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A low frequency phase shift method

Ronnie Carroll McMillan

A LOW FREQUENCY PHASE SHIFT METHOD

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Approved by

Richard Anderson (advisor) Otto S. Hill

P. L. Loeffler, Jr.

ABSTRACT

The phase shift method of measuring atomic lifetimes is particularly useful because it allows us to make measurements in the steady state mode instead of in the transient mode. High frequency phase shift techniques cause many problems. The equipment, which was designed and built to make low frequency phase shift techniques possible, is shown schematically and discussed. The new experimental techniques employed in our apparatus are a phase multiplier and a unique method of using a photomultiplier as a mixer. In the photomultiplier the final dynode is used as the grid of a triode. The next to the last dynode may be thought of as the cathode of a triode normally used as a mixer. Using the photomultiplier in this fashion, we have a low noise, extremely efficient mixer. The phase multiplier is essential to accurately measure small phase angles. The problems of high frequency techniques are cited and references given.

The speed of light was measured as a test of the equipment. The experimental values resulted in a .95 confidence interval containing the generally accepted value of the speed of light in air. The average value measured was $(2.937 \pm .033) \times 10^8$ meters per second, where .033 is the standard deviation for 48 trials. This average value

represents an error of 1.61%. From the results we have established the feasibility of this low frequency method of measuring atomic lifetimes.

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INTRODUCTION

In order to study the lifetime of certain unstable atomic states of a given atom, these states must be populated by external excitation. In the present studies a particular desired state was optically pumped by radiation of the appropriate wavelength. By modulating the pumping radiation we may measure how long a state remains excited by monitoring the phase difference of the pumping and emitted light. Lifetime measurements of excited states employing the phase shift method have been reported by Brewer[1], Link[2,3], Tummerman[4], Bailey and Rollefson[5], Gottling[6], Lawrence[7,8], Hesser and Dressler[9], Ware[10], Muller[11], Hesser[12], and Pink and Welge[13].

For states whose lifetimes are very short this method requires a pumping light modulated at a very high repitition rate (greater than 10^6 kc). The reason, of course, is that the phase difference between the pumping radiation light and emitted light will be very small for pumping that is modulated at a lower rate[1].

Modulating the light at a high frequency often causes troublesome RF pick-up problems[2,3]. Moreover, in instances where an electro-optic light modulator is used as the light modulator via the Pochels-effect, high modulation frequencies heat the crystal and force one to adopt elaborate

cooling methods[11].

By using phase multiplication, one is able to use much lower modulating frequencies, thus avoiding the above mentioned shortcoming of the phase shift method.

The equipment used in this experiment was designed for measuring atomic lifetimes. This equipment was tested by measuring the speed of light as has been done by others[1,2]. The speed of light experiment is an excellent test of the quality of the equipment used.

THEORY

Associated with modulated light is some phase angle. If we view the light at a certain place, we will observe a waveform with a phase angle which we can arbitrarily call zero. Further down the light path we will observe the same waveform but this one will lag the other by an angle depending on the distance between the observations. This can be seen as follows.

Consider two waveforms of the same period. One lags the other by a phase angle ϕ. The relationship of this phase lag to the time lag is

$$\frac{\phi}{t} = \frac{2\pi}{1/f} = 2\pi f,$$

where f is the frequency of the waveform, t is the time, and ϕ is the phase angle.

Solving for the phase we have

$$\phi = 2\pi ft = 2\pi f \frac{d}{c} = \frac{360° fd}{c},$$

where d is the distance between observations, and c is the speed of light. ϕ is measured in degrees. Now, we have

$$c = \frac{360° fd}{\phi}.$$

Knowing the frequency of the modulated light and the distance between the two observation points and measuring

the phase angle we can make a calculation of the speed of light.

If we optically pump an atomic state with modulated resonant light, we will observe a fluorescence which is produced by the excited atoms as they decay back into the ground state. This fluorescence is modulated at the same frequency as the pumping radiation. By monitoring the pumping light (which is arbitrarily said to have zero phase) at a point after the modulation of the light and the steady state emitted fluorescence at the same time, we observe a difference in phase angle between the two signals.

This phase angle is associated with the apparent lifetime of the state and an inherent instrumental phase shift. The inherent instrument phase shift correction is obtained by replacing the absorption cell with either a mirror or the absorption cell filled with a colliodal suspension scatterer, while maintaining all other conditions the same as in the lifetime experiment. The scatterer is preferred to mirrors because of reflection phase shifts at the surface in the latter case.

The monitored apparent lifetime τ may not be the true lifetime of the excited state because of resonant imprisonment. When an excited atom decays by emitting radiation of a certain wavelength, this radiation may be absorbed by an unexcited atom and subsequently reemitted. This process of

absorption and reemission continues until the emitted light can escape from the cell. By making lifetime measurements at several pressures and then extrapolating back to zero pressure, we may obtain the true lifetime of the state. However, this extrapolation must be performed over the region in which the fluorescent intensity varies linearly with pressure because at higher pressures radiation imprisonment may predominate. At lower pressures diffusion mechanisms bias the lifetime measurements. Diffusion effects are seen at low pressures because the excited atoms will sometimes collide with the cell walls causing the atom to prematurely decay. This means we will measure an apparent lifetime which is shorter than the true lifetime.

If the resonant imprisonment effects are negligible, instrument phase corrections have been made, and the lifetime of the excited state is short enough so that diffusion effects can be neglected, then the relationship between the phase angle and the apparent lifetime of the excited state is given by

$$\phi = \tan^{-1} \omega\tau$$

This can be seen by the following argument.

Consider two atomic states. The j-th state is the unexcited state and the i-th state is the excited state.

