II. MICROWAVE PHYSICS

A. MICROWAVE SPECTROSCOPY

Staff: Professor A. G. Hill R. Hillger J. G. Ingersoll Dr. R. L. Kyhl R. B. Lawrance M. W. P. Strandberg T. Wentink, Jr.

1. Microwave-Frequency Bridge

Staff: M. W. P. Strandberg J. G. Ingersoll

Attempts have been made to detect, with the familiar sweeping technique, those absorption lines of oxygen which occur in the 5-mm region. This has been unsuccessful. Two considerations account for this failure. The bridge has the sensitivity to detect absorptions of the order of $\frac{1}{2}$ db/km when operated as a point-by-point balance system, but when used with a sweeping technique, this sensitivity is decreased because of the rapidity with which information is obtained. If the data are taken slowly enough to use a bandwidth as narrow as that employed in a point-by-point scheme, minute variations in the waveguide impedance over the frequency range swept (~ 5 Mc/sec) set a pseudo-noise level much higher than NkTAv. These facts have been established experimentally and for the present the oxygen work is considered completed.

Work is in progress measuring the higher rotational absorptions in COSe; i.e., J, $5 \rightarrow 6$, $6 \rightarrow 7$, $8 \rightarrow 9$, to yield data on the rotational distortion of the molecule.

2. Sweep Spectrometer

Staff: M. W. P. Strandberg R. E. Hillger T. Wentink, Jr.

Measurements of the spectrum of COSe in the 1-cm region are under way. Lines for all the stable selenium isotopes have been identified together with the much weaker lines due to the naturally occuring C^{13} in the molecule with 0^{16} and $\mathrm{Se}^{78,80}$. Rotational lines perturbed by the symmetric and bonding vibrations of this molecule have also been measured. Stark measurements have been made on the rotational lines in both the ground and excited vibrational states. The electric dipole moment has been observed not to change with the selenium isotope, within present experimental error, but it does decrease in the excited symmetrical vibration state, giving moments of $0.752 \pm .007$ and $0.728 \pm .007$ Debye units in the ground and excited

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states, respectively. From intensity ratios the bonding (v_2 =1) frequency has been determined as 440 cm⁻¹.

An absorption cell which can be varied in temperature from -200° C to + 350°C has been constructed and work continues in an effort to find the rotational lines perturbed by the asymmetrical vibration and to detect the naturally occuring 0^{18} in the 0^{18} C⁴²Se^{78,80} molecule.

The successful conclusion of these measurements should allow a fairly accurate determination of the potential constants of this molecule up to third-order terms. This information will allow a reasonably accurate determination of the interatomic distances, and, consequently, the selenium mass values.

3. Miscellaneous Problems

Dr. R. L. Kyhl

(1) The d-c electric field has been calculated for a waveguide containing a Stark septum. Assuming a TE_{Ol} -mode pattern for the microwave field the shape of a Stark component of an absorption line has been calculated. The position of the line center is exactly the same as for a uniform d-c field. The line broadening due to field inhomogeneity is about $l\frac{1}{2}$ per cent of the Stark splitting.

(2) Some estimates have been computed of the accuracy required to obtain selenium mass values from the COSe spectrum (in conjuction with M. Strandberg). Molecules containing both carbon and oxygen isotopes must be measured. To obtain packing fractions to 10 per cent, it appears that over-all measurement accuracy of \pm 10 kc is necessary and the rotation constants must be known to 1 in 1000 (or perhaps only the variation of these constants to 1 per cent).

(3) An automatic frequency control circuit has been built for a new K/2 search system. It uses a 15-Mc channel to go with the 30-Mc channel for the doubled signal. A doubler-mixer has been built which it is hoped will be more rugged mechanically and suitable over a broader band than the present types. Tests are being made on these components.

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II. B. MOLECULAR BEAM RESEARCH

- Staff: Professor J. R. Zacharias Professor B. T. Feld L. Davis, Jr. Dr. R. S. Julian Dr. D. E. Nagle C. W. Zabel
- 1. Hyperfine Structure of K⁴⁰

Staff: Professor J. R. Zacharias L. Davis, Jr.

