The use of scintillation counters for determining absolute radioactive decay rates by coincidence methods

by Philip Charles Finch

A THESIS Submitted to the Graduate Faculty in partial fulfillment of the requirements for the degree of Master of Science in Engineering Physics
Montana State University

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ABSTRACT

A method of determining the absolute decay rate of a radioactive substance is described, wherein use is made of three counting rates. The difficulties resulting from the application of scintillation counters to this problem are discussed, and methods for avoiding some of these problems are presented. Circuit diagrams of a gain-stabilized photomultiplier circuit, an amplifier circuit, and a coincidence circuit are shown. The results obtained by applying the method to two samples of Co^{60} indicate that reasonable accuracy may be expected, although a definite statement as to the accuracy of the method must await a more complete investigation into the effects of satellite after-pulsing in photomultiplier tubes.

INTRODUCTION

Coincidence counting methods have been used extensively since 1930 as a means of studying cosmic radiations, Compton effect, and in the determination of quantum energies. All of these experiments involve the "simultaneous" triggering of two counting devices by a single particle or quantum. "Simultaneous" events are registered by some coincidence device, and any two events that occur within the resolving time of this device are considered as simultaneous events.

In 1939, Feather and Dunworth (5) began coincidence measurements of a somewhat different nature for a new purpose--the detection of the simultaneous emission of two particles or quanta by an atomic nucleus. A portion of the experiments was directed towards the determination of the absolute efficiencies of the Geiger-Müller counters for various gamma radiations, and towards the determination of the absolute intensity of the sources.

Recent advances in the design of photomultiplier tubes have permitted the scintillation counter to replace the Geiger-Müller counter for many counting purposes. The application of scintillation counters to the problem of determining the absolute decay rate of a Co^{60} source by the Dunworth method is the subject of this report.

DERIVATION OF THE EXPRESSION FOR ABSOLUTE DECAY RATES

Coincidence counting involves registering the detection of two events that occur within a predetermined time interval. Such events are considered to be simultaneous events.[1] In the problem of determining absolute decay rates, the simultaneous events to be detected are the emission of two ionizing particles or quanta by a single nucleus in the process of its radioactive decay. Indicated coincident events may result from one of three different processes. These processes may be described in the following manner:

1. A single particle may pass through both detectors and activate both.
2. The detectors may be activated by separate particles which are the result of a single nuclear decay.
3. The detectors may be activated by particles which have their origins in separate, uncorrelated events that have occurred within the necessary time interval.

Coincidences of the first type are the result of cosmic radiation. The position of the two detectors relative to one another determines to a large extent how significant the error introduced by coincidences of this type may be. By arranging the detectors in a horizontal plane, the

1. The time interval (the resolving time) within which two events must occur to be considered as coincident or simultaneous events may vary within rather wide limits according to the requirements of any problem. Resolving times on the order of 10^{-7} seconds are easily obtainable, and Z. Bay (2) has shown that resolving times as short as 3×10^{-10} seconds are obtainable with the proper choice of equipment.

probability of such an event is reduced to the extent that such coincidences do not introduce an appreciable error.

Coincidences of the second type are the genuine or true coincidences to be investigated.

Coincidences of the third kind are the result of the finite resolving time of the equipment. The error introduced by these chance coincidences is determined by the resolving time of the coincidence circuits and by the activity of the source. In most cases, it is desirable to keep the number of these coincidences at a minimum.

Consider the case in which a nucleus undergoes a radioactive transformation with the simultaneous emission of two particles. Let ϵ_1^{1} denote the total probability that particle number one will be detected by counter number one, and ϵ_2^{1} the probability that particle number one will be detected by counter number two. Let ϵ_1^{11} and ϵ_2^{11} represent the corresponding probabilities for particle number two. These probabilities are dependent upon the solid angles subtended at the source by the two detectors, the efficiency of the detectors, the sensitivity of the electronic circuits involved, the efficiency of the optical coupling, etc. With the probabilities defined as above, the following relationships may be written:

$$N_1 = N\,(\epsilon_1^{1} + \epsilon_1^{11}) \qquad (1)$$

$$N_2 = N\,(\epsilon_2^{1} + \epsilon_2^{11}) \qquad (2)$$

where N_1 represents the counts per second by counter number 1, N_2 the counts per second by counter number 2, and N the number of decays per second.