Let N_i be the number of excited atoms in state i at time t, B is the Einstein absorption coefficient for excitation from state j to state i, in units of m^3 $\rho(t)$ is the incident photon density of frequency ν, in units of $\frac{1}{m^3 t}$, τ is the lifetime of the i-th state and $\frac{1}{\tau} = \sum_k A_{ik}$, where k are states of lower energies than i. $B\rho$ is the probability that the i-th state will be excited and has the units of reciprocal time.

The rate equation of the process is

$$dN_{i/dt} = N_o \rho\ B - N_{i/\tau}$$

where N_o is the number of unexcited atoms.

Let us assume that the optical pumping light is modulated so that

$$\rho(t) = \rho_M \cos\ \omega t,$$

where ρ_M is the maximum number of incident photons of frequency ν, $\omega = 2\pi f$ is the angular frequency of modulation, and f is the frequency of modulation.

Now we must solve the following first order differential equation (inhomogeneous):

$$dN_i/dt = BN_o\ \rho_M \cos\ \omega t - N_{i/\tau}$$

Let $K = BN_o \rho_M$. Then we have,

$$dN_i/dt = K\cos\ \omega t - N_{i/\tau}$$

The reduced equation is

$$dN_i/dt + N_{i/\tau} = 0$$

whose solution is

$$N_i = C \exp(-t/\tau)$$

which is also the complementary function of the original inhomogeneous equation. Now we find a particular solution of the inhomogeneous differential equation and the sume of the complementary and the particular solutions will be the general solution we are seeking.

Using the integration constant $e^{t/\tau}$, we have

$$\frac{dN_i}{dt} e^{t/\tau} + \frac{N_i}{\tau} e^{t/\tau} = d(N_i e^{t/\tau})/dt = ke^{t/\tau}\cos \omega t.$$

Separating the variables and integrating we get,

$$N_i = e^{-t/\tau} \int e^{t/\tau} k \cos \omega t \, dt.$$

From the integral tables

$$\int e^{t/\tau} \cos \omega t \, dt = \frac{e^{t/\tau}(\frac{1}{\tau} \cos \omega t + \omega \sin \omega t)}{(\frac{1}{\tau})^2 + \omega^2} .$$

Hence,

$$N_i = \frac{k(\frac{1}{\tau} \cos \omega t + \omega \sin \omega t)}{(\frac{1}{\tau})^2 + \omega^2} .$$

Defining

$$\frac{1}{\tau} = \cos \phi, \ \omega = \sin \phi, \ \tan \phi = \omega\tau$$

and using the trigonometric relation

$$\cos(\omega t-\phi) = \cos\phi\cos\omega t + \sin\phi\sin\omega t$$

we get

$$N_i = \frac{K\cos(\omega t-\phi)}{(\frac{1}{\tau})^2 + \omega^2} \quad , \quad \phi = \tan^{-1}(\omega\tau).$$

The general solution of our differential equation is

$$N_i(t) = Ce^{-t/\tau} + \frac{BN_o\,\rho_M\cos(\omega t-\phi)}{(\frac{1}{\tau})^2 + \omega^2} .$$

Let τ be the natural lifetime of the excited state i. τ is less than 10^{-5} sec. Hence, the transient term in our solution, $Ce^{-t/\tau}$, goes quickly towards zero and can be neglected after a very short time. Hence,

$$N_i(t) = \frac{BN_o\,\rho_M\cos(\omega t-\phi)}{(\frac{1}{\tau})^2 + \omega^2}, \quad \phi = \tan^{-1}(\omega\tau)$$

In consequence, a measurement of the steady state phase difference between incident and reemitted light provides us with the lifetime of the i-th state by means of the simple relation.

$$\phi = \tan^{-1}(\omega\tau).$$

EXPERIMENTAL PROCEDURE

For the speed of light measurements the equipment was arranged as indicated in Fig. 1. The electro optic light modulator polarizers, and 1/4 wave plate were aligned as explained later. The crystal was modulated at 510 kc with a 1000 volt peak to peak sine wave. A light signal from a laser was sent through the modulator and was reflected from a good quality front surfaced mirror and collected into a 9558Q EMI photomultipler. It was mixed in the photomultiplier with a 502 kc signal to obtain an 8 kc signal containing the phase information . This signal was amplified and filtered and then multiplied by 9 in the phase multiplying system. A reference 8 kc signal from the 8 kc oscillator was multipled by 8 and then mixed with the 72 kc signal containing the phase angle 9ϕ to obtain an 8 kc signal with phase angle of 9ϕ. This was then sent into the signal channel of a PAR Lock-in amplifier model JB-4. A reference 8 kc signal from the 8 kc oscillator was sent through the phase shift generator into the reference channel of the lock-in amplifier. The signal amplitude was monitored and set to 4 volts peak to peak and the phase angle dialed to zero at the lock-in amplifier. The reading on the phase shift generator was recorded. Next, the mirror was moved a known distance.

