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RESEARCH WITH CRYSTAL COUNTERS AND
SCINTILLATION COUNTERS

FINAL REPORT

Prepared by
George Goldsmith and Lee Aukerman

Contract N₇ONR-39413

between

OFFICE OF NAVAL RESEARCH

and

PURDUE RESEARCH FOUNDATION
Purdue University
Lafayette, Indiana
February 13, 1952

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RESEARCH WITH CRYSTAL COUNTERS AND SCINTILLATION COUNTERS

I. Introduction

The purposes of the experiments carried out under this program are: (a) to develop efficient and reproducible counters and (b) to investigate the physical phenomena involved in the counting process as an aid in understanding further luminescence in molecular systems. Of particular interest under part (b) of the program is the mechanism of energy transfer in the systems under consideration.

A number of aspects of this program have been under investigation in this laboratory previously. Among these are the development of P-N barrier (germanium) counters, measurement of the fluorescence spectrum of anthracene at room temperature, investigation of conductivity counting in organic phosphors, scintillation counting with solutions of organic materials, and the measurement of some of the electrical properties of cadmium sulfide crystals under various conditions. This project affords an opportunity to place more emphasis on a systematic study of these problems.

During the contract period being reported herein, the greater portion of the time and personnel has been devoted to the development and construction of equipment suitable for making the desired measurements. Much effort has been spent on instrumentation which will provide adequate precision for all anticipated situations.

The extent of the field under consideration in the program necessitated the selection of several separate experiments chosen to suit the ability and

interests of the personnel and to utilize efficiently the available facilities. These are summarized briefly below.

A. Crystal Growing

In addition to providing the essential materials of research, the crystal growing experiments are concerned with the development of methods for growing crystals of mixed composition.

B. Fluorescence Intensity Measurements

A sensitive vacuum tube electrometer has been constructed for the measurement of the total current produced in a multiplier phototube by the luminescence light. This apparatus is arranged for the convenient measurement of liquid, solid, and plastic phosphors under rigidly reproducible conditions. Among the experiments planned for this equipment is the measurement of the influence of the viscosity on the luminescence output of solutions of organic phosphors.

C. Pulse Measurements

The apparatus for the measurement of the pulse characteristics of the phosphors is designed to minimize the effects of the phototube background pulses. It also provides for the analysis of the pulse height distribution through use of a single-channel pulse-height analyzer.

D. Photoconductivity Measurements

The photoconductivity of solutions of organic materials in organic solvents has been observed previously (1) though no modern measurements have been published. It is hoped, through the performance of such measurements

with more refined techniques than were available at the time of the original measurements, that useful information regarding the process itself and the associated problem of energy transfer can be obtained. The apparatus nearing completion consists of a suitable cell, a vacuum tube electrometer capable of measuring currents as low as 10^{-15} amperes, and a source of ultraviolet light.

E. Spectra

Indications from experiments of L. Roth carried out in this laboratory in 1948 are that the mode of excitation (alpha, beta, gamma, X and ultraviolet radiation) influences the structure of the fluorescence spectrum of anthracene. Interpretation of this influence can lead to a further understanding of the phenomena involved. These earlier measurements were not sufficiently precise to permit valid conclusions to be drawn. Since then this laboratory has become equipped with excellent low temperature equipment, and it is intended to repeat these measurements at very low temperatures where the fluorescence bands are resolved into rather discrete lines (2).

F. P-N Barriers and CdS Counters

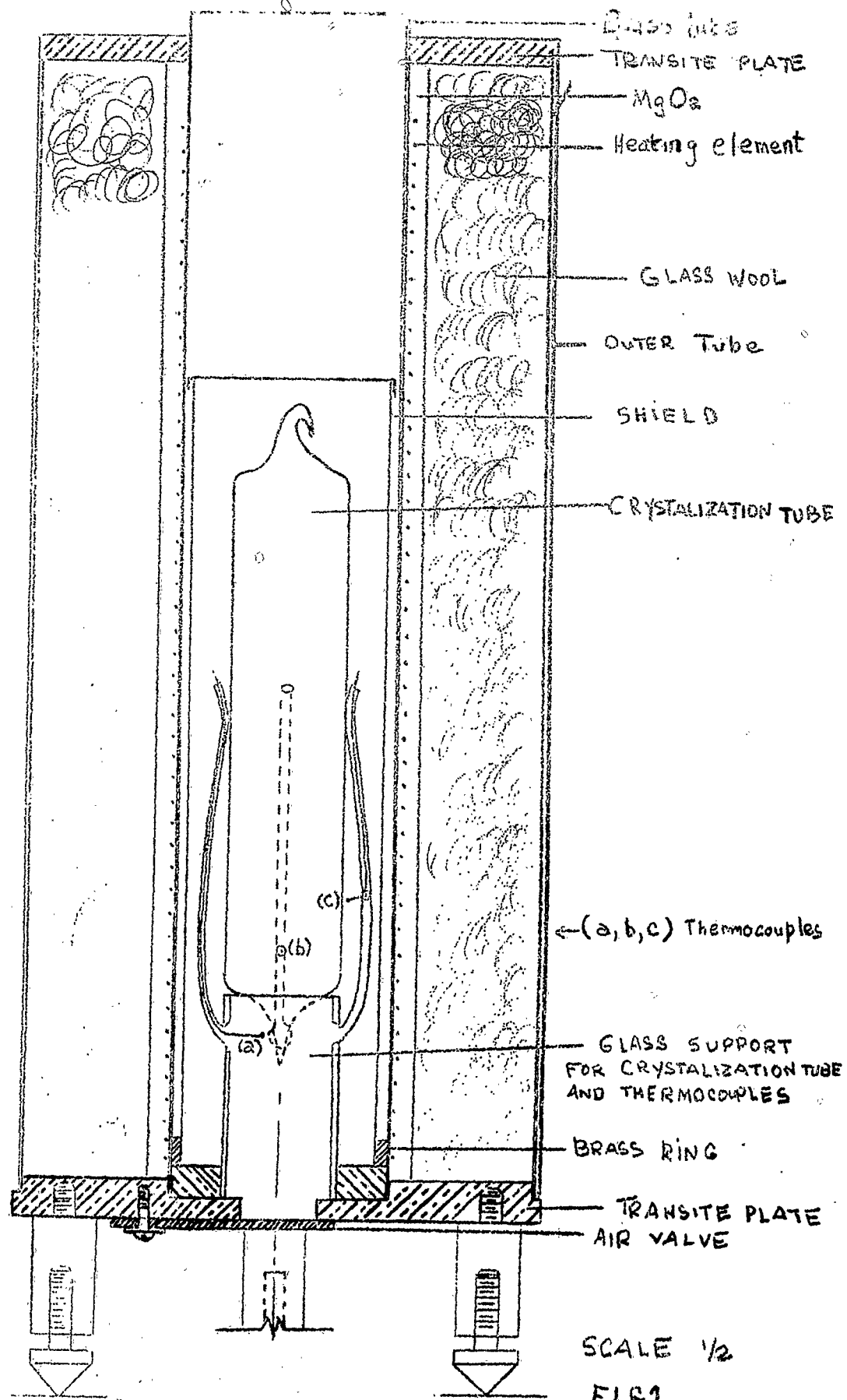
As noted above measurements on these types of counters have been under way in this laboratory previous to the beginning of this particular program. (3, 4) Both materials show promise as useful counters which have the advantage of being small and relatively simple. The work performed during the period reported is a continuation of this work, concentrating on the measurements of the properties of P-N barrier counters.

II. Crystal Growing

Large single crystals of organic phosphors are conventionally grown from the melt by a slow cooling. Numerous procedures have been proposed for accomplishing this. A technique described by Huber, Humbel, Schneider and Steffen (5) was selected as being the simplest and most economical. In this method the material to be crystallized is melted in a conventional glass crystallization tube which has been either evacuated or filled with inert gas at a reduced temperature. The furnace (Figure 1) is designed to have a continual thermal gradient, increasing in temperature toward the top (Figure 2). Growth of the crystal is brought about by slowly cooling the entire furnace by reducing the applied voltage with a motor driven Variac (Figure 3).

At the beginning of the crystallization run, the entire furnace is brought to a temperature slightly above the melting point of the phosphor and held there until the sample is completely melted. Cooling is then started at a rate of about one degree per hour. Because of the thermal gradient, nucleation will take place in the constricted bottom of the crystallization tube. In this manner only one crystal may form. To ensure further that the thermal gradient remains undisturbed during the crystallization period, the tube is surrounded with an aluminum radiation shield. During the run the temperature and the gradient (which should be about $1^{\circ}\text{C}/\text{cm}$) is checked periodically by means of thermocouples inserted along the length of the tube.

Pretreatment of the material in preparation for crystallization depends on the phosphor itself and upon the source from which it is obtained.