True coincidence counts may arise in two different fashions. Particle number one may be detected by counter number one and the associated particle number two by counter number two. The probability of this event would be $\epsilon_1^{1}\epsilon_2^{11}$. Particle number one may be detected by counter number two and the associated particle by counter number one. The probability of this event is $\epsilon_1^{11}\epsilon_2^{1}$. The probable number of true coincidence counts is given by the expression:

$$N_c \text{ (coincidence counts)} = (\epsilon_1^{1}\epsilon_2^{11} + \epsilon_1^{11}\epsilon_2^{1})N \qquad (3)$$

If the relative efficiencies of the counters towards the two particles are known, such that:

$$\epsilon_1^{11} = K\epsilon_1^{1} \qquad (4)$$

$$\epsilon_2^{11} = K\epsilon_2^{1} \qquad (5)$$

Then: $$N_1 = N\epsilon_1^{1}(1 + K) \qquad N_2 = N\epsilon_2^{1}(1 + K) \qquad N_c = 2NK\epsilon_1^{1}\epsilon_2^{1} \qquad (6)$$

The decay scheme of Co^{60} involves the emission of a β-particle followed by the emission of two gamma rays with energies of 1.17 Mev. and 1.33 Mev. By screening out the β-particles by means of aluminum shields, the problem of determining the activity of a Co^{60} source is reduced to the problem discussed above. Furthermore, since the two gamma rays are of very nearly the same energy, it may be stated with little error that K, the relative efficiency, is equal to unity. The equations then become: $$N_1 = 2N\epsilon_1^{1} \qquad N_2 = 2N\epsilon_2^{1} \qquad N_c = 2N\epsilon_1^{1}\epsilon_2^{1} \qquad (7)$$

An expression for $N = \frac{N_1 N_2}{2N_c}$ is then obtained.

The above expression for the number of radioactive transformations per second does not take into account the chance coincidences, the background coincidences, or the individual background counting rates of the counters.

The background coincidence rate and the individual background counting rates may be measured directly. The determination of the chance coincidence rate requires a knowledge of the resolving time of the coincidence circuits. Bleuler and Goldsmith (4) and Barnothy and Farro (1) have outlined a method for determining the resolving time, wherein the two counters are isolated and set to counting radiations from uncorrelated sources. The resulting coincidence counts are chance coincidences only, and the resolving time may be calculated from the expression $N_c = 2N_1N_2\tau$, where τ represents the resolving time of the equipment. If the following notation is used:

n_1 = background counts on counter number one (8)

n_2 = background counts on counter number two (9)

n_c = background counts on coincidence counter plus chance coincidence counts. (10)

The expression for the absolute decay rate then becomes:

$$N = \frac{(N_1 - n_1)(N_2 - n_2)}{2(N_c - n_c)} \qquad (11)$$

THE CHOICE OF THE PHOTOMULTIPLIER TUBE AND PHOSPHOR

Scintillation counters are no newcomers to the field of particle counting. Crookes and Regener introduced the technique of visual scintillation counting in 1908, and this type of counter has played an important role in establishing the nature and charge of the α-particle.

The fact that the scintillations must be detected by the eye of a human observer places serious limitations on the above technique, however. The rate of counting is limited to about 60 scintillations per minute. In addition, only α-particles can be detected by the counter, since the weak ionizations of β-particles and γ-rays do not produce scintillations of sufficient intensity to be seen by the human observer. Because of these limitations, the gas-ionization counter had largely replaced the visual scintillation counter by the late 1930's.

The years since 1947 have seen a tremendous revival of interest in the possibilities of the scintillation counter. The advances in design of reliable photomultiplier tubes that are sensitive to very small amounts of light have eliminated the necessity of the human observer and have extended the use of the scintillation counter to the study of the weakly ionizing β-particles and γ-rays.

Of the photomultiplier tubes developed in recent years, the tube that has found the widest application in scintillation counting work in this country is the RCA type 5819. The tube was developed especially for scintillation counting purposes and combines a number of desirable characteristics, two of them being a very large light sensitive surface and a comparatively low noise level. Morton (9) and Birks (3) list detailed information on the tube. Two of these tubes were used in the equipment constructed for the present problem, the determination of the decay rates of two samples of Co^{60}.