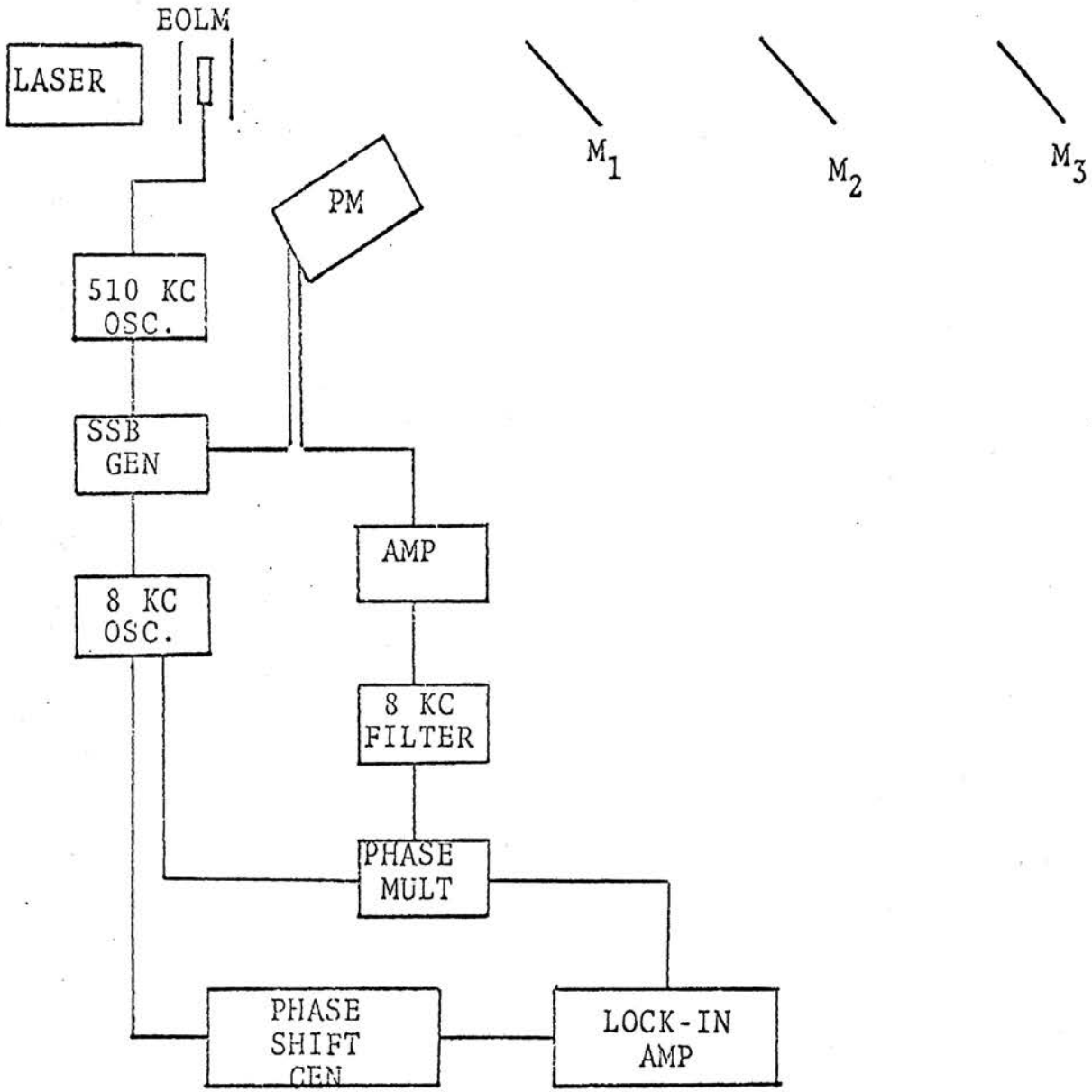


Fig. 1 Experimental Arrangement

The signal amplitude was again set to 4 volts peak to peak and this time the phase was zeroed by the phase shift generator. This reading on the phase shift generator was recorded. The values of the phase angles were calculated and substracted giving the phase shift from which the speed of light can be calculated.

When the lock-in amplifier is used as a phase detector, its output is proportional to the cosine of the phase difference between the signal and the referece inputs. This can be seen by the following argument. A sine wave is sent into the signal channel of the lock-in amplifier. This signal goes through a selective amplifier and then into the phase sensitive detector which is a product device (a balanced mixer). This signal we will call

$$S' = A' \cos(\omega t + \phi),$$

where A' is the amplitude, ω is the angular frequency, and ϕ is the phase angle associated with a position of the mirror. At the same time a reference signal is sent into the reference channel of the lock-in amplifier. This signal passes through a phase shift generator and thus into the phase sensitive detector. This signal we will call

$$S'' = A'' \cos(\omega t + \alpha),$$

where A" is the amplitude, ω is the angular frequency

which is the same as for S', α is the phase angle of this signal and is variable. The phase sensitive detector, being a product device, mixes the two signals giving a signal of the form

$$S''' = S'S'' = A'A'' \cos(\omega t+\phi) \cos(\omega t+\alpha)$$

Using a trignometric idenity, this is

$$S''' = \frac{A'A''}{2} [\cos(2\omega t+\phi+\alpha) + \cos(\phi-\alpha)]$$

This signal is sent through a low pass filter and only the DC term, $\cos(\phi-\alpha)$ survives. Now, we have finally,

$$S^{IV} = \frac{A'A''}{2} \cos(\phi-\alpha).$$

With the mirror in the first position, as explained above, we set $S^{IV} = 0$. That is, we vary α so that

$$\alpha_1 = \phi_1 - \pi/2$$

With the mirror moved to the second position we again set $S^{IV} = 0$ with the calibrated phase shift generator as explained above. Now we have

$$\alpha_2 = \phi_2 - \pi/2$$

where α_1, α_2, and ϕ_1, ϕ_2 are the phase angles associated with the reference and signal inputs respectively corresponding to mirror positions one and two. We are interested in the phase difference $\alpha_1 - \alpha_2$ or $\phi_1 - \phi_2$. It is

$$\alpha_1 - \alpha_2 = \Phi = \phi_1 - \phi_2 ,$$

which gives the phase shift associated with the known light path difference.