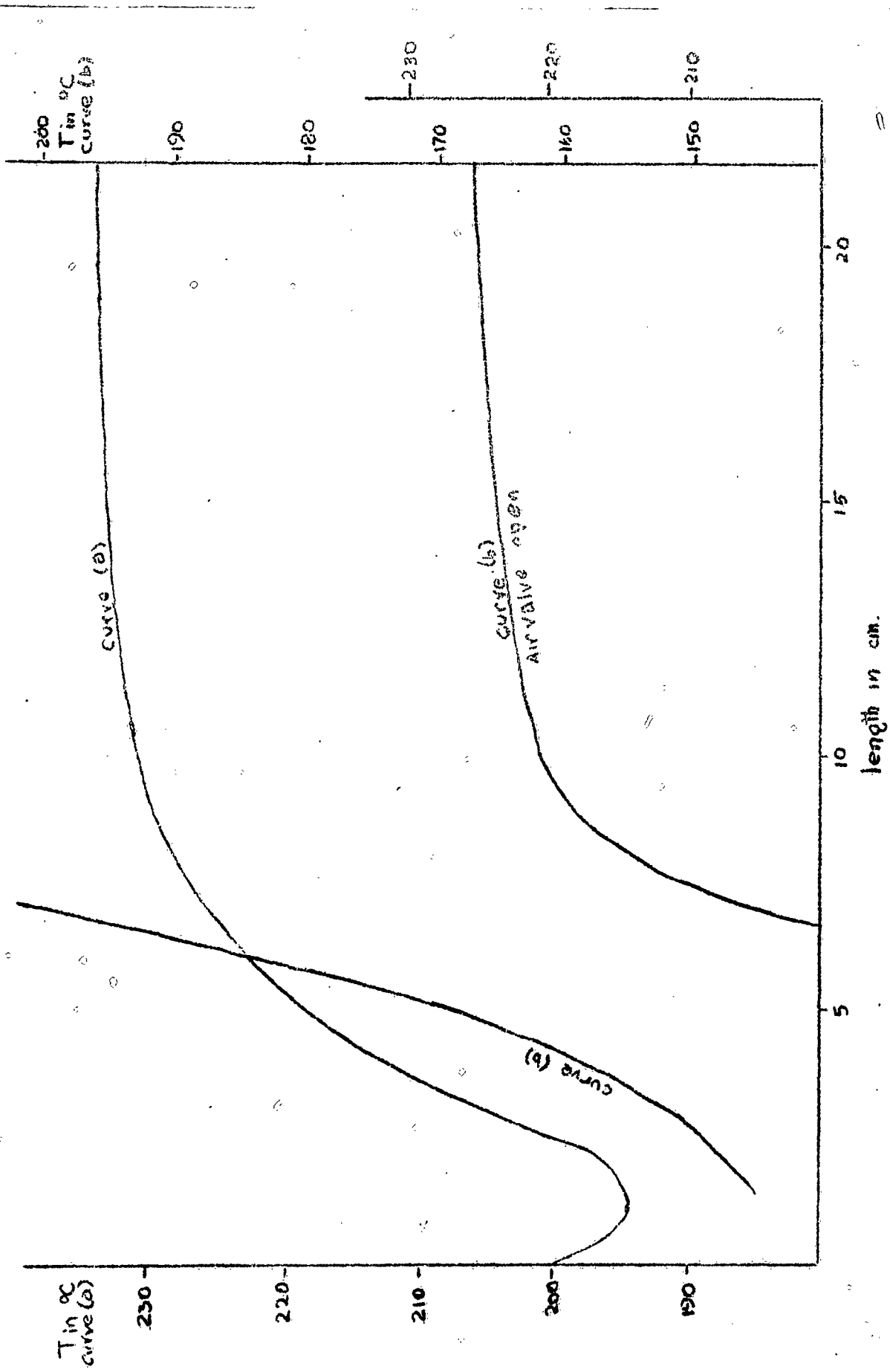


FIG. 2

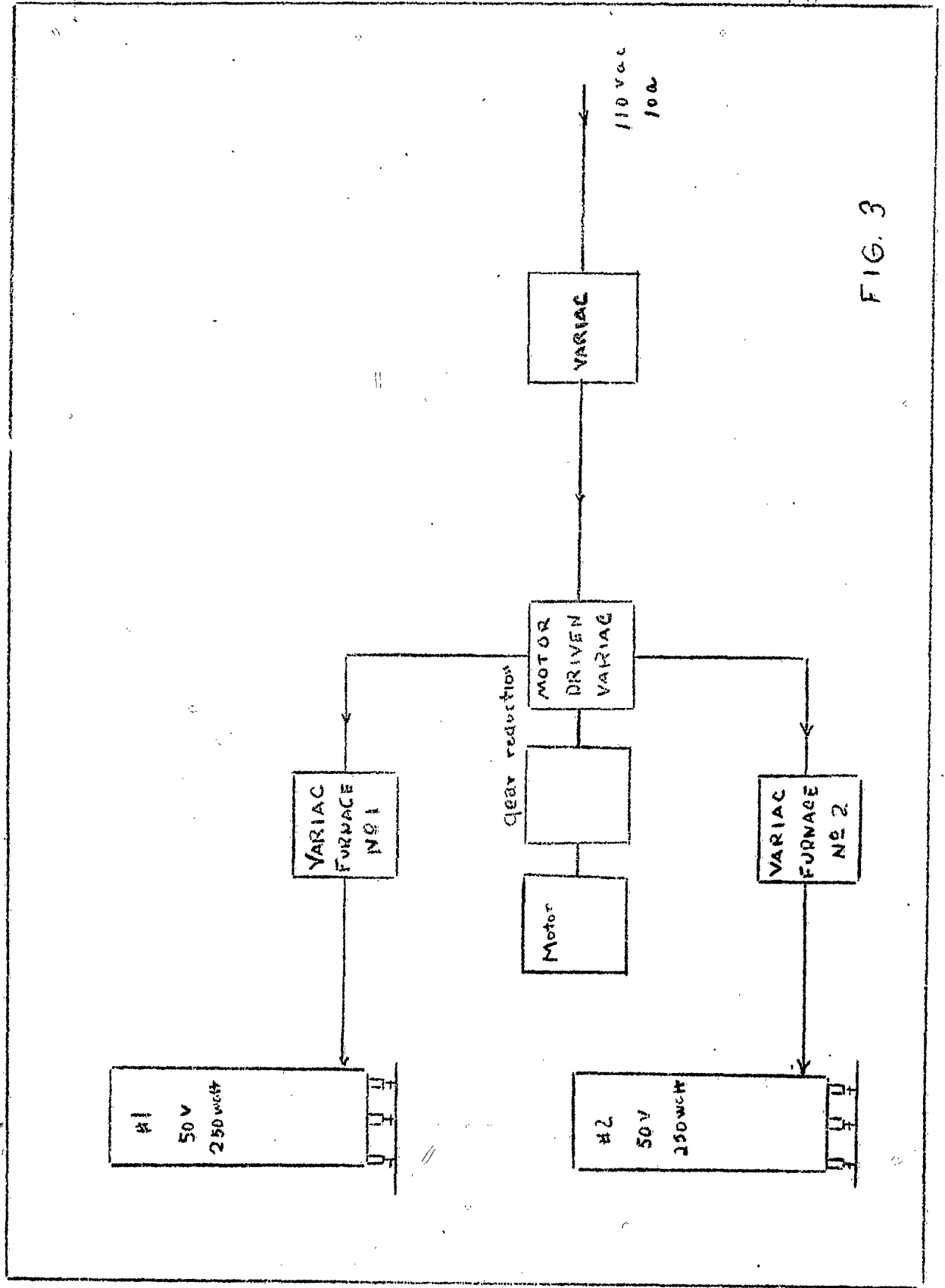


FIG. 3

Anthracene of "scintillation grade" is obtained from Riley Coal Tar Products Company. This material has been highly purified and requires only a single resublimation prior to recrystallization. The principal purpose of this treatment is to remove residual solvent. Anthracene must be sublimed with considerable care because it undergoes polymerization readily at elevated temperatures and is subject to oxidation if heated in air. The sublimation is carried out under continual pumping to remove the solvents and to prevent oxidation. The apparatus (Figure 4) consists of a large glass test tube with a ground fitting on its open end. A second tube is inserted into the apparatus to facilitate removal of the sublimate which collects on the walls of the inner tube. The lower end is heated by means of a heating jacket and the upper end is cooled with a stream of cold air. The sublimation temperature is about 125° C which yields a sublimation rate of 10 g/day. More rapid sublimation results in a yellowish product evidencing excessive polymerization.

It has not been possible to obtain terphenyl of purity equalling that of the anthracene. This material must be recrystallized from benzene and then resublimed as described above.

Transtilbene as obtained from Larco Nuclear Engineering Company appears to be of sufficient purity to introduce immediately into the crystallization tube. There is, however, evidence that this material also contains residual solvent.

The above phosphors are the only materials currently under consideration since they show the greatest usefulness.