<u>Summary</u>. The atomic beam magnetic resonance method has been used to determine the hyperfine structure Δv of K^{40} and to make a positive assignment of the results to the nucleus of mass 40. The apparatus used in these measurements was similar to that described by Zacharias¹, but with a mass spectrometer in place of the usual surface ionization detector, which in this case serves as the source of ions for the mass spectrometer. The mass spectrometer reduces the general background due to other alkali atoms in the apparatus and other isotopes of potassium in the beam. The entire apparatus has been designed for measurements with artificially radioactive nuclei, such as Na²², and a special oven with a long exit canal was used to conserve material.

At mass number 40 transitions were observed at weak field and low frequencies (2 to 8 Mc) which correspond to an alkali atom with a nuclear spin of 4. A resonance at 1285.745 Mc in a field of 2.58 gauss and another at 1285.774 Mc in a field of 3.37 gauss when weighted yield a value of the Δv for K⁴⁰ of 1285.73 \pm .05 Mc. This confirms previous results.

<u>Theory</u>. The theory of this experiment has been thoroughly described in a previous paper by Zacharias². The frequency of the $F = i + \frac{1}{2}$, $m_f = -(i+\frac{1}{2}) \leftrightarrow m_f = -(i-\frac{1}{2})$ transition can be shown to be approximately $v_{40} = 2v_{39}/(i+\frac{1}{2})$ for very weak fields. By observation of this line, and that of K^{39} the nuclear angular momentum i may be directly determined.

At weak magnetic fields the hyperfine structure Δv splits into 32 lines (for i=4), 31 of which are observable in this apparatus; and for very weak fields 16 lines are seen, 15 of which are degenerate. The frequencies of the $F = i+\frac{1}{2}$, $m_f = \pm \frac{1}{2} \leftrightarrow F = i-\frac{1}{2}$, $m_f = \pm \frac{1}{2}$ transitions are degenerate and almost independent of magnetic field at very weak fields. These lines may hence be used for a very accurate determination of the hyperfine structure Δv .

<u>Apparatus</u>. A schematic diagram of the K⁴⁰ apparatus is shown in Fig. 1. Atoms from the source pass through the apparatus as shown, around the

^{1.} J. R. Zacharias, Phys. Rev., <u>61</u>, 270 (1942).

^{2.} Ibid.



Figure 1. Schematic diagram of K⁴⁰ apparatus.

obstacle wire, and if their space quantization has made the necessary change in the r-f field, fall on the detector. This apparatus is in principle the same as that used and described by Zacharias¹ except for the source and detector.

The oven source designed primarily for Na²² has been constructed to conserve as much of the material under observation as possible. A canal .010 in. x .010 in. in cross section and 1 in. long has been cut in the face of a stainless steel block which, when covered by another block of metal, forms the exit slit. This oven has been found to have a beam intensity which is down to 40 per cent of maximum at both ends of the detector filament, and a width at half intensity of l_2^1 degrees. As the intensity in the forward direction is independent of the length of canal for lengths less than a mean free path, a measure of the amount of material saved is in the ratio of the total effusion rates for an extremely short canal and for one of length 1. The ratio, g, of gas flow through such a thin-walled hole to that through the long canal as given by Smoluchowski² can be shown to be g = 3l/4d where d is the side of the square cross section of the canal. By this means $l\frac{1}{2}$ per cent as much material is needed for such a canal as for a hole in a thin wall. A cold trap with a small hole for the beam is placed directly in front of the exit canal to recover most of the material from the oven.

The detector, as shown in Fig. 1, utilizes a hot wire surface ionization detector as the ion source for the mass spectrometer. The detector used here is unusually large for such an apparatus, being

^{1.} J. R. Zacharias, loc. cit.

^{2.} M. V. Smoluchowski, Annaler der Physik, 33, 1559 (1910).