In order to modulate the light, a Baird-Atomic-Electro-Optic Light Modulator (E.O.L.M.) model JW-1 was used and it modulated the light from a University Laboratories 1/4 milliwatt model 240 He-Ne laser. The basic unit of the E.O.L.M., which can be considered a crystalline analogue of the Kerr cell, consists of a Z cut (001) plate of potassium dihydrogen phosphate (KDP). The crystal plate is placed between electrodes which allow light to pass in the same direction as the applied electric field. For normally incident collimated light the unit has the properties of a polarization retardation plate, with the magnitude of its retardation directly proportional to the applied voltage. On placing the crystal unit between polarizers, a light beam can be intensity modulated in accordance with the voltage applied to frequencies well beyond the video region.

By placing a 1/4 wave retardation plate adjacent to the crystal, the transmission is biased to 50% of the maximum. Now signals of irregular shape are reproduced linearly.

The maximum transmission of the E.O.L.M. is determined by the transmission of the crystal and the two polaroids and by reflection losses. Since the transmission

of the parallel polaroids is about 35%, the maximum for the E.O.L.M will be 10-25% depending on the wavelength and the number of reflection surfaces in the optical system[14].

The minimum transmission will be determined by the efficiency of the crossed polarizers and the angular field used. Since the crystals are naturally birefringent, there will be some retardation, and therefore transmission, for light that is not passing exactly along the optic axis. For a 100/1 ratio between maximum and minimum transmission the angular field must be limited to a few degrees. In other words the more divergent the beam through the crystal the smaller the percent modulation.

For maximum operating efficiency the E.O.L.M. should be operated at around 2000 volts peak to peak[14]. However, we were not capable of that voltage at 510 kc. Therefore, since we were already in a situation where the percent modulation could be bad, we could not afford to use a beam that was very divergent. A University Laboratories 1/4 milliwat model 240 He-Ne laser was therefore obtained allowing us to modulate the beam with a 50% efficiency.

The alignment procedure for the modulation system is extremely critical. Almost perfect alignment is required to maximize the percent modulation. It is necessary to orient the polarizing axes of the polaroids parallel to

the sides of the square crystal plates. Placing a piece of white paper in the light beam after the second polarizer, we observe the characteristic polarization interference pattern of a uniaxial crystal, a black cross defining the polarizer axes and a series of concentric colored rings. This enables one to determine whether the colliminated light is parallel to the optic axis of the crystal (source centered in the pattern), and whether the size of the source is such as to be within the useful angular field of the E.O.L.M. Further increase of the source area will not increase the amount of modulated light, there will be a reduction in modulation percentage.

When inserting the 1/4 wave plate into position, its principal axes of refraction are to be oriented along the diagonals of the crystal plate, i.e., at 45 degrees to the polarizer axes. The 1/4 wave plate axes are located by observing extinction between crossed polarizers of known axes. If the polarizer's axes are not marked, they can be determined by observing the angle at which crossed extinction is not affected by revolving the polarizer 180 degrees about a vertical axis.

A 510 kc oscillator and power amplifier was built to operate the E.O.L.M. as shown in Fig. 2. A 1000 volt peak to peak sine wave was produced which allowed 50% modulation of the laser beam.