SUBLIMATION TUBE

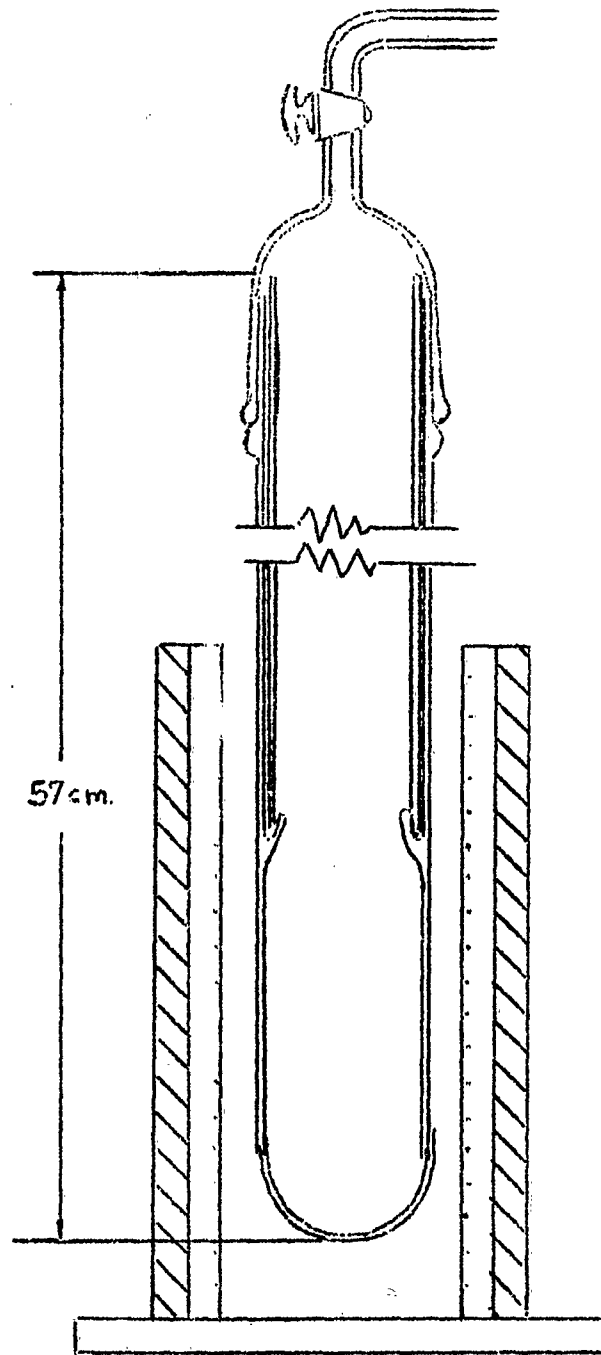


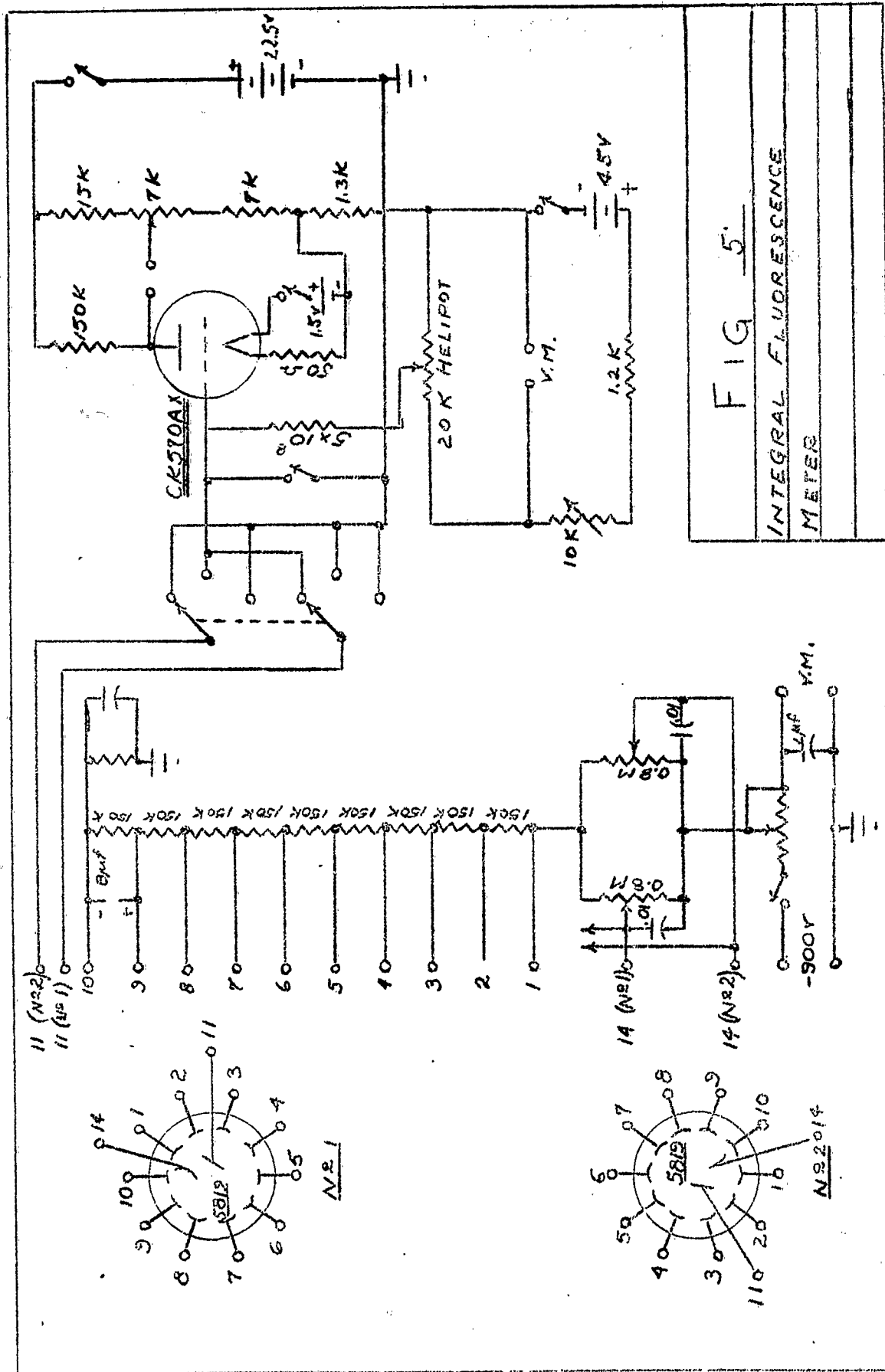
FIG 4
SCALE 1/2

At the present time two furnaces are in operation and two additional furnaces are under construction. Though the crystals obtained by this method are generally satisfactory, they are not as transparent as might be desired. Work is in progress to improve the quality of the product.

Cadmium sulfide crystals are produced by the vapor-phase condensation method of R. Frerichs (6). The apparatus was constructed previous to the inception of this project, but it had to be reassembled because of changes in the laboratory location. It is not currently in operation.

III. Fluorescence Intensity Measurements

The apparatus (Figure 5) for the total fluorescence intensity measurements consists of two RCA Type 5819 multiplier phototubes either of which can be switched to a conventional triode electrometer circuit. The voltage developed by the photocurrent in the grid resistor is compared with the potential developed by a battery across a 20,000 ohm helipot, hence the electrometer is used as a null point detector. The multiplier tubes have glass cylinders cemented to the top of the bulb into which is placed the phosphor under investigation. In this manner efficient light collection can be achieved. The entire assembly is battery operated including the high voltage supply for the multiplier tubes. Provision is made for accurate check of all significant operating voltages. The multiplier tubes are protected from the effects of stray magnetic fields (to which the Type 5819 multiplier is very sensitive) through use of mu metal shields.



Despite the precautions taken to ensure complete reproducibility of the operating conditions from day to day, measurements indicate that there is considerable fluctuation of the photocurrent. This may be due partially to fluctuations in the room temperature which certainly accounts for variations in the dark current. Such large fluctuations as have been observed cannot, however, be accounted for entirely by this temperature dependence--especially since the dark current is subtracted from each measurement. It must be concluded, then that the efficiency of the photocathode does not remain constant. This situation indicates the necessity for establishing some sort of calibration method to be used with each measurement. The method currently under investigation is that of illuminating the photocathode from the underside with a small incandescent light source. There appears to be, however, some difficulty with this technique since it is not certain that the output from an incandescent light is exactly reproducible considering the sensitivity of the multiplier tubes both to intensity and to spectral distribution of the incident light.

This apparatus is intended to be used principally with plastic and liquid phosphors.

IV. Pulse Measurements

The pulse measuring equipment (Figure 6) consists of a coincidence gate circuit and a differential pulse height analyzer. The circuit is so designed as to pass for analysis only those pulses which are coincident between the two multipliers, thus minimizing the background due to multiplier noise and permitting analysis of pulses of very low amplitude. Descriptions of the individual units follow.

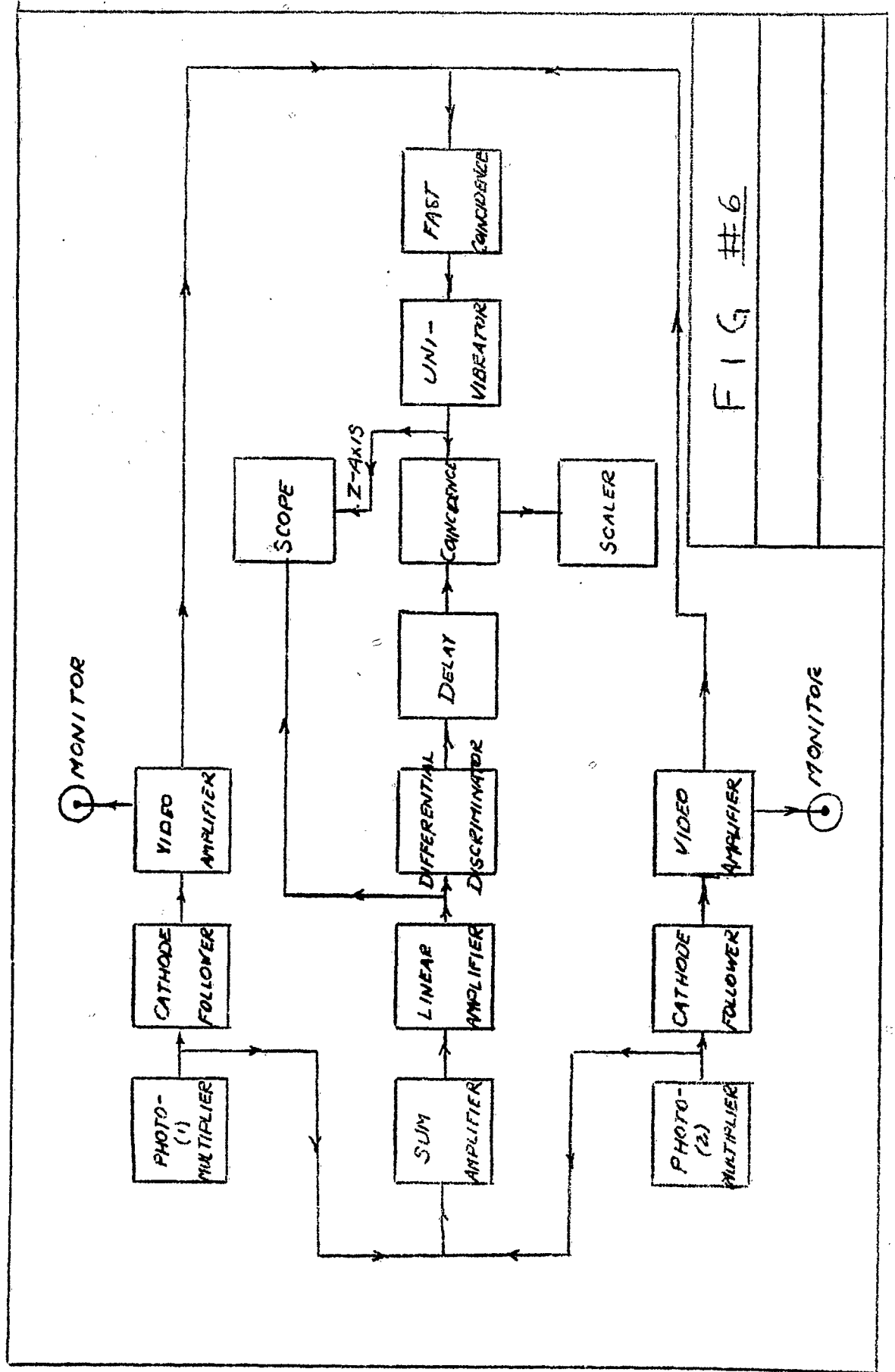


FIG #6

A. Multiplier unit. The multiplier unit consists of the two RCA type 5819 multiplier phototubes, the cathode followers, and the sum mixer.

The multipliers are mounted horizontally facing each other on movable carriages. To ensure efficient light collection, 1/2" Lucite light pipes are coupled to the tube faces by means of a thin layer of cedar oil (Figure 7). This eliminates the troublesome problem of achieving good optical contact to the curved faces of the tubes. The light collection assembly is attached to the multiplier tubes by means of strips of "Scotch" electrical tape. This method is not particularly satisfactory since the cedar oil attacks the tape somewhat. Other cementing techniques are being sought.

The cathode followers (Figure 8) are also mounted on the individual carriages in order to minimize wiring capacitance.

The sum mixer, a double cathode follower, is mounted beneath the plate on which the carriage assembly rests.

B. Coincidence circuits. Because of the fact that it is desirable to observe very small pulses, it was necessary to employ some amplification of the output pulses prior to coincidence mixing. In order to extend the lower limit of the measurable pulse height a useful amount, it is necessary that these amplifiers be relatively "fast" that is they must have rise times of the order of 10^{-8} seconds. It is not necessary that these be "linear" amplifiers, but they must have constant rise time and pulse width over a wide range of amplitudes. Attempts to construct simple RC amplifiers having these characteristics by using a large number of stages with small gain per stage proved unsuccessful. The amplifiers finally

LUCITE
POLISHED ALUMINUM
OIL SPACE
FILLING TUBE
TUBE
OUTER TAPE
M-METAL SHIELD
INNER TAPE

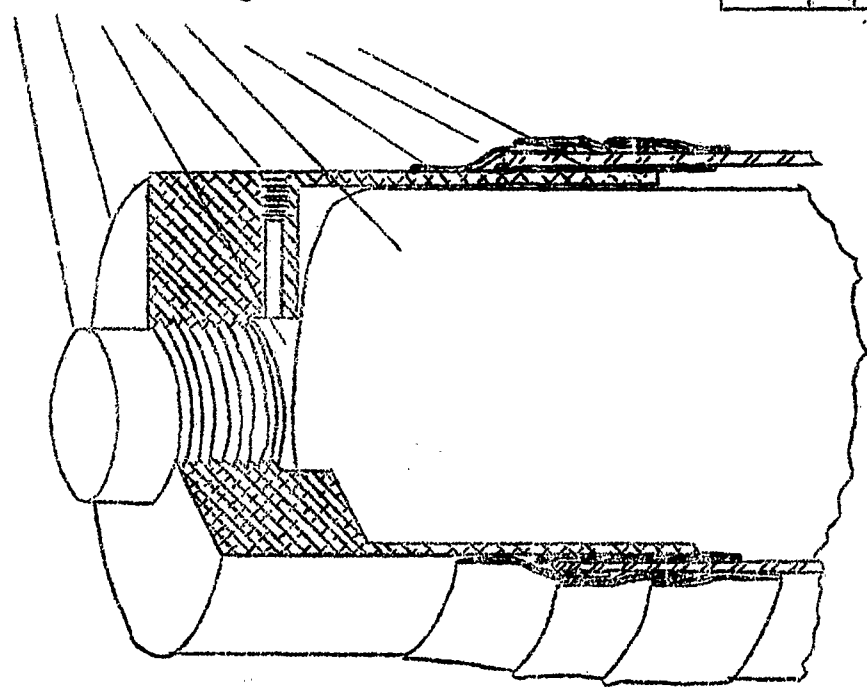


FIG 7

LIQUID LIGHT PIPE AND
MULTIPLIER SHIELD

(NOT TO SCALE)