The development of highly sensitive photomultipliers has made possible the use of many substances as phosphors. The zinc sulfide preparations used as phosphors in the visual scintillation counters have many disadvantages, some of the most serious being a long decay time for the light flash (10^{-5} seconds or longer) and a low transparency to the emitted light.

Sodium iodide crystals, activated with thallium, were used as phosphors in this work. The NaI (Tl) crystal possesses a number of properties that combine to make it a very useful phosphor. The light yield is the best of the known phosphors with the exception of ZnS, and while its decay time (about 2.5×10^{-7} seconds) is not as fast as some of the organic phosphors, it is still suitable for most counting work. The crystal has been shown by Hofstadter and Milton (7) to be very transparent to its emission spectrum, making possible the use of large crystals. The crystals are deliquescent, however, and must be used in air-tight surroundings. These air-tight surroundings are provided for in the case of the Harshaw crystals[2] by surrounding the crystal on three sides by thin aluminum and on the fourth by a transparent plastic. The "canned" crystals were taped directly to the end of the photomultipliers, with a thin

2. The crystals are the Harshaw Scintillation Crystal, serial number 292, type 16.

film of mineral oil included to improve the optical coupling of the apparatus.

THE ELECTRONIC CIRCUITRY

The Photomultiplier and Preamplifier Circuit

The most frequent cause of instability in scintillation counters is the dependence of photomultiplier gain upon the applied voltage.[3] Unless suitable precautions are taken, any variation in applied voltage is accompanied by a large change in the counting rate.

To date there exist two methods of stabilizing scintillation counters. The first involves the use of extremely well regulated high voltage supplies with output voltages stable to about 0.01 per cent for short periods of time (hours) and 0.1 per cent for periods of days. Such stable supplies are very difficult to obtain, either commercially or by construction.

A much simpler solution to this problem is described by Sherr (10). A plateau is introduced in the gain versus high voltage characteristics of the RCA 5819 tube as a result of stabilizing the voltage between the fifth and sixth dynodes of the tube. In the circuit described by Sherr this is accomplished by means of a voltage regulator tube. Green and Paul (6) indicate that while this circuit has excellent gain characteristics, the resulting current drain on the high-voltage supply is excessive. The same effect may be obtained by the circuit of figure 1, in which the required stabilization is obtained through the use of batteries. The gain characteristics of this circuit show a slight improvement over those

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3. Green and Paul (6) state that the gain of the RCA 5819 tube is dependent upon the 8th power of the applied voltage.