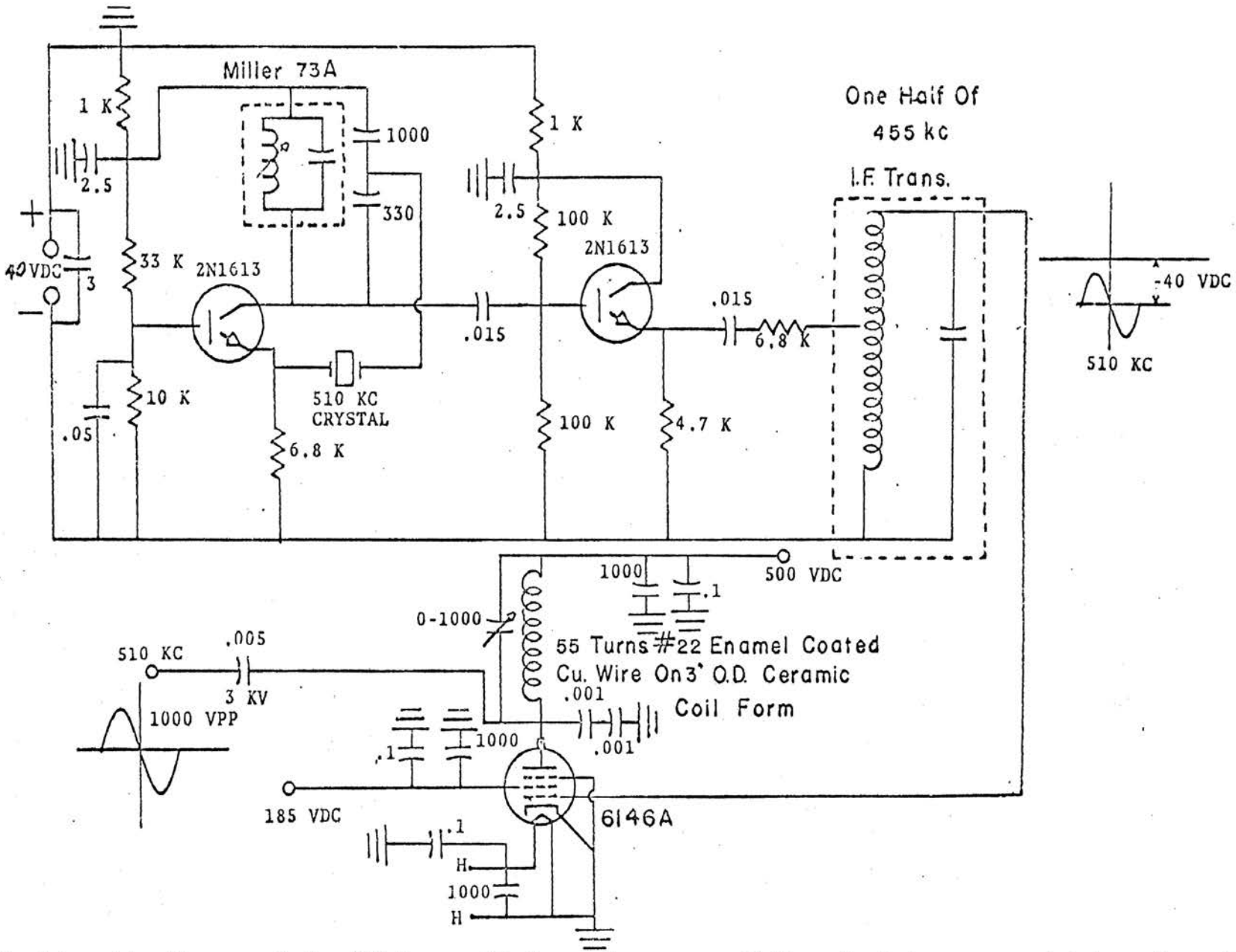


Figure 2. Schematic diagram of the 510 kc oscillator and power amplifier. Resistances are in ohms. Capacitor values greater than 3 are in picofarads. Capacitor values of 3 or less are in microfarads.

A 9558 EMI photomultiplier was used as both a signal detector and as a mixer as shown in Fig. 3. The laser beam (modulated at 510 kc) is detected in the usual manner by the photomultiplier. However, the last dynode of the photomulitplier is modulated by a 502 kc signal from a single side band generator shown in Fig. 4. As far as the electrons are concerned, the last two dynodes and the plate of the photomultiplier constitute a triode. A mixing effect is seen and can be quantitatively analyzed as in any mixing tube[15]. This procedure of employing the photomultipler as a mixer is believed to be unique. It results in a high performance, low noise mixer. The advantage of simplicity is obvious.

The phase multiplier which was built is shown schematically in Fig. 5. It is similar to the one built by Noble and Cook except that it was designed for 8 kc signals rather than 10 kc[16]. The tubes used were 12AU7's. Resistances are in ohms. Capacitor values greater than 1 are in picofarads. Capacitor values less than 1 are in microfarads. The phase multiplier consists of two frequency multipliers; one provides a factor multiplication of 8 and the other a factor of 9. The reference 8 kc signal with phase angle 0° is multiplied in frequency by 8. The information 8 kc signal with phase angle ϕ is multiplied in frequency by 9. These two signals are then

