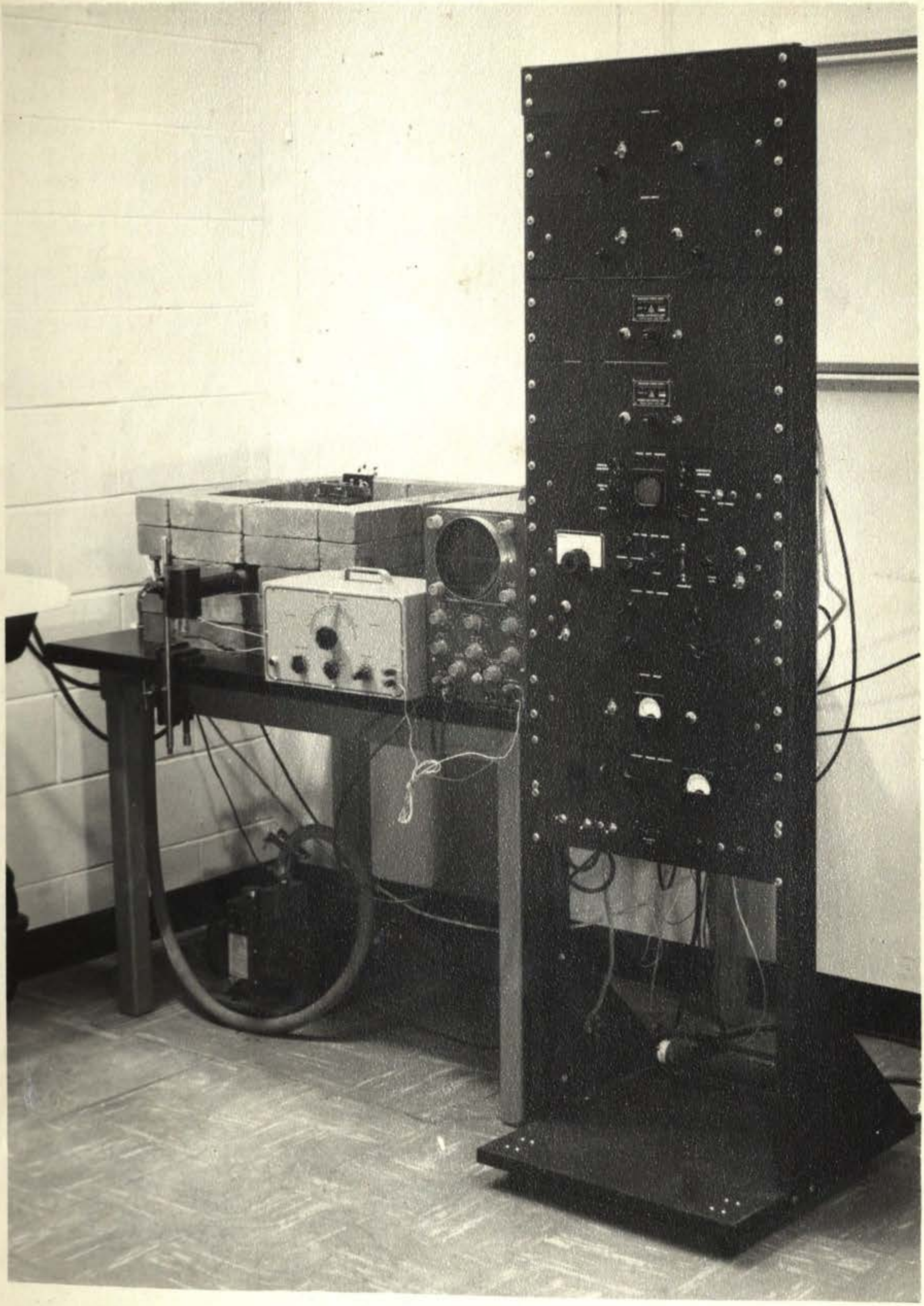


Fig. 9. A General View of the Ultra-Centrifuge and Associated Equipment

CHAPTER IV

RESULTS AND CONCLUSION

None of the few attempts to reduce adhesion between the copper film and the rotor were very successful. Several oils and greases were tried but these either prevented the plating from sticking at all (silicone grease) or did not reduce adhesion enough to allow measurement since the rotor would explode before the film would fly off. As a rule of thumb, the film had to be free enough to be scraped from the rotor with the fingernail before adhesion could be measured using rotors 0.635 centimeter in diameter and of Brinell hardness number 56.