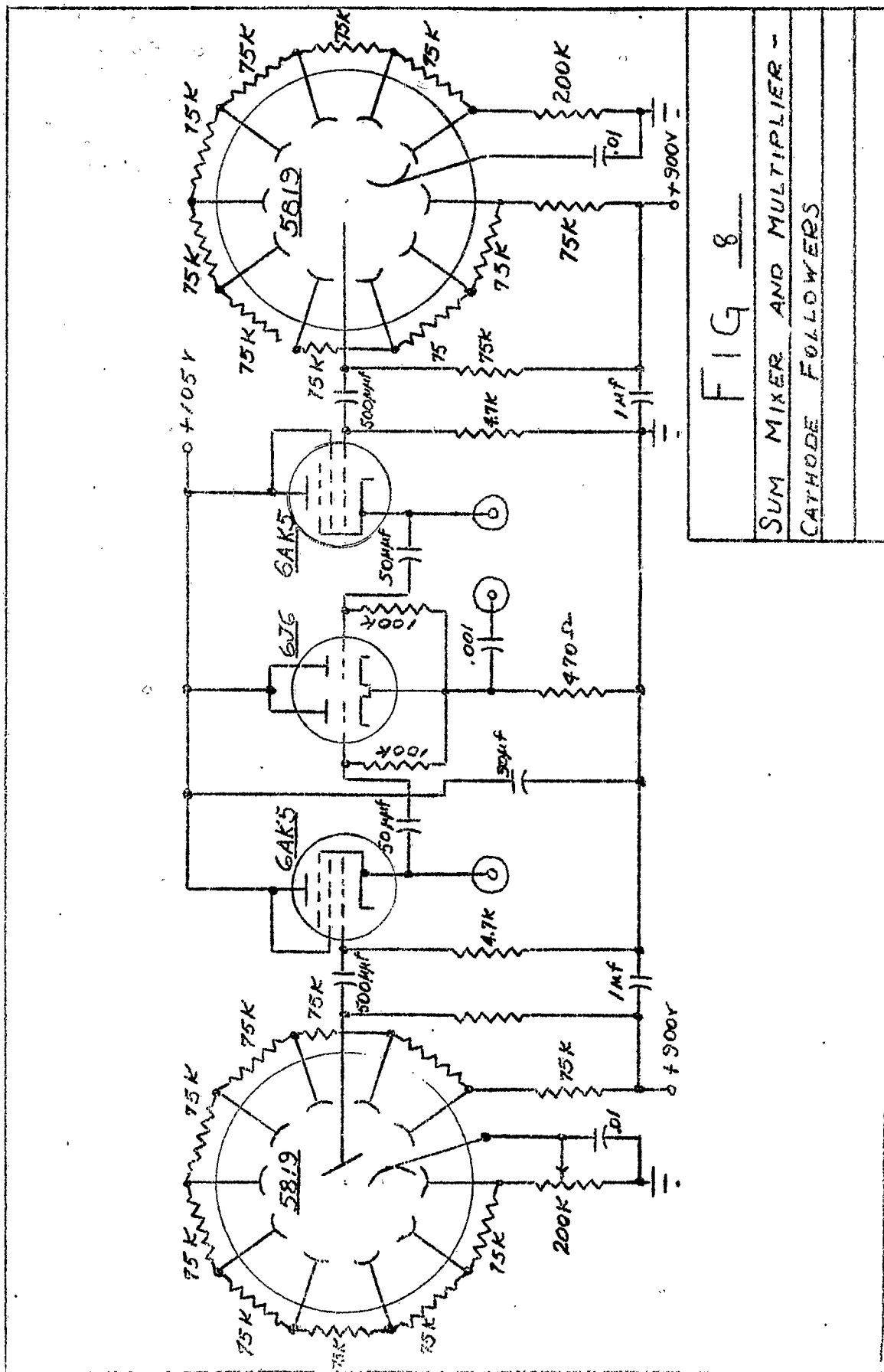


FIG 8
 SUM MIXER AND MULTIPLIER -
 CATHODE FOLLOWERS

employed (Figure 9) are modified "noise" amplifiers from Navy Radar jamming equipment. The rise time of these amplifiers (Figure 10) is of the order of 5×10^{-8} seconds and the gain, of the order of 10^3 . Under conditions of very low pulse amplitude, additional preamplifiers are used (Figure 11).

The coincidence circuit is a modified Rossi circuit designed by Garwin (7). This circuit which features high resolution and high sensitivity is ideally suited to our problem. The coincidences are selected by a Schmitt-Trigger discriminator (Figure 12).

C. Linear Amplifier and Differential Analyzer, Analysis of the scintillation pulses is performed on the sum of the outputs from the two multiplier tubes. In this manner the effect of self absorption of the fluorescence radiation in the phosphor can be minimized and the useful photosensitive area doubled.

The sum pulse is amplified by a commercially available linear amplifier (Atomic Instruments Company Model 204B). As originally constructed, this amplifier has on its output a precision integral pulse height analyzer. The following modifications convert this into a single channel differential discriminator (Figures 13 and 14).

A second integral pulse height selector was constructed identical with the one in the commercial amplifier. The bias of the discriminator tube in this circuit is obtained from the same voltage divider which supplies the bias for the first discriminator. Provision is made for adjusting the "zero" level for this second discriminator at some value above that of the first unit. Thus pulses of sufficient amplitude to trigger the first discriminator will not necessarily trigger