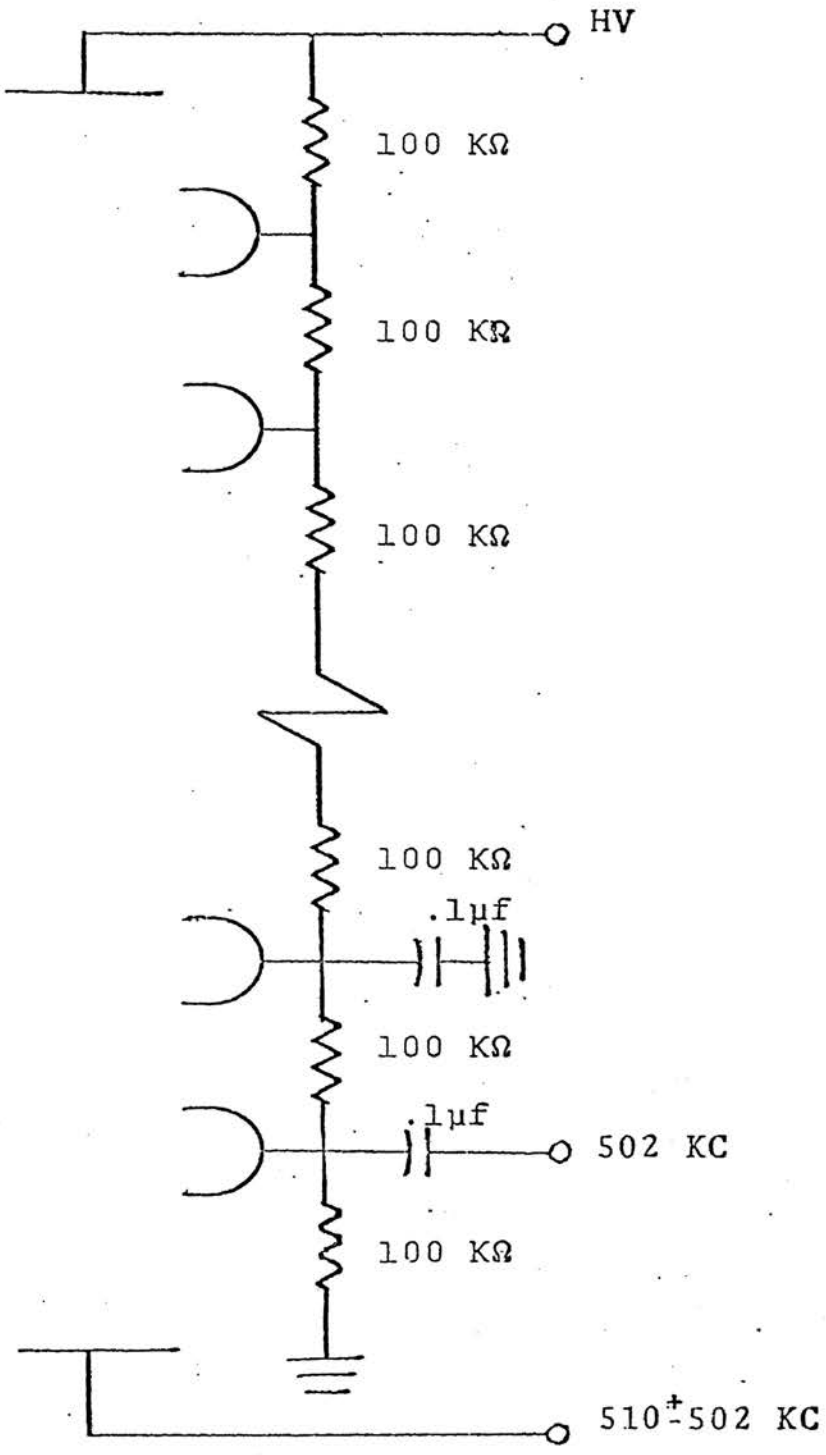
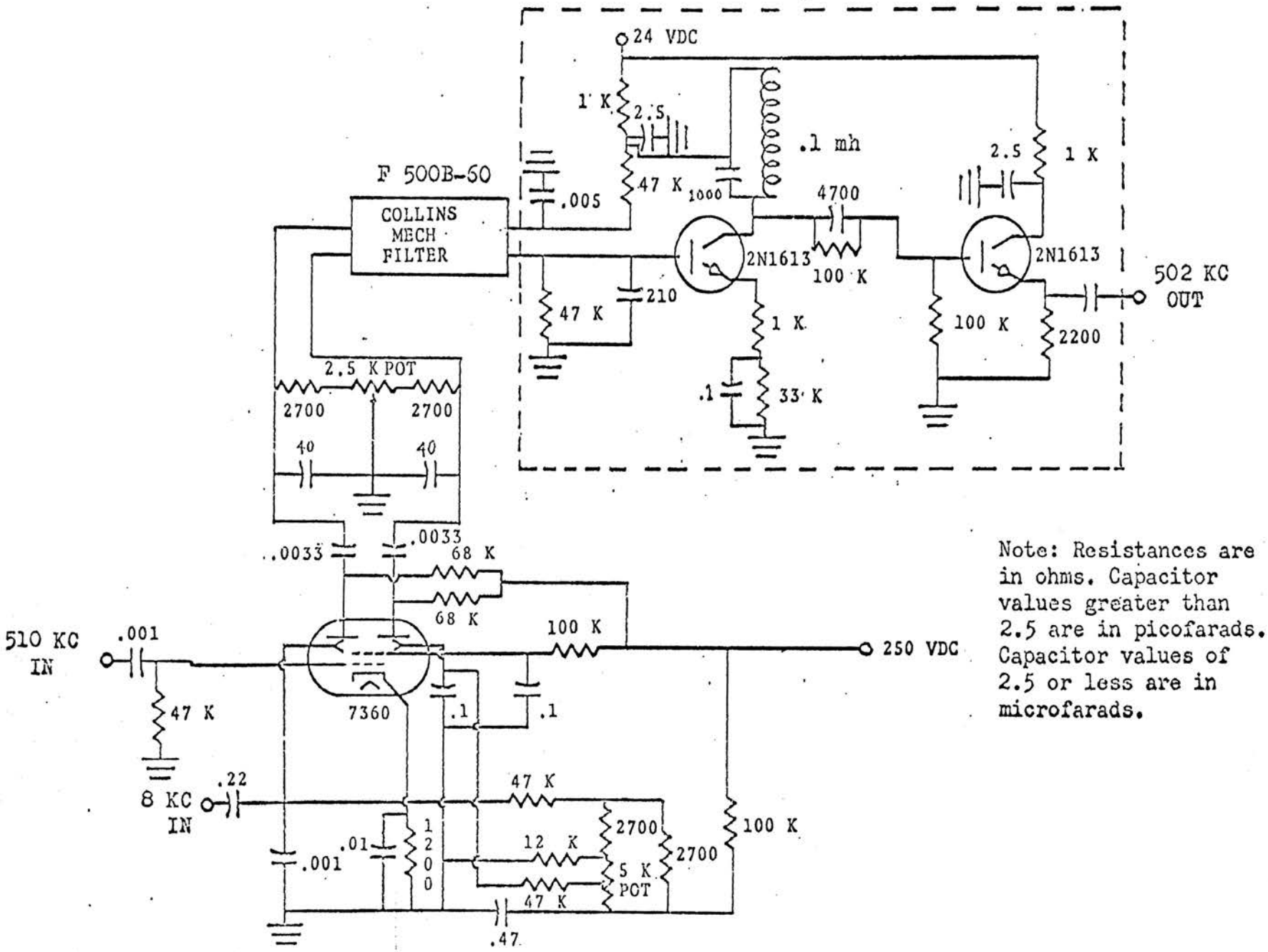


Figure 3. Photomultiplier used as a mixer.



Note: Resistances are in ohms. Capacitor values greater than 2.5 are in picofarads. Capacitor values of 2.5 or less are in microfarads.

Figure 4. Schematic diagram of the single side band generator.