0.010 in. x 1.20 in. and requiring a hot wire 0.015 in. wide. The ion accelerating plate is cylindrical in order to focus the ions in a vertical direction so as to pass through the 1-cm gap of the magnet. An 8-cm radius, 60-degree wedge magnet is used, with a Faraday cage as a collector. An FP-54 electrometer tube circuit measures collector current with a sensitivity of 0.6 x 10^{-16} amps/mm having about 1 x 10^{-16} amps as the minimum detectable current in the noise background. The calculated aberrations of the mass spectrometer³ indicate that good resolution may be achieved at mass number 22, but that resolution is decreasing rapidly about mass number 40. This can be shown to be mostly due to the detector width and height so that for higher mass numbers a smaller detector area is necessary. The use of the mass spectrometer decreases the general background from alkalies in the apparatus, that from other isotopes in the beam, and determines the mass number under observation.

<u>Method</u>. The low-frequency lines of K_{40} and K_{39} were first observed to determine the nuclear angular momentum i of K_{40} which was found to be 4. At very weak fields and at high frequency the 16 lines in the neighborhood of Δv were observed unresolved, and yielded a crude determination of $\Delta v = 1285.76 \pm 0.3$ Mc/sec. Proceeding to a field of 2.58 gauss sufficient to separate the lines by about 0.7 Mc/sec, a line was observed at 1285.745 Mc/sec as shown in Fig. 2. At 3.37 gauss the line was observed



at 1285.774 Mc/sec. These observations show the line to be field-insensitive and yield a value of $\Delta v = 1285.73 \pm .05$ Mc/sec.

2. Hyperfine Structure of Chlorine

Staff: Professor J. R. Zacharias Professor B. T. Feld L. Davis, Jr. C. W. Zabel

A successful attempt to detect a beam of chlorine atoms by electron attachment has resulted in a preliminary investigation of the hyperfine structure of Cl^{35} and Cl^{37} . Several transitions have been found, all of which are consistent with a nuclear spin of 3/2 for both isotopes.

The ground states of chlorine, J = 3/2, is split into 2I+1 levels, F = 0, 1, 2, 3 by the interaction of the nucleus with the electrons. Each of these levels breaks up into 2F+1 levels with the application of a magnetic field. Figure 3 shows, in a rough way, the energy of each state as a



PARAMETER PROPORTIONAL TO MAGNETIC FIELD

Figure 3. Sketch showing the dependence of the energy levels of chlorine on the magnetic field.

function of the magnetic field. The exact solutions of the secular equations which give the energy levels are being made. The transitions indicated in Fig. 3 should be observable in the experiment preformed. The two low-frequency transitions and the several lines between F=2 and F=1 were observed. The transition between F=3 and F=2 has not yet been found. From the exact solution of the secular equations and the frequencies of the transitions observed, an approximate value of the $(3,-1) \leftrightarrow (2,-1)$ frequency may be calculated. It is hoped the line will then be found. With this added data the dipole and quadrupole interaction constants are easily obtained. The frequency of the transition from F=2 to F=1 at zero applied field is 355.23 Mc/sec for $C1^{35}$ and 298.12 Mc/sec for $C1^{37}$.

The apparatus used is the same as described in the K^{40} experiment. In the case of chlorine the surface ionization tungsten ribbon forms negative ions since the halogens have a very large electron affinity. The source of chlorine atoms is the same source as was used in the hydrogen experiments described in the July 15, 1947 Progress Report.

3. Hyperfine Structure of Na²²

Staff: L. Davis, Jr.

<u>Apparatus</u>. The apparatus as shown in Fig. 4 is essentially the same as that described for the K^{40} experiment. The oven is the same, but instead of distilling Na²² from the metal, calcium chips are added to refine the Na from NaCl. For very small quantities of Na²² (about 10^{-9} moles) it is found that this process proceeds rather slowly and that the detector sensitivity must be increased. The electron-multiplier described next in this report is being tried as a collector for the mass spectrometer, but corona back-ground has so far prevented its use.

4. Electron Multiplier

Staff: H. Lew

<u>Electron Multiplier for Ion Detection</u>. The most satisfactory way of detecting a beam of atoms or molecules after it has passed through a magnetic resonance apparatus is to ionize it and measure the ion current. The most sensitive current measuring device that has been used in molecular beam work up to now is the electrometer tube (FP-54). It has been considered desirable, however, expecially in the study of radioactive isotopes which often come in very minute quantities, to find an even more sensitive way of measuring ion currents. To this end, a secondary emission multiplier with a very low background or dark current and sensitive to ions has been constructed. Rough tests have indicated that its efficiency in the detection of potassium ions of 3600-volt energy is very high; i.e., between



Figure 4.