Of the substances tried, only albumin, which had greatly reduced the adhesion between rotors and silver films,[3] reduced adhesion to the point where it could be measured with the ultra-centrifuge. With a rotor which had been dipped into a solution of 0.2 gram/liter of bovine albumin before plating, pieces of film could be observed to fly off at indicated frequencies ranging from 11 to 20 kilocycles/second (20 kilocycles/second was the maximum speed to which any rotor was driven after one rotor exploded at a frequency of 24 kilocycles/second; see Fig. 10). The film removed ranged from one-fourth to two-thirds of the total and the film appeared to have come off in small pieces. One rotor was dipped in filtered egg albumin (not diluted) and it behaved like those treated with bovine albumin except that only about one-tenth of the film was removed. The adhesion values corresponding to 11 to 20 kilocycles/second are $5.5 \times 10^6 - 18.3 \times 10^6$ dynes/square centimeter. Since these values are nearly two orders of magnitude greater than those desired for tensile strength measurements of very thin films, albumin cannot be used to reduce adhesion between copper films and rotors in

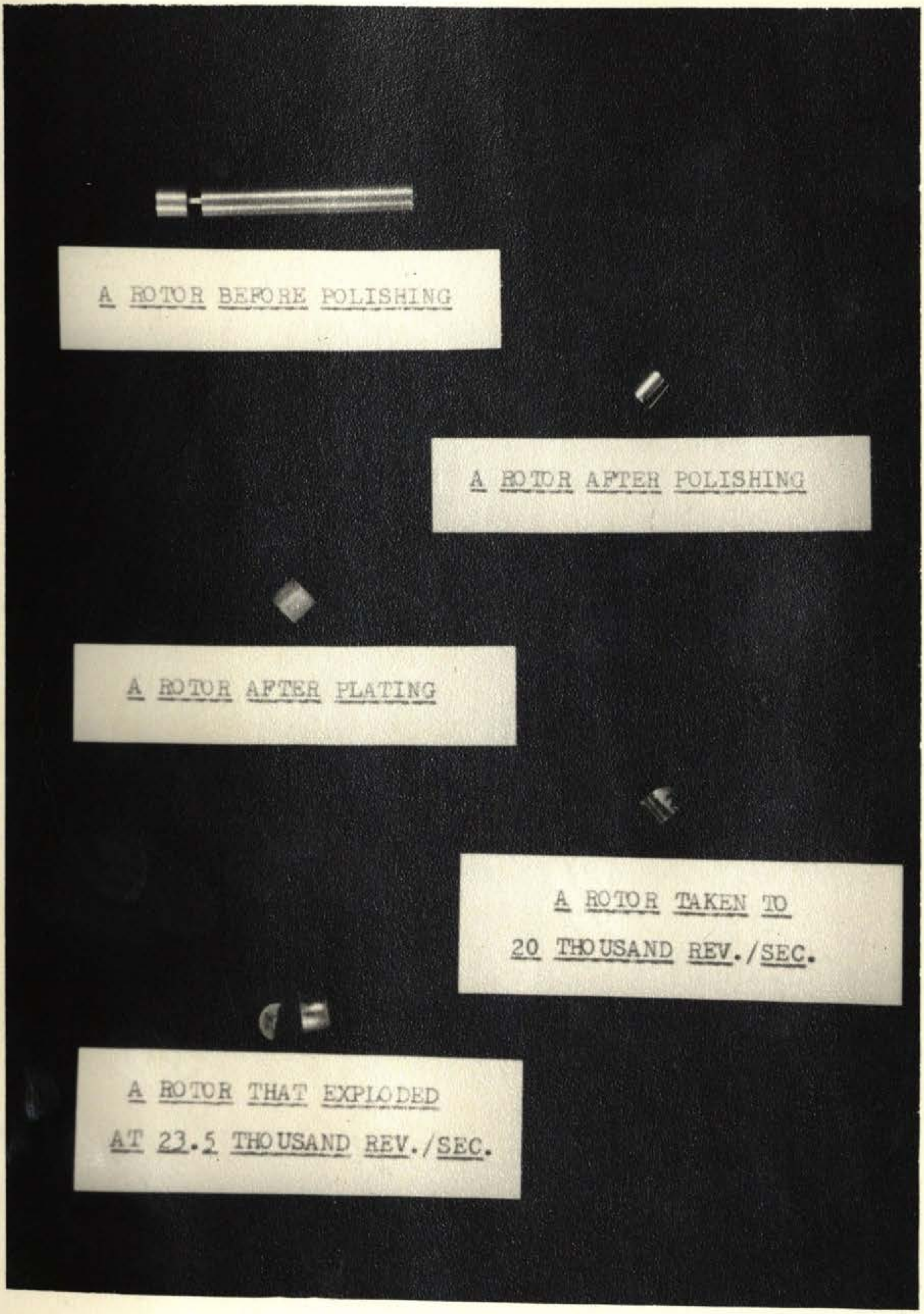


Fig. 10. Rotors

this case.

In trying to eliminate the tension in the film, the cutting of the rotor probably did not help since the film at the edges of the cut seemed to adhere better than the rest of the film did. This would make it necessary to overcome the tensile strength of the film before the film could leave the rotor no matter how small the adhesion might be.