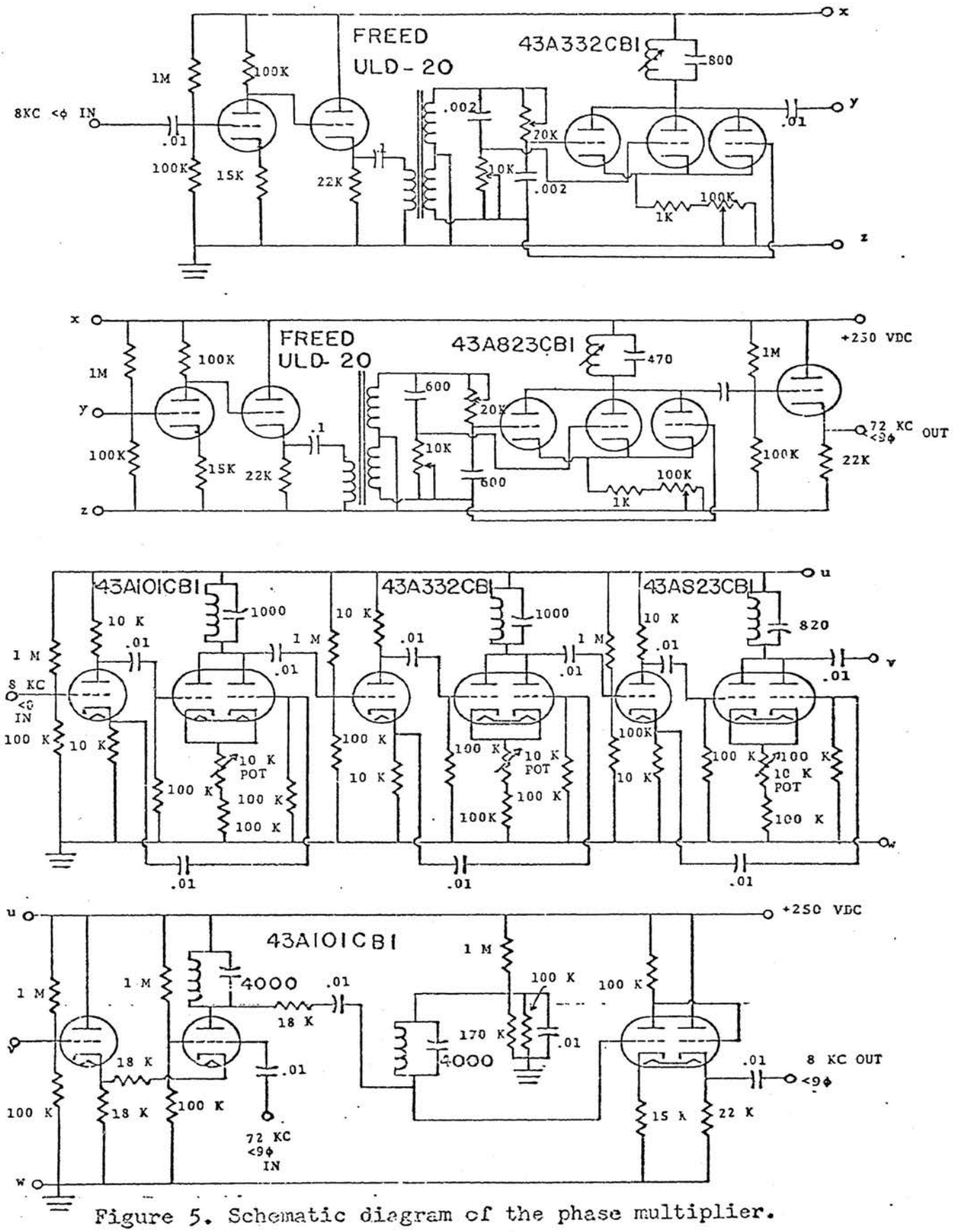


Figure 5. Schematic diagram of the phase multiplier.

mixed and filtered giving a signal of 8 kc and phase angle of 9 ϕ. Now we can measure small phase angle differences associated with low frequency modulation in the phase shift method.

The phase shifter consists of an RC network with a precision varible resistance as shown in Fig. 6. It is similar to those of Rollefson[5] and Ware[10]. The phase angle ϕ corresponding to a given resistance setting of the phase shifter is calculated from $\tan(\phi/2)=2\pi f'RC$. When $f'=8$ kc, this is $\tan(\phi/2) = .10048R$, where R is measured in Kohms.

For making lifetime measurements the experimental arrangement will be basically the same as in Fig. 1. A cell containing atoms of a given element will be substituted for the mirrors. The fluorescent emitted light will be detected at 90° from the pumping light in order to reduce the amount of scattered light.