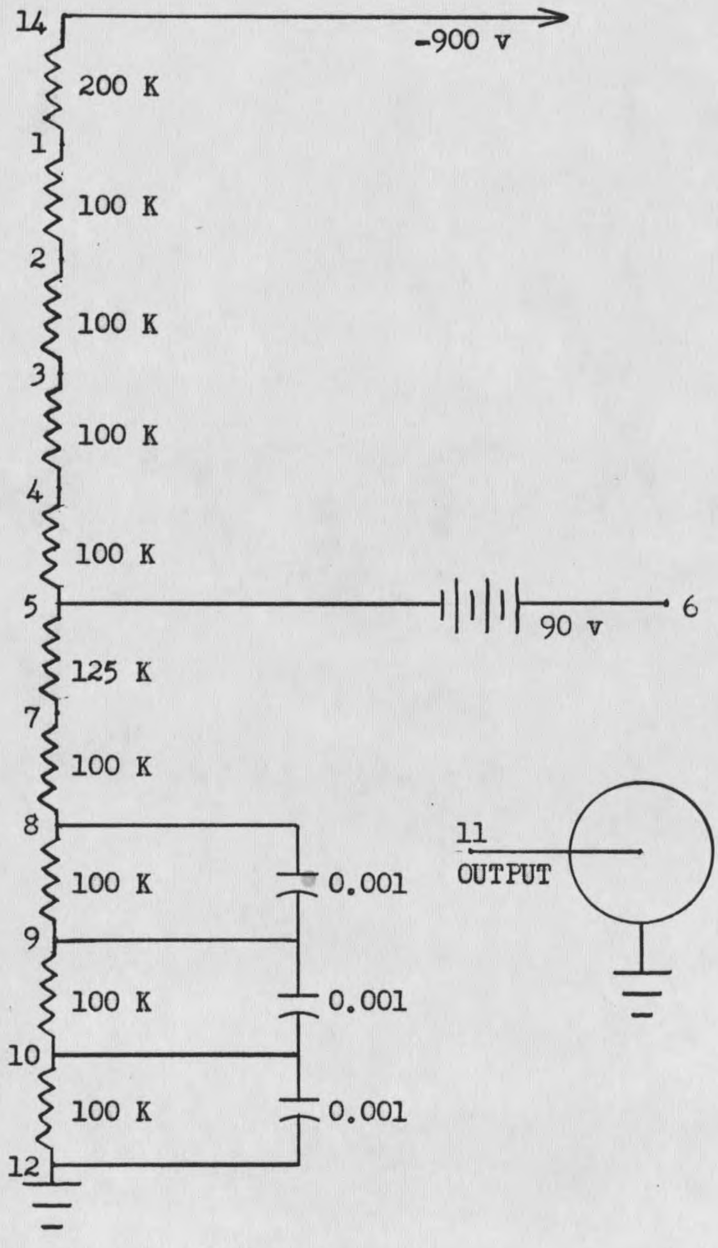

Figure 1

A Schematic Diagram of the Gain-Stabilized Photomultiplier Circuit of the original VR tube circuit. A flat plateau extends over the range from 880 volts to 980 volts.

No theoretical basis for the existence of this plateau is offered either by Sherr or by Green and Paul. An earlier article by Morton (9) mentions the effect and attributes it to the possible occurrence of a focusing loss at the sixth dynode. The effect of the focusing loss is more pronounced at higher voltages, and partially compensates for the increased gain that accompanies higher tube voltage in a standard photomultiplier circuit.

Two of the circuits of figure 1 were constructed for the purposes of this experiment. While no extensive gain studies were carried out on these circuits, an optimum high voltage value was obtained by conducting a "counts versus high voltage" test for each photomultiplier and its associated preamplifier and amplifier. The results of these tests are shown in figure 2 and Table I.

In the process of these tests, the high-voltage measurements were made with a Triplett multimeter, Model 625-N, 10,000 ohms per volt. The meter was placed directly across the output of the high-voltage supply and remained an integral part of the circuit throughout the conduction of these and all other tests.

Figure 3 shows the complete schematic diagram for the photomultiplier and preamplifier unit, and figure 4 shows the diagram for the amplifier and coincidence unit.

The cathode follower preamplifier provides the necessary impedance

TABLE I

Results of Tests on Plateau Region in Gain Characteristics of Photomultiplier Tube

Volts	Counts, Scaler No. 1 Divided by 64	Counts, Scaler No. 2 Divided by 64
820	37	65
840	108	98
860	153	129
890	163	137
910	167	137
930	174	126
940	169	127
955	231	121