Even though preliminary measurements did not turn out well from the standpoint of greatly reducing adhesion, the vacuum ultra-centrifuge was demonstrated to be well fitted for these measurements. The quarter-inch rotor is satisfactory for measuring the low values of adhesion sought, but high values of adhesion could easily be measured by reducing the rotor diameter.

It is my hope that someone will continue the investigation for which this instrument was designed.

APPENDIX I

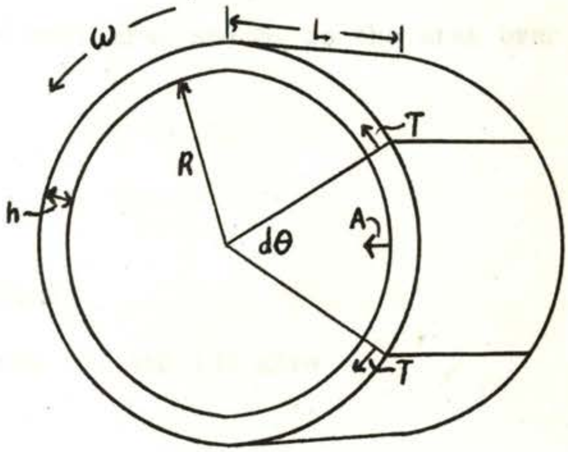
FORCES ACTING ON THE PLATED FILM

Consider a rotor of radius R and length L with a film of thickness h plated on its surface. When the rotor spins about its axis of revolution, the centrifugal force on an increment of mass a distance r from the axis of the rotor is given by

(1) $(dF_c) = a(dm)$,

where $a = \omega^2 r$.

Now, if d is the density of the material, the increment of mass is just



(2) $(dm) = rLd(d\theta)(dr)$

and equation (1) becomes

(3) $(dF_c) = \omega^2 Ldr^2(d\theta)(dr)$.

By integrating equation (3) over the thickness of the film

$$F_c = \omega^2 Ld(d\theta) \int_R^{R+h} r^2(dr)$$

Fig. 11

or

(4) $$F_c = \omega^2 Ld(d\theta)\left[R^2 h + Rh^2 + \frac{h^3}{3}\right].$$

This centrifugal force will be balanced by a centripital force due to both the adhesion of the film to the rotor and the tension in the film, or

(5) $F_c = F_a + F_t$.

If the slight irregularities caused by edges of the film at the ends of the rotor are neglected, the centripital force on an element of mass due to the tension in the film is

(6) $$F_t = 2TS_t \sin(\frac{d\theta}{2}),$$

where T is the tensing force per unit area and S_t is the area over which T is applied. From Fig. 11, it can be seen that

(7) $$\sin(\frac{d\theta}{2}) = \frac{R(\frac{d\theta}{2})}{R} .$$

Thus, equation (6) becomes

(8) $$F_t = TS_t(d\theta)$$

If A is the adhesive force per unit area and S_a is the area over which A is applied then

(9) $$F_a = AS_a.$$

As shown by Fig. 11,

(10) $$S_t = hL \quad \text{and} \quad S_a = LR(d\theta)$$

which when substituted into equations (8) and (9) give

(11) $$F_t = ThL(d\theta)$$

and

(12) $$F_a = ALR(d\theta).$$

If equations (4), (12), and (13) are substituted into equation (5),

$$\omega^2 Ld(d\theta)\left[R^2h + Rh^2 + \frac{h^3}{3}\right] = ALR(d\theta) + ThL(d\theta)$$

or by simplifying,

(13) $$\omega^2R^2d\left[1 + \frac{h}{R} + \frac{h^2}{3R^2}\right] = A\frac{R}{h} + T.$$

If $R >> h$, then equation (13) becomes

(14) $$\omega^2R^2d = A\frac{R}{h} + T$$

which is the equation used to determine adhesion and tensile strength when working with thin films.

BIBLIOGRAPHY

[1]Beams, J. W., J. L. Young, III, and J. W. Moore, J. Appl. Phys., 17 (1946), 886.

[2]Beams, J. W., W. E. Walker, and H. S. Morton, Jr., Phys. Rev., 87 (1952), 524.

[3]Beams, J. W., J. B. Breazeale, and W. L. Bart, Phys. Rev., 100 (1955), 1657.

[4]Beams, J. W., "Mechanical Properties of Thin Films of Gold and Silver", Structure and Properties of Thin Films (New York, N.Y., 1959), 183-192.

[5]Mentor, J. W. and D. W. Pashley, "The Micro Structure and Mechanical Properties of Thin Films", Structure and Properties of Thin Films (New York, N.Y., 1959), 149.

[6]Beams, J. W., Tech. Proc. Am. Electroplaters Soc., 43 (1956)

[7]Handbook of Chemistry and Physics, 36th ed. (Cleveland, Ohio: Chemical Rubber Publishing Co., 1954)

[8]Blum, W. and G. B. Hogaboom, Principles of Electroplating and Electroforming, 2nd ed. (New York, N.Y., 1930), 228.