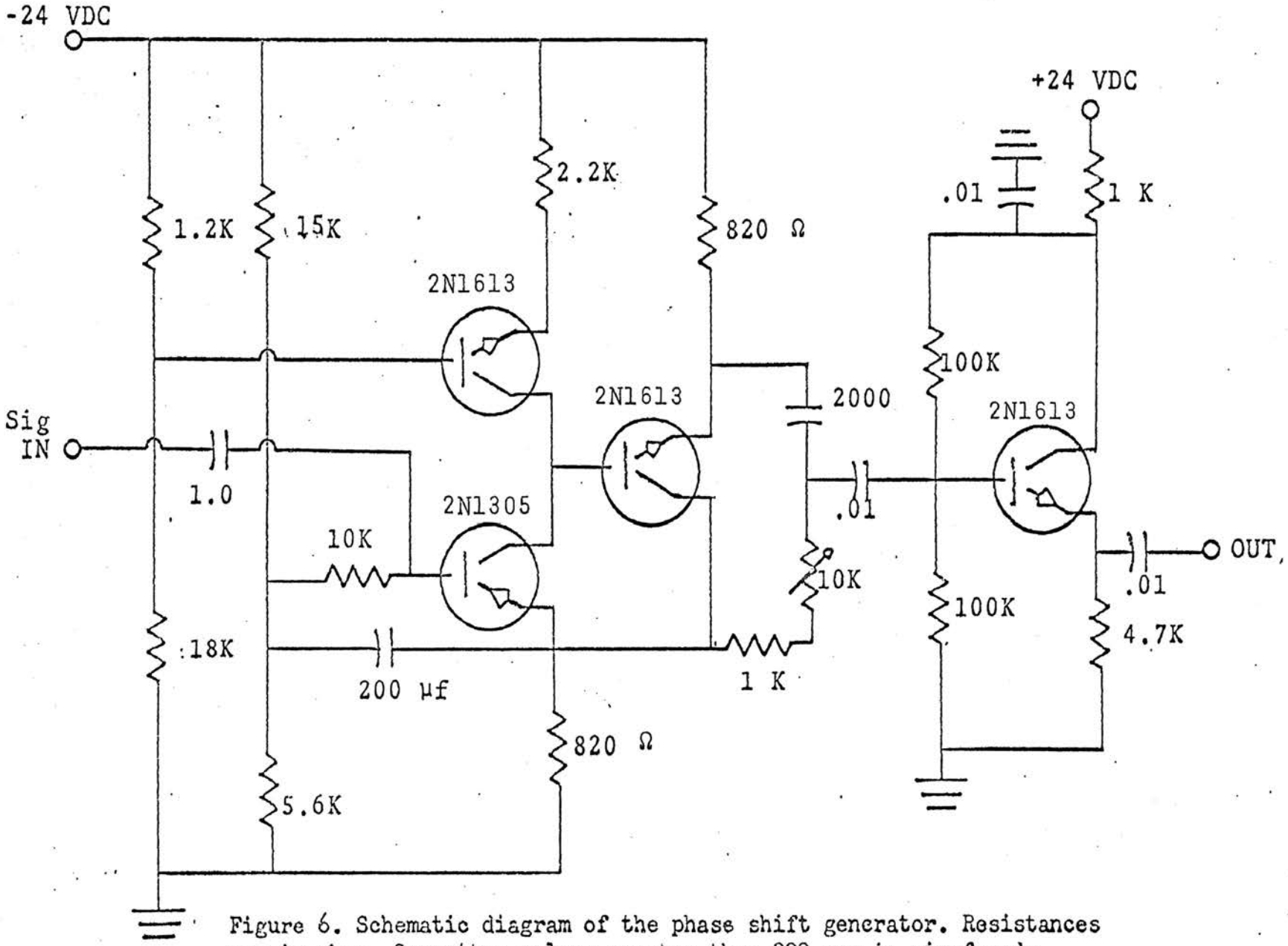


Figure 6. Schematic diagram of the phase shift generator. Resistances are in ohms. Capacitor values greater than 200 are in picofarads. Capacitor values of 200 or less are in microfarads.

RESULTS AND DISCUSSION

The phase shift was measured 48 times over a two week period when the mirror was moved a distance of two meters. The results of these measurements are seen in Table 1. For each value of ϕ in Table 1 a value for the speed of light was calculated from

$$C = \frac{360°fd}{\phi} .$$

These values are shown in Table II. The average value for the speed of light measured was found to be $\bar{c}_{mes} = (2.937 \pm .033) \times 10^8$ m/sec. The value 0.033 is given by $\frac{\sigma}{\sqrt{n}}$, where σ is the standard deviation, and $n = 48$. This represents an error of 1.61% from the accepted value of 2.985×10^8 meters per second for the speed of light in air.

A .95 confidence interval was constructed as follows:

$$\bar{c}_{mes} \pm 2.012 \times .033$$

or

$$\bar{c}_m \pm .066$$

which is

$$2.871 \times 10^8 < \bar{c}_{mes} < 3.003 \times 10^8 .$$

The value of 2.985×10^8 m/sec certainly falls within this interval.

TABLE I

ϕ is the phase angle difference measured with a distance change of 2 meters.

ϕ(degrees)	ϕ(degrees)
1.228	1.349
1.361	1.240
1.313	1.393
1.228	1.330
1.172	1.202
1.396	1.209
1.173	1.244
1.194	1.286
1.150	1.269
1.161	1.228
1.167	1.373
1.492	1.018
1.367	1.486
1.258	1.354
1.193	1.327
1.447	1.141
1.357	1.208
1.217	1.255
1.161	1.258
1.096	1.159
1.183	1.241
1.148	1.227
1.330	1.345
1.312	1.227

ϕ ave, = 1.258 $\pm$ 0.0142

TABLE II

Individual values for the measured speed of light in units of 10^8 meters per second.

2.990	2.722
2.912	2.961
2.797	2.636
2.990	2.761
3.133	3.055
2.636	3.037
3.130	2.952
3.075	2.855
3.193	2.894
3.163	2.990
3.147	2.674
2.461	3.607
2.686	2.471
2.919	2.712
3.078	2.767
2.538	3.218
2.706	3.040
3.017	2.926
3.163	2.919
3.350	3.168
3.104	2.959
3.199	2.933
2.761	2.730
2.799	2.993

Suppose we use the experiment as a test of a hypothesis. The hypothesis is that our equipment is of good enough quality to perform lifetime measurements. Our experimental results yield at .95 confidence interval which contains the accepted value of the speed of light in air. On this basis we can accept the hypothesis.

We feel that the error came mainly from the measurement of the path length. This error may have been as much as six or seven parts in a thousand.

CONCLUSION

Although the individual values were not always good, in the average a very good result was obtained. It is felt that the speed of light was measured to an accuracy and with a confidence commensurate with the best results obtained in the past by others who have used the phase shift method. Therefore, employment of phase multiplier and the use of the photomultiplier as a mixing tube are novel and useful techniques in the phase shift method. From this experiment we can now say that we are ready to make lifetime measurements using the phase shift method with low frequency optical pumping of the excited state and with final mixing in the last dynode state of the photomultiplier.

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VITA

Ronnie Carroll McMillan was born July 7, 1943, in Mena, Arkansas. He received his primary and secondary education in Nashville, Arkansas. He received his college education from Arkansas Polytechnic College, in Russellville, Arkansas, and the University of Missouri - Rolla. He received a Bachelor of Science Degree in physics and a Master of Science Degree in physics from the University of Missouri - Rolla in May 1967 and January 1969 respectively.

He has been enrolled in the Graduate School of the University of Missouri - Rolla since September 1967 and has had a research assistantship for the period from June 1967 until June 1968.