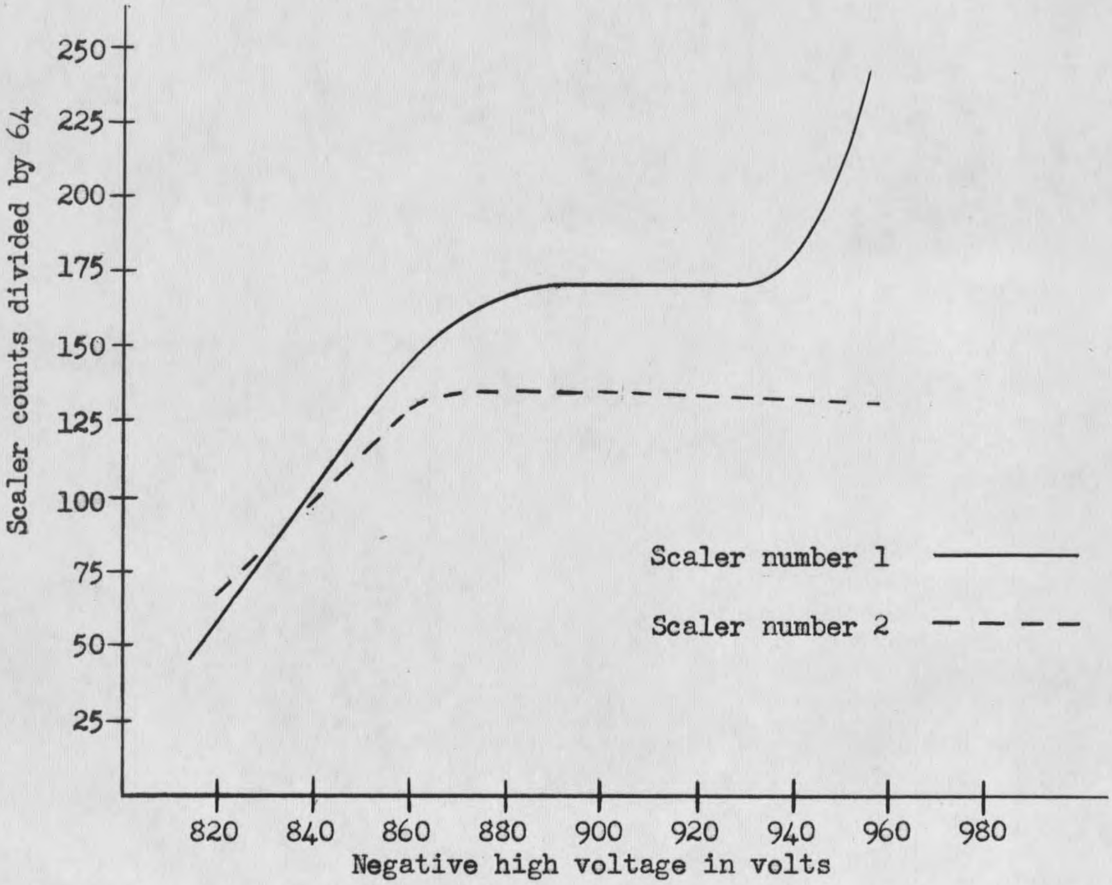

Figure 2. A plot of scaler counts vs. negative high voltage.

match between the photomultiplier and the coaxial cable used to couple to the main amplifier.

The Amplifier and Coincidence Circuit

The amplifier consists of a series of 12AU7 twin triodes and incorporates the principle of direct coupling throughout. The schematic diagram for this amplifier was furnished by Mr. W. Farrand of Richland, Washington. Complete information as to its performance was not available at the time and has not been published since. However, the amplifier was reputed to possess excellent discrimination control and stability.

The two amplifier units constructed by the author did not give the performance that had been anticipated. The 50-ohm potentiometer which supposedly provides the discrimination control had no discernable effect on the counting rate. The counting rate was very dependent upon the supply voltage, with higher counting rates occurring at lower supply voltages. In addition, a pronounced drift in counting rate appeared in one of the units, although the other could be depended upon to provide consistent results. The reputation of the amplifier is well established however, and these unfortunate characteristics can only be attributed to faulty construction and/or faulty components, although thorough tests on the components indicated that they were in good condition.

The nature of the output pulses from the two amplifier stages (very uniform pulse amplitude, sharp leading and trailing edges, 50 volt amplitude, less than 1 micro-second pulse width) was felt to be sufficient advantage to warrant the use of the amplifiers. Such uniform, high-voltage pulses made the design of the coincidence network a simple matter.

The coincidence circuit used was of the Rossi type, using a twin-triode 6J6 tube. The action of the circuit may be described as follows. A signal (negative) from one of the amplifiers will cut off one-half of the tube. The resultant tendency for a reduced current through the common-cathode resistor, R25, reduces the grid to cathode voltage for the other section of the tube. The conduction of this half of the tube then increases, and the effect is one of negative feedback for a signal from only one amplifier. If, however, simultaneous signals from both amplifiers are present at the grids of the coincidence tube, both halves are cut off and a large negative-going pulse appears across the cathode resistor R25 (figure 4).