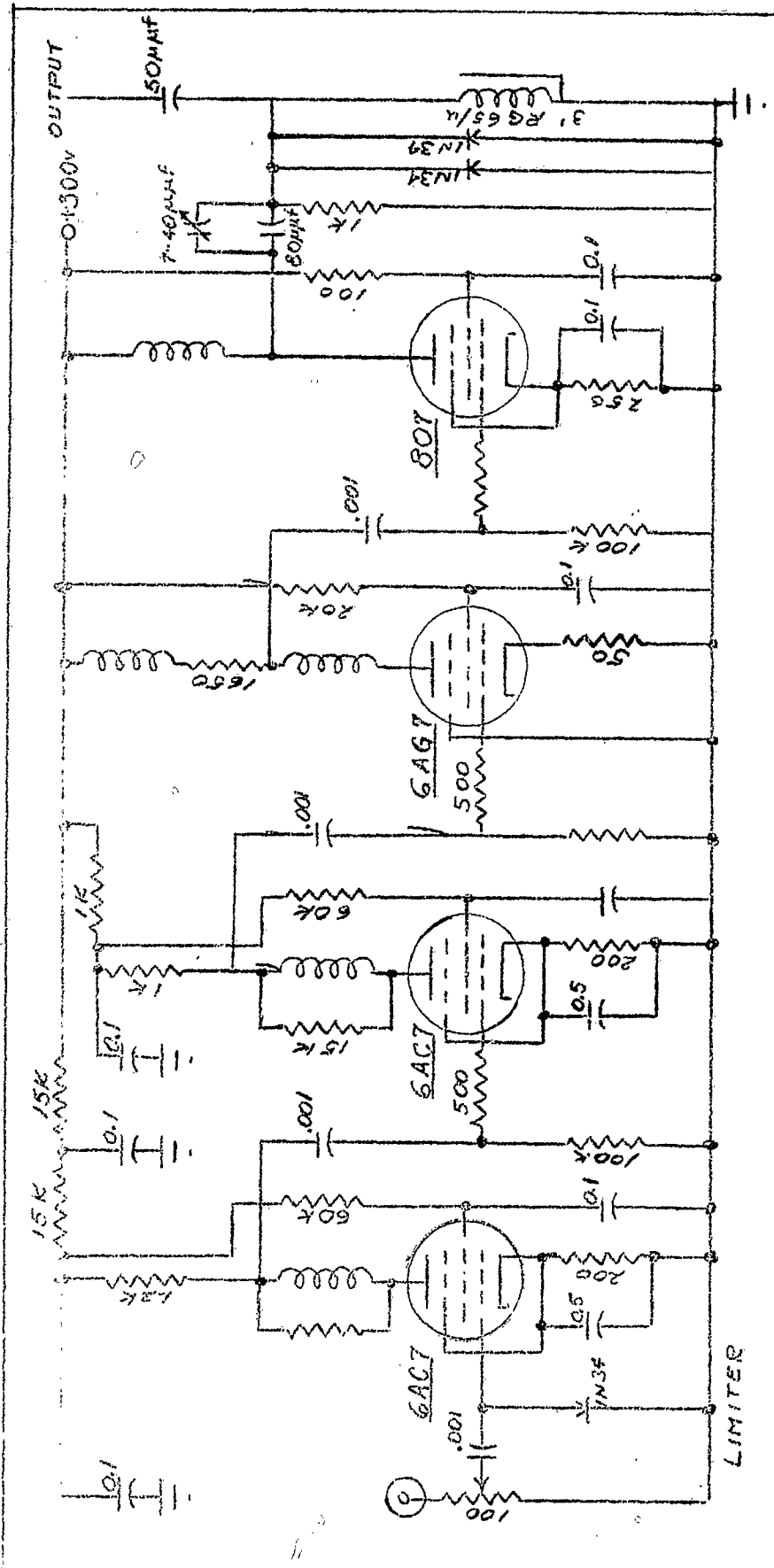


FIG 9

FAST COINCIDENCE AMPLIFIER

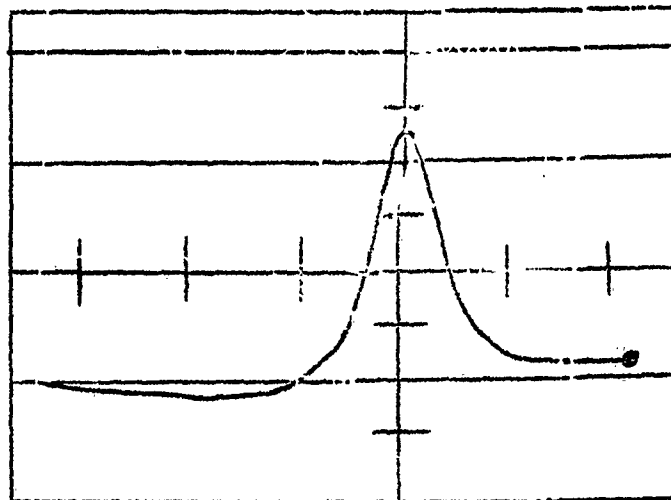
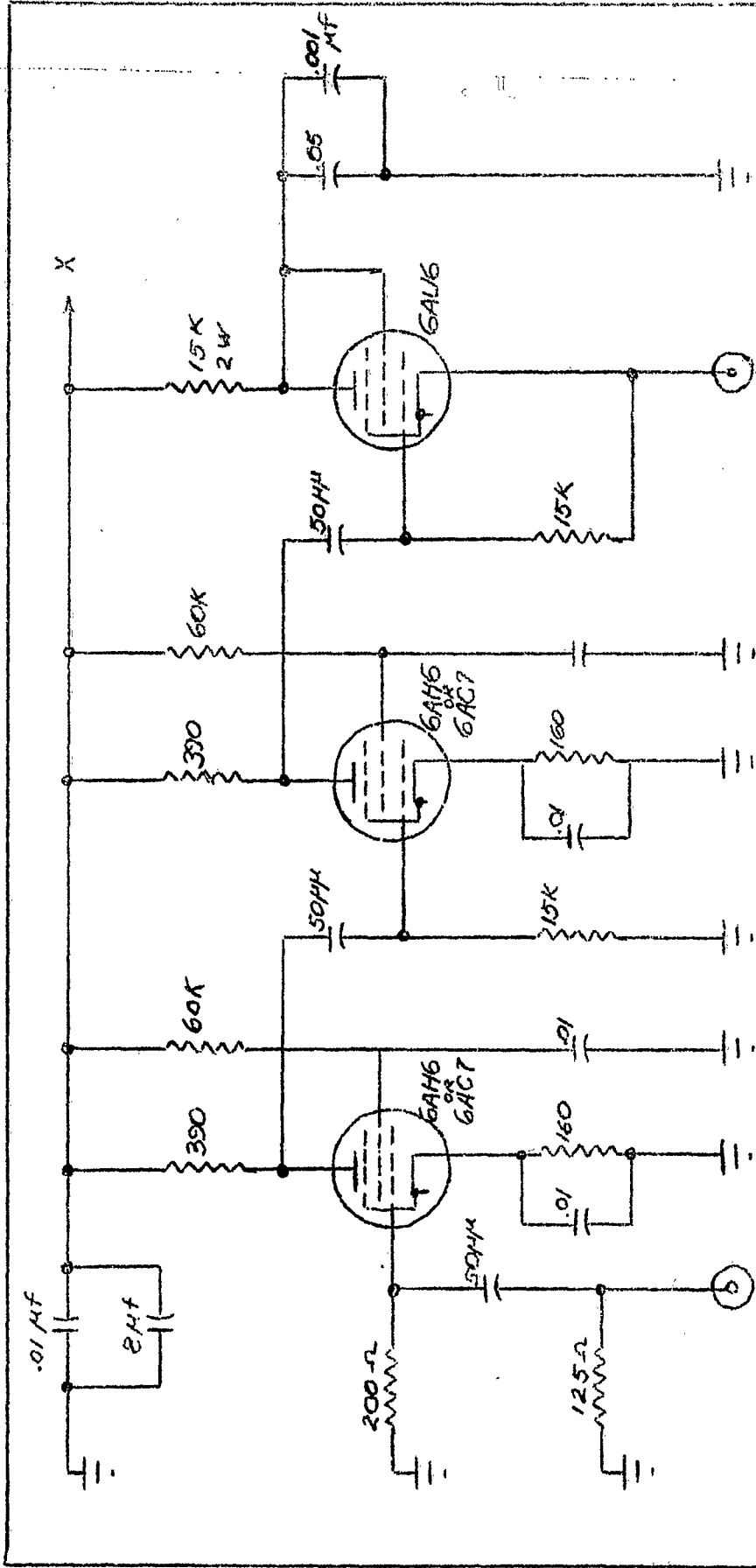


Fig. 10

Signal from coincidence amplifiers

(Sweep from right to left, 0.1 micro-seconds between markers--traced from photograph.)



NOTE: ALL INTERSTAGE LEADS AS SHORT AS POSSIBLE AND NOT CLOSE TO CHASSIS

FIG II

PRE - AMPLIFIER

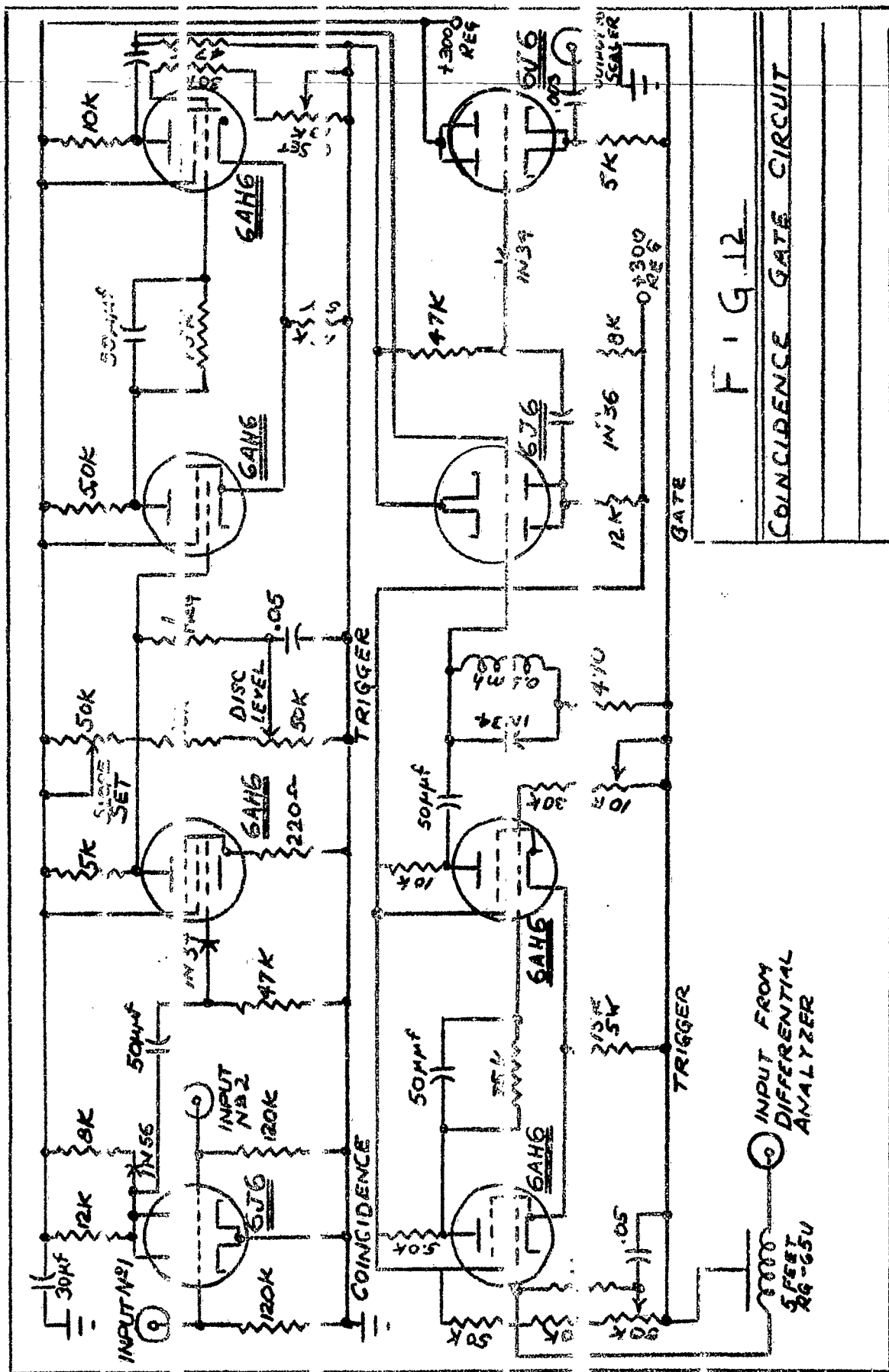


FIG. 12
COINCIDENCE GATE CIRCUIT

INPUT FROM
DIFFERENTIAL
ANALYZER

200V
50Hz
24-65V

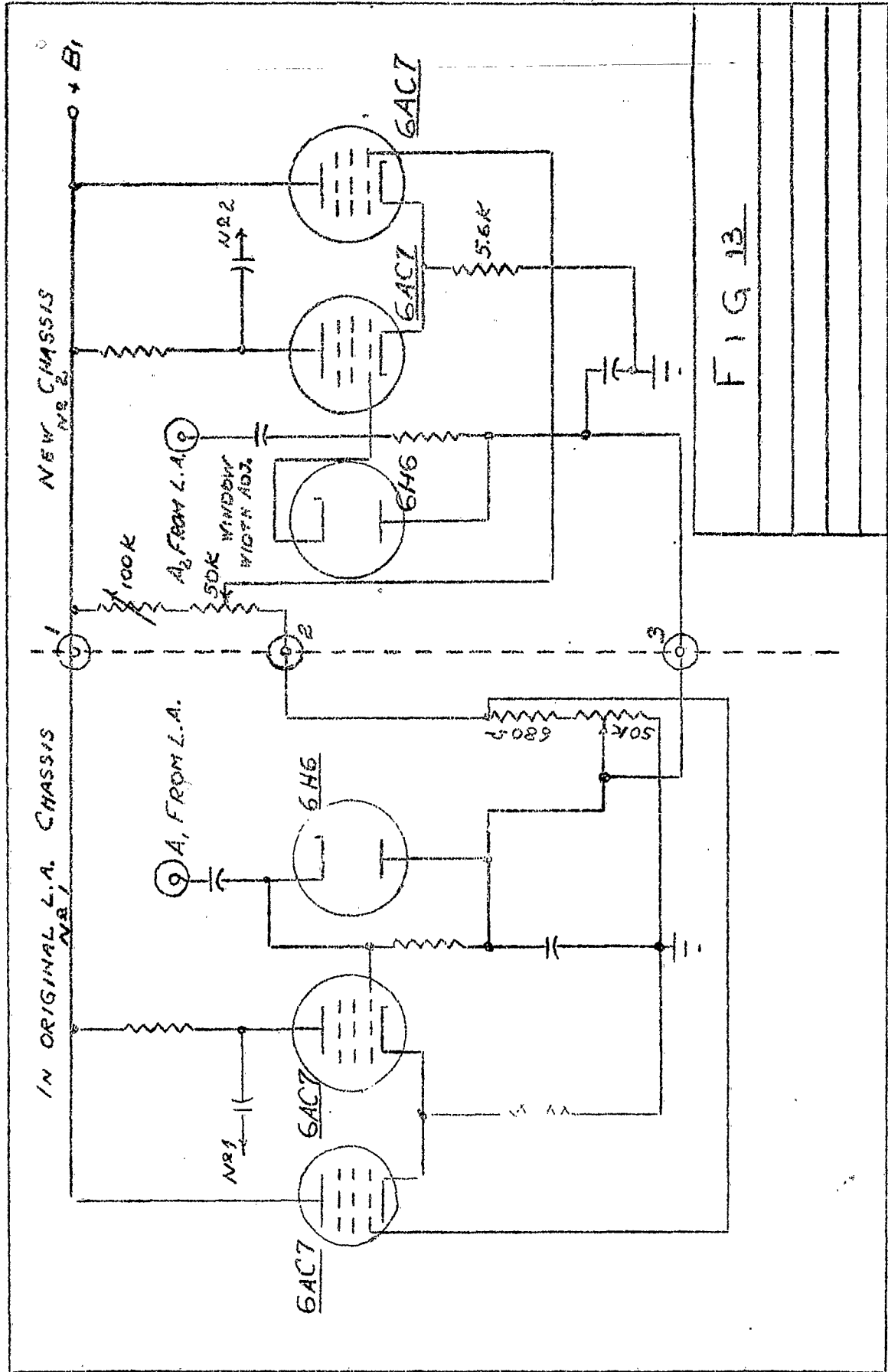


FIG 13

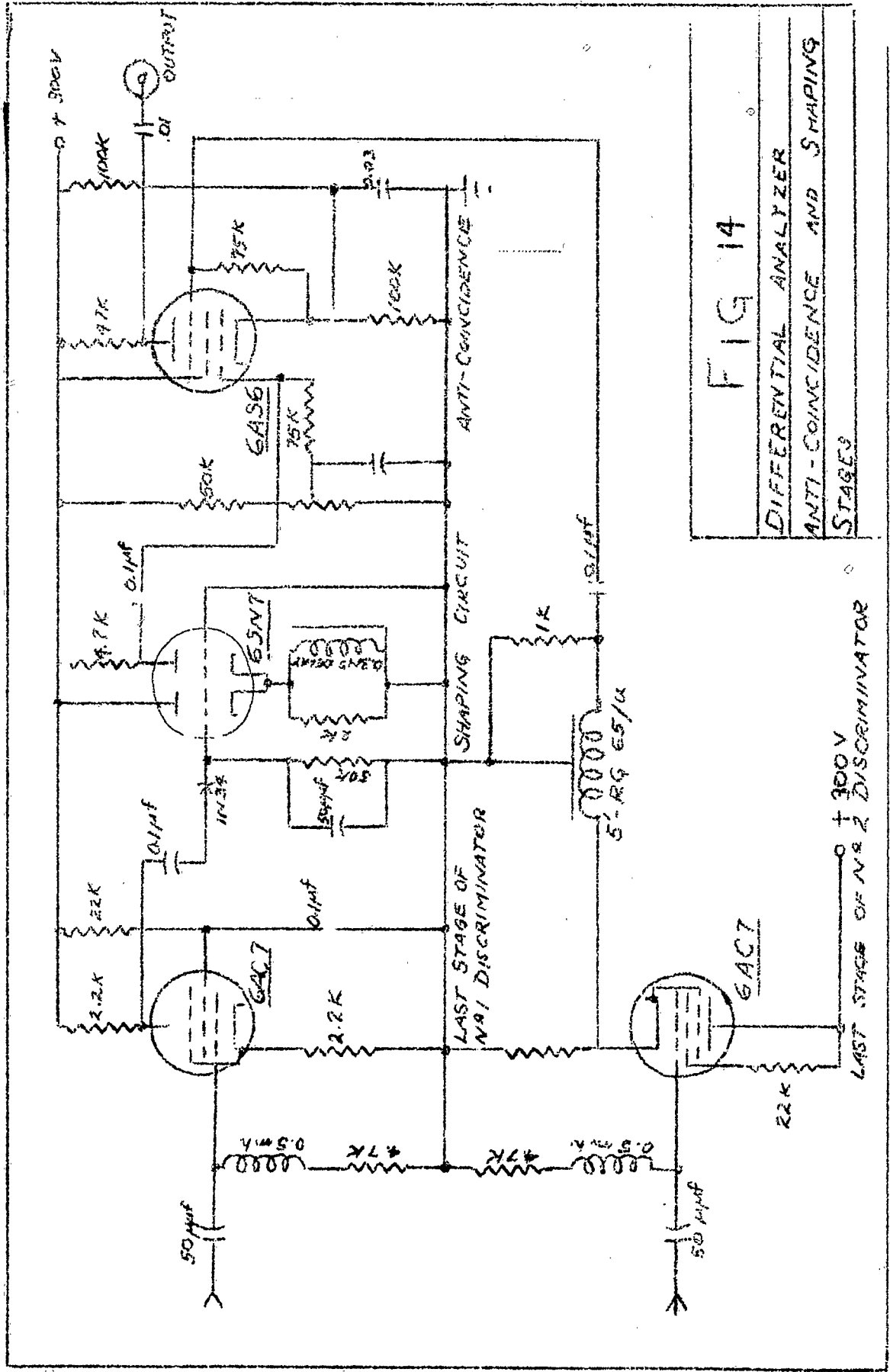


FIG 14

DIFFERENTIAL ANALYZER
ANTI-COINCIDENCE AND SHAPING
STAGES

LAST STAGE OF N° 2 DISCRIMINATOR + 300 V

the second discriminator unless the amplitude exceeds the threshold level for both. Anti-coincidence circuits, together with suitable pulse shaping and delay networks to account for the time difference in triggering the upper and lower discriminators, select those pulses which are large enough to trigger the lower discriminator but not the upper. This, then, gives the pulses falling within a certain interval between V and $V + V$, V being selected by adjustment of the "zero" level of the upper discriminator.

The output of the differential analyzer and the gating pulse from the original coincidence are mixed in a second Garwin coincidence tube which serves as the gate (Figure 12). The output of the gate is then fed to a scaling circuit.

D. Operating Characteristics of the Circuits. The shape and constancy of the "window" of the differential discriminator have been checked by application of a step wave of constant known amplitude to the input of the linear amplifier. The window shape is a true Gaussian (Figure 15) and is constant in width and form over a range of inputs from 2 mv to 1 v. Further check of the operation of this circuit was made by analyzing the X-ray spectrum from the conversion electrons of In^{114} with a proportional counter. This measurement indicates satisfactory operation.

The resolution time of the coincidence circuit was measured by delaying the input through one of the channels from a single channel (Figure 16). The resolving time with negligible losses is about 6×10^{-8} seconds. This can be improved considerably by acceptance of a larger percentage of losses of true coincidences. The useful range, however, is not too large since limitation of the

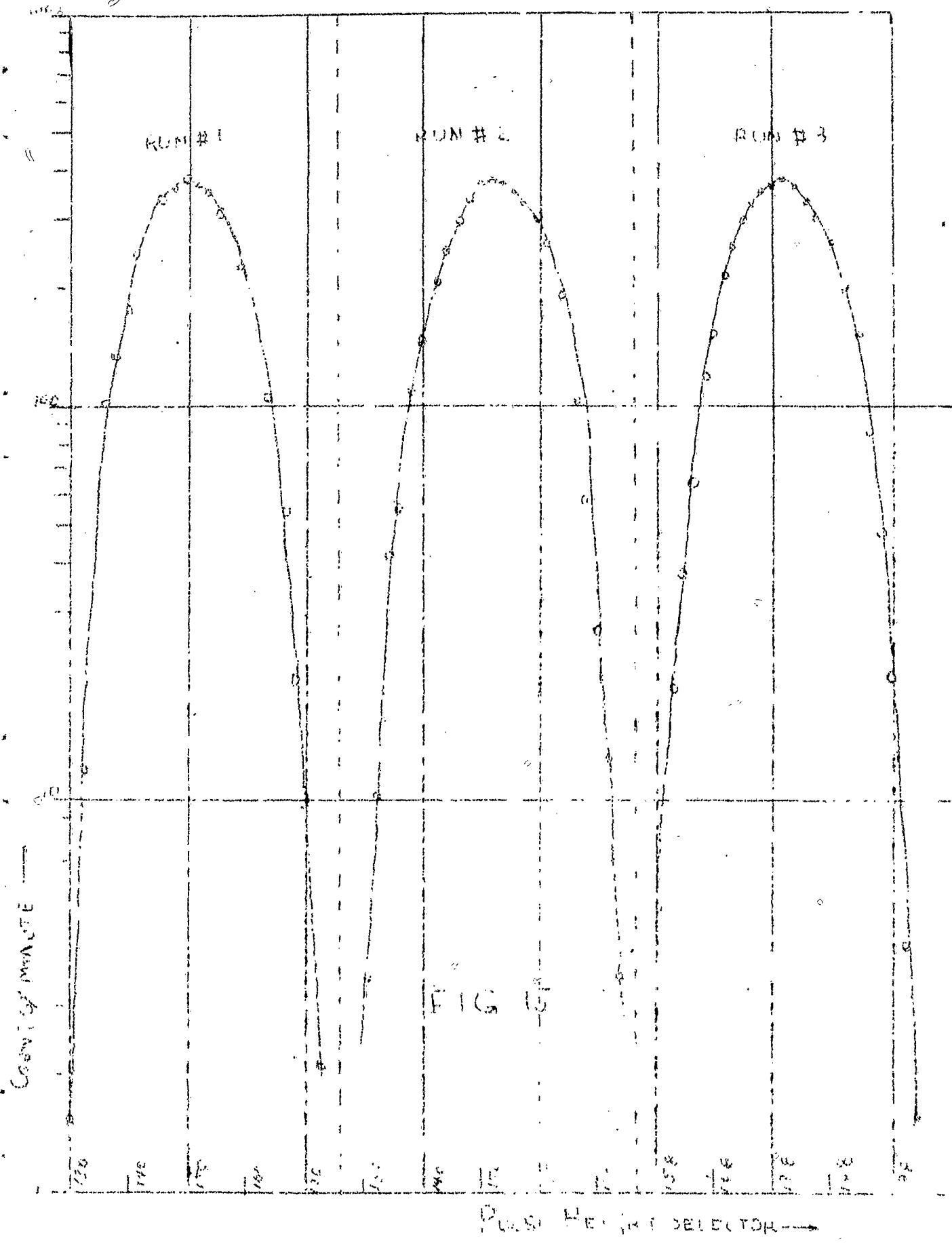
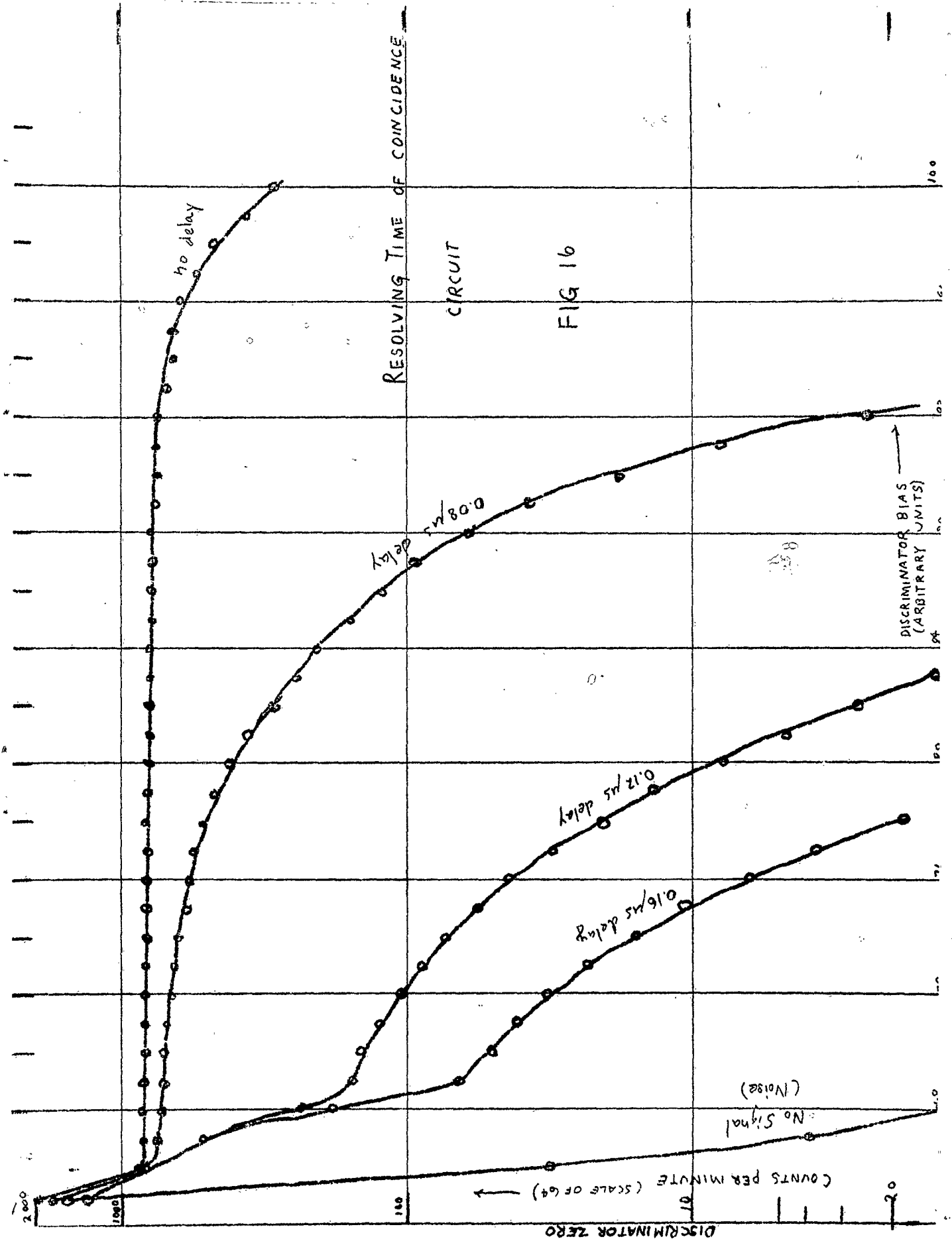