The ratio of the output resulting from coincident pulses to that resulting from non-coincident pulses for the circuit shown in figure 4 is 2.5/1. While this value does not compare with the 10/1 ratio obtainable from some Rossi-type circuits, the use of additional discriminating stages has increased the ratio to 25/1. Coincident pulses result in a negative-going pulse across resistor R34 of 125 volts, whereas non-coincident pulses result in signals of only 5 volts. The output from the coincidence circuit is reduced by resistors R35 and R36 to a value consistent with the requirements of the scaler units. The outputs from the amplifiers are reduced in the same fashion.

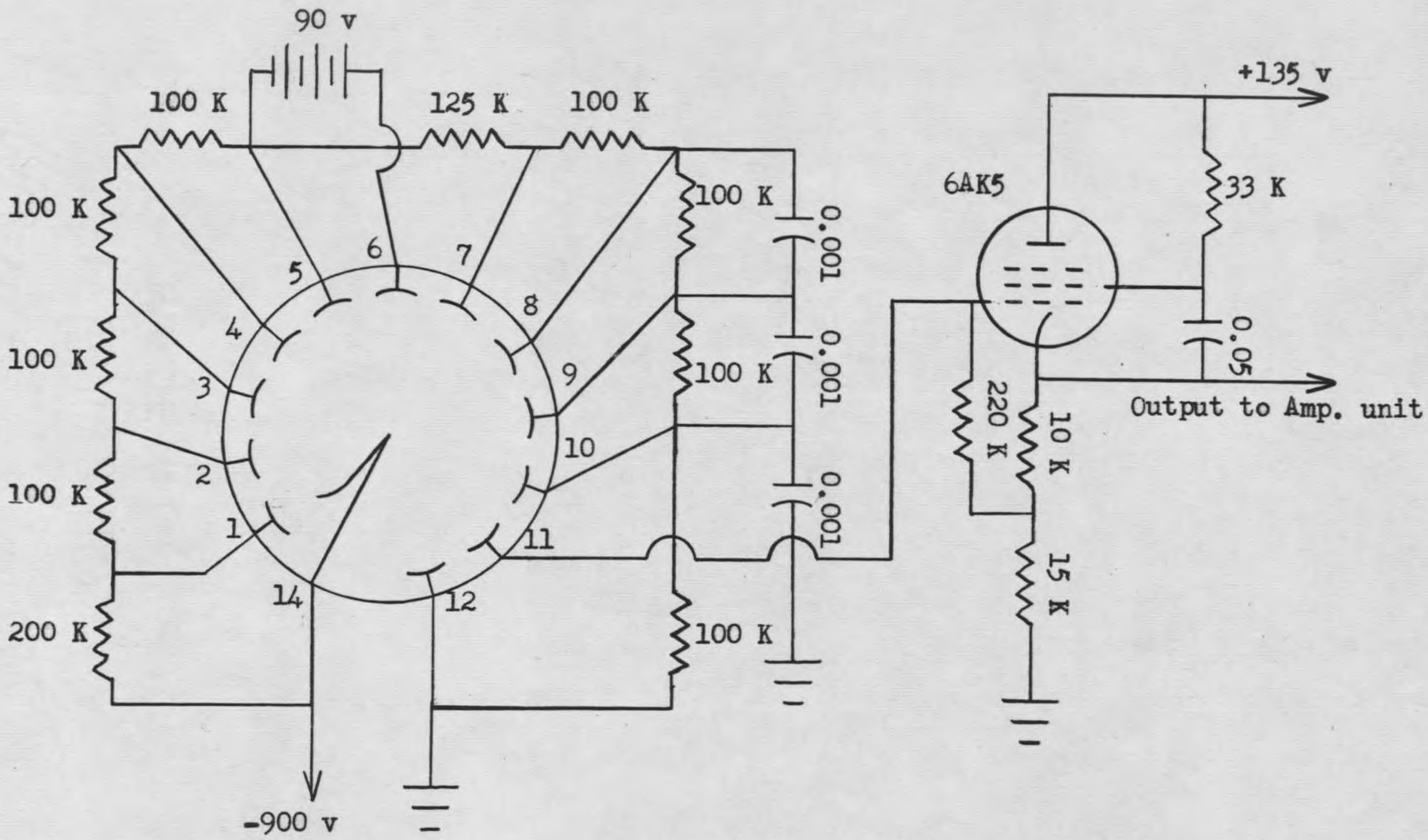

Figure 3. Schematic Diagram of the Photomultiplier and Preamplifier Circuit

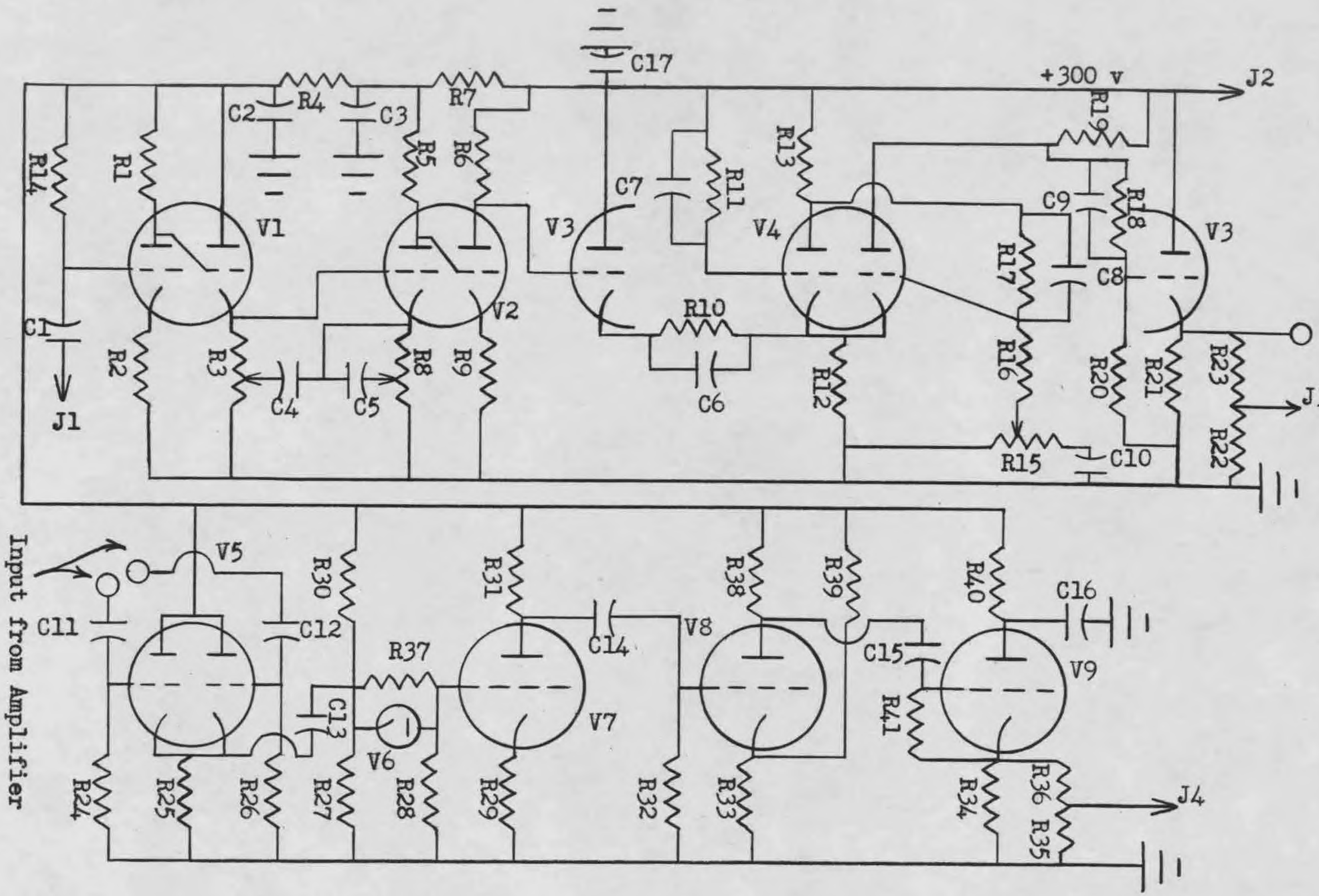

Figure 4. Schematic Diagram of the Amplifier and Coincidence Circuits