FIG 6



number of losses is set by the lower limit of pulse amplitudes which one wishes to measure.

The circuit does detect the smallest pulses as is evidenced by the plateau curve (Figure 17). Though the pulse amplitude, which is dependent on the 10th power of the applied voltage, varies considerably as the voltage is changed, the counting rate changes very little.

E. Auxilliary Equipment. The high voltage power supply (Figure 18) is designed along conventional lines. The minor modifications appear to result in greater stability and dependability.

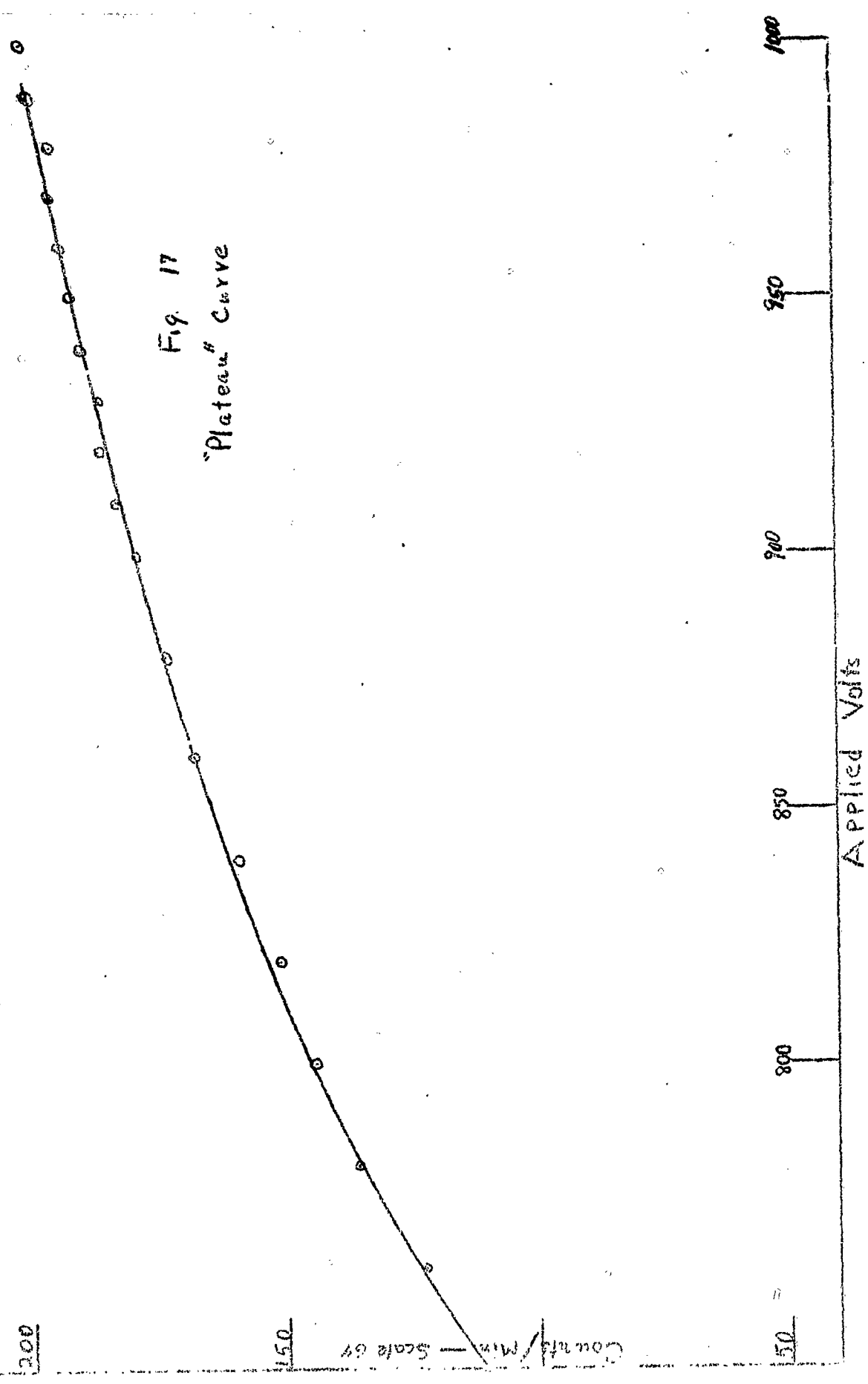
For the measurement of liquid phosphors, a glass cell has been constructed having a length of 4 in. and a diameter of 1.5 in. The ends of the cell are ground flat and parallel and the unit is covered with aluminum foil to minimize light losses. This cell is inserted between the two multipliers being cemented to the light pipes with Canada Balsam.

V. Spectra

Because of limitations of personnel, these experiments were abandoned early in the contract period. There is still considerable interest in the problem, but work on it must be postponed.

VI. Photoconductivity of Solutions

The apparatus for the photoconductivity measurements consists of a vacuum tube electrometer (Figure 19), a fused silica cell, and a source of ultra-violet light.



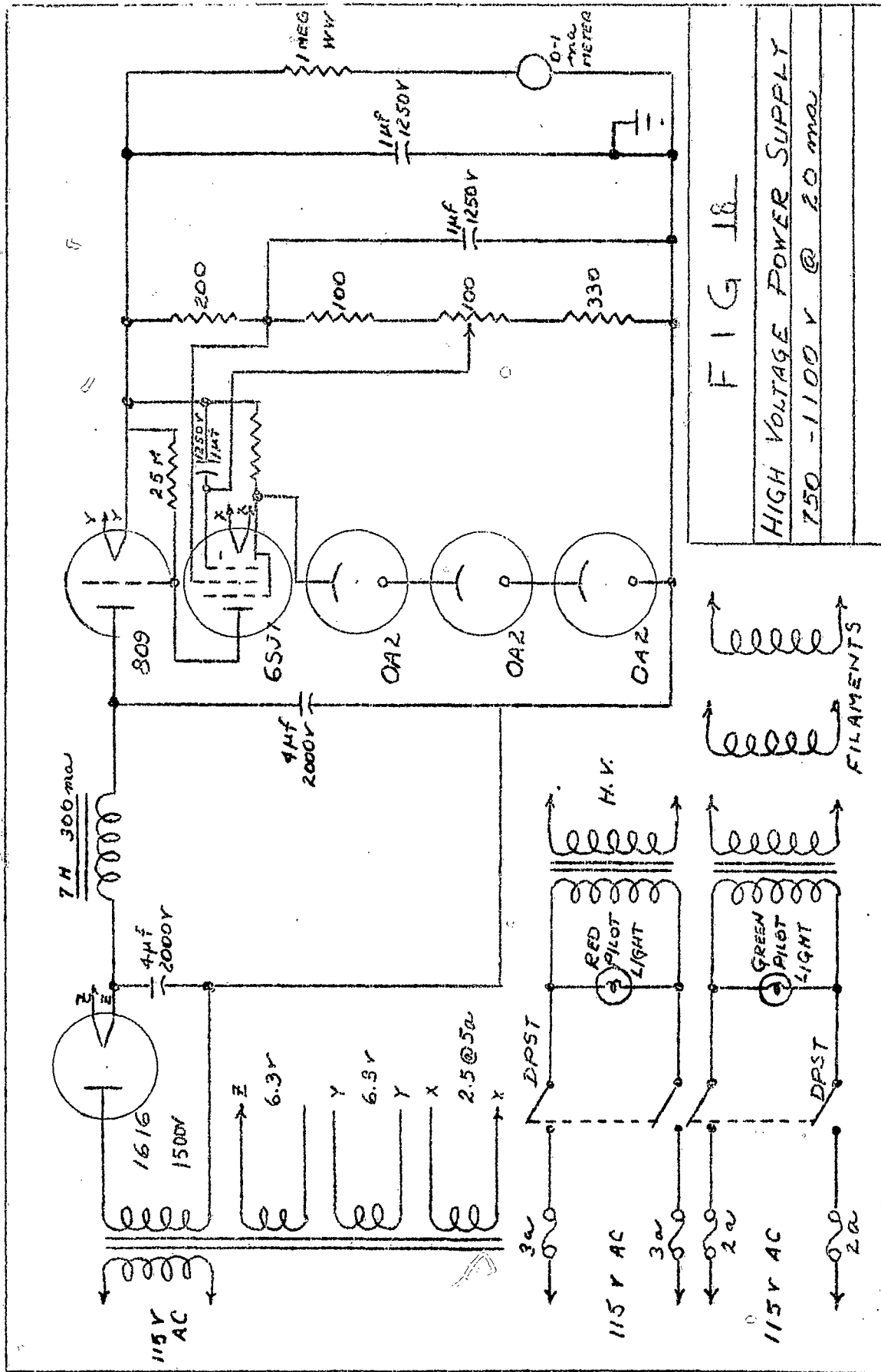


FIG 16

HIGH VOLTAGE POWER SUPPLY
 750 - 1100 V @ 20 ma

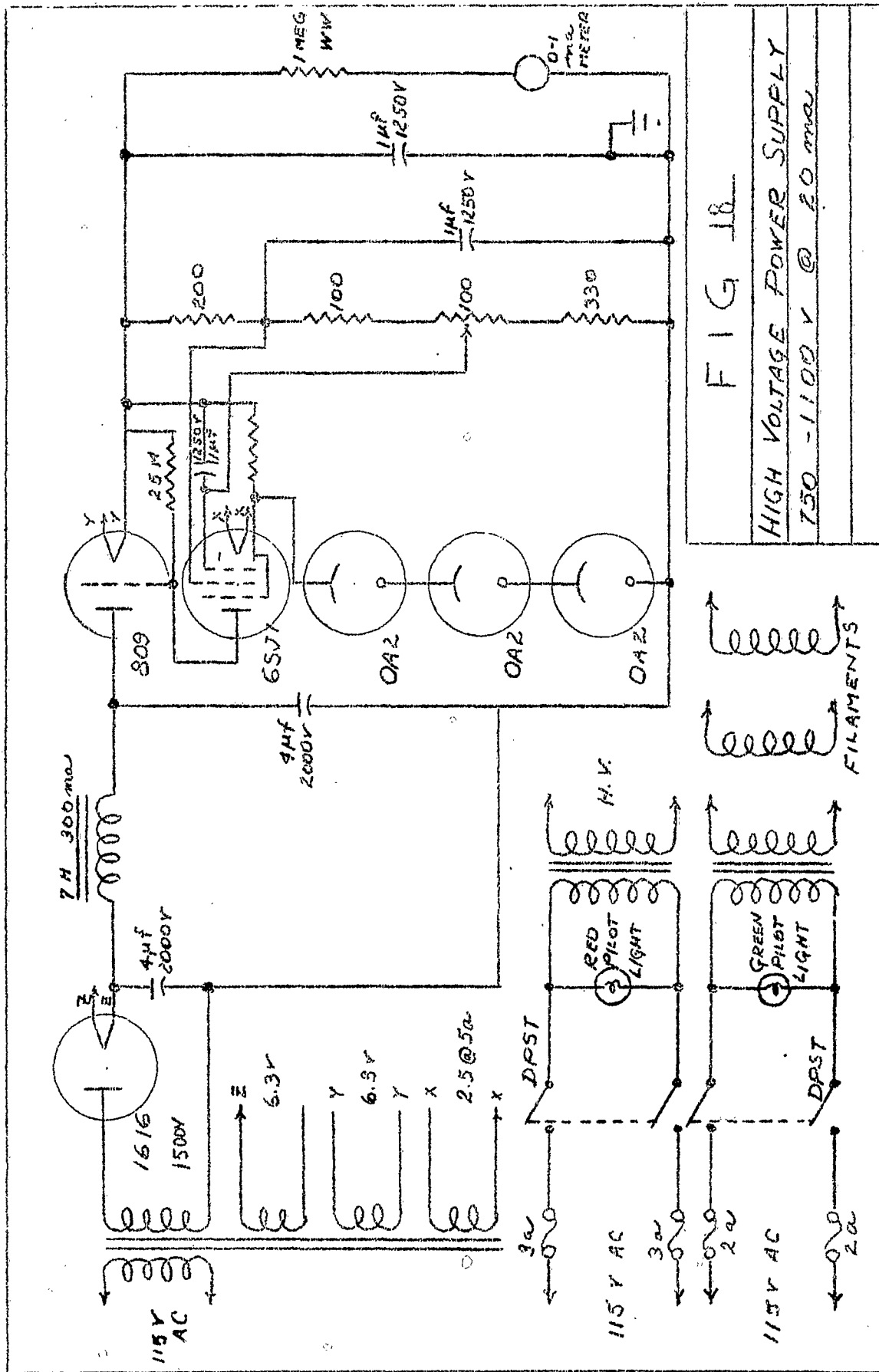
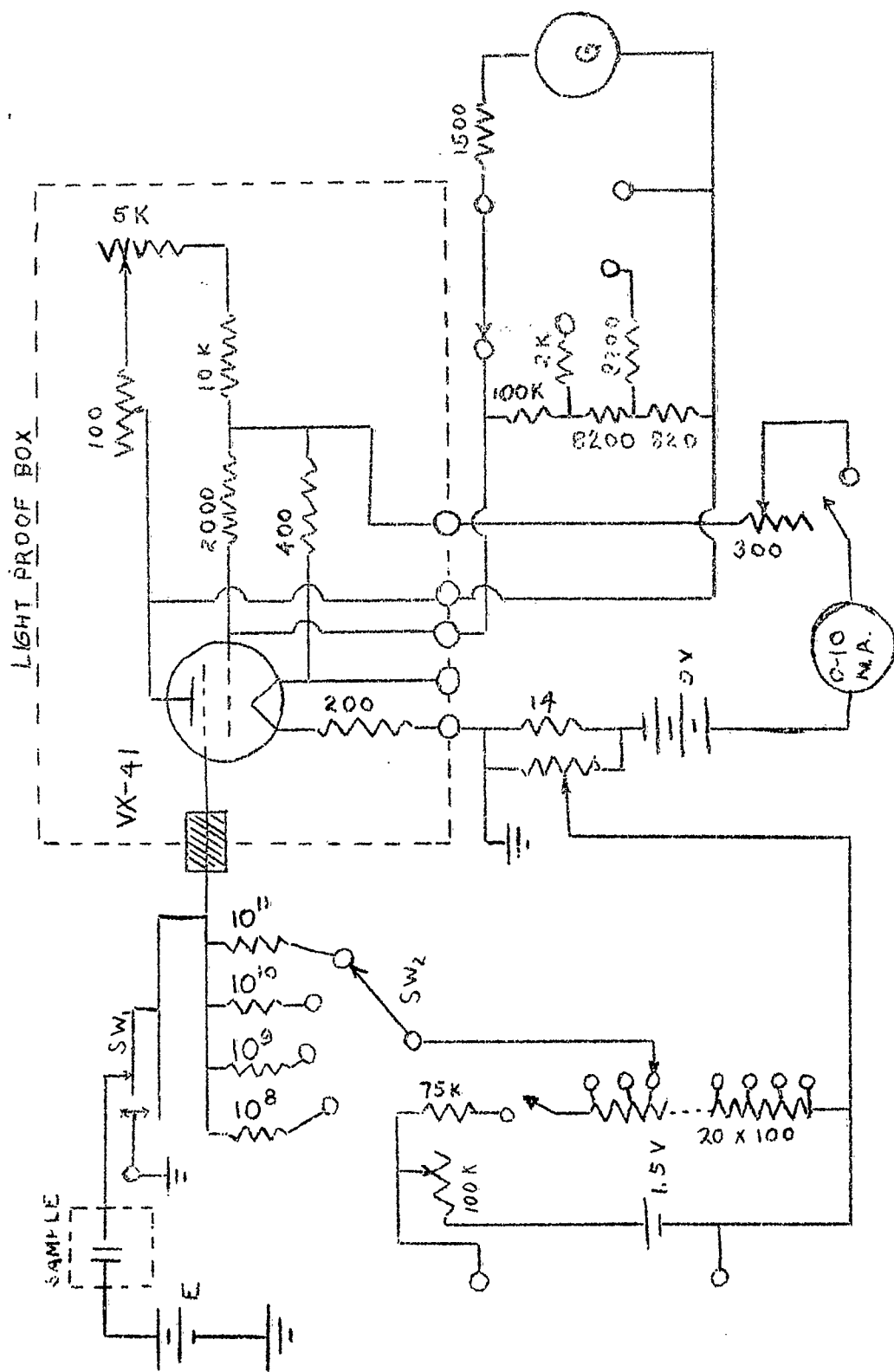


FIG 18

HIGH VOLTAGE POWER SUPPLY

750 - 1100 V @ 20 ma



ELECTROMETER CIRCUIT DIAGRAM

FIG 19

The electrometer, as it is now arranged, is capable of measuring currents of the order of 5×10^{-15} amperes. The measurements cited in reference (1) indicate that the solution of anthracene in hexane that was investigated had a dark current-density of 6×10^{-15} . The current-density upon illumination was of the order of 100×10^{-14} , under a field, in each case, of 500 volts/cm. It was, therefore, concluded that this apparatus would be suitable for carrying out the measurements.

There are, however, some large discrepancies between these early measurements and the data at hand. The International Critical Tables give as the resistivity of pure hexane a value of greater than 10^{18} ohm-cm. The solutions under investigation show a dark resistivity of approximately 4×10^{15} ohm-cm, and there is little difference between this value and the resistivity found here for the purified hexane.

The cell contains two platinum electrodes with a spacing of 2 mm, and an area of 1 cm^2 , reproducing the conditions reported by Volmer exactly. Illumination is by a GE-h4 high pressure Hg arc which has had the glass bulb removed.

Measurements up to this time show no photoconductivity of the order of magnitude reported by Volmer and measurable effects attributable to electrode reactions rather than to true photoconductivity.

The following reasons are being investigated for the lack of photoconductive effect. (a) The light source may be too weak in the ultraviolet component responsible for the photoconductivity. (b) Impurities such as dissolved oxygen may be "quenching" the effect. (c) The measurements of Volmer may be invalid due to the presence of undetermined impurities. This last conclusion is suggested from

the fact that the Volmer solutions had a relatively high conductivity. It is felt that there should be a true photoeffect in these solutions, though the early measurements may have not been a true indication of its extent.

VII. P-N Barrier Counters

This phase of the program is the only one on which there was little construction of equipment since the apparatus had been built previous to the beginning of the contract period.

The investigations involve the measurement of the energy response and the effect of temperature and applied field on the counting characteristics. A complete separate report on this work has been prepared.

VIII. Summary

Work on scintillation and crystal counters during the contract period has been devoted nearly exclusively to the development of suitable instrumentation for carrying out the proposed experiments. The various instruments and their operating characteristics are described in detail.

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