50 and 100 per cent. The background is about one count a second. Thus an incident ion current of two or three ions a second should be readily detectable. This corresponds to a current of 10^{-19} amperes. The multiplier geometry is exactly that of the RCA 931-A but the electrodes are made of beryllium copper. The treatment of the electrodes consists in cleaning them with emery paper, dilute hydrochloric acid, and acetone before assembly and then, after assembly, firing in an atmosphere of hydrogen at 600° C for 15 minutes.

For use with the multiplier, amplifiers and scalers have been constructed.

Extension of Surface Ionization Method. The applicability of the molecular beam technique is greatly limited by the difficulty of ionizing atoms or molecules with any efficiency. So far, the surface ionization method is the only one that has been used to any extent. It has been applied successfully only to the alkalies and to indium and gallium. Work is now progressing to see whether the method could be applied to other atoms. It is hoped that, even if the efficiency of conversion of atoms to ions is very low in these cases, the sensitivity of the secondary emission multiplier mentioned above may make the method practicable.

5. Hyperfine Structure of Hydrogen

Staff: Dr. K. S. Julian Dr. D. E. Nagle, Jr.

<u>Apparatus</u>. A better determination of the Δv of hydrogen and deuterium is planned requiring considerably greater resolution. To this end the new apparatus will be used in order that a longer homogeneous field and greater separation between magnets may be used. New magnets have been designed and are in construction which will give a much longer and more homogeneous field than that used previously.

6. Third Molecular Beam Apparatus

Staff: H. Lew

Work has begun on a third molecular beam apparatus. The vacuumtight can for this will be of cast brass, differing from the second apparatus only in detail. It is proposed to study molecules with this third apparatus.

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II. B. 7. Nuclear Quadrupole Effects in the Molecular Beam Spectra

Staff: Professor B. T. Feld M. J. Klein

An analysis of the theory of nuclear quadrupole effects in the molecular beam spectra of diatomic molecules has been carried out. A theory of these effects has been given by Feld and Lamb¹. This theory treats the rotational motion of the molecule classically by assuming that the ratio of the quantum numbers M_J/J is a continuous variable z whose range is -1 to + 1. The predictions of this theory have been compared with the results of exact calculations (to second order in perturbation theory) for particular cases of finite J values. Approximations valid to order 1/J and $1/J^2$ have been developed to supplement the theory of Feld and Lamb for the two situations of experimental interest (strong and vanishing magnetic fields). Calculations of line shapes which will show the effects of the various approximations are in progress.

^{1.} Phys. Rev. <u>67</u>, 16 (1945).

II. C. LOW-PRESSURE GAS DISCHARGES

Staff: Professor S. C. Brown Professor W. P. Allis M. A. Biondi E. Everhart M. A. Herlin Donald E. Kerr

<u>Transient Discharge Characteristics</u>. The failure of measured values of the ambipolar diffusion coefficient to agree with theoretically predicted behavior has led to an investigation of the diffusion problem at transition densities. It is found that for a given container and gas within the container, there is a minimum density of positive ions and electrons which must be present in the gas in order that the conditions for ambipolar diffusion be met all the way to the walls of the container. The conditions for ambipolar diffusion are that the positive ion density be nearly equal to the electron density (within 10 per cent for example) and that the vector drift velocities of the electrons and ions be nearly the same.

A simple calculation shows that the field which must be set up by the small charge density difference at each point to maintain ambipolar diffusion at that point is

$$\mathbf{E} = -\frac{\mathbf{D}_{-}}{\boldsymbol{\mu}_{-}} \frac{\boldsymbol{\nabla}\mathbf{n}}{\mathbf{n}}$$

where D_ and $\mu_$ are the electron diffusion coefficient and mobility coefficient respectively, and n refers to either positive ion or electron density.

For parallel plate geometry the space charge necessary to set up this field is

$$(n_{+} - n_{-}) = \frac{2}{3} \left(\frac{\pi}{a!}\right)^{2} \frac{\epsilon_{0}}{e} V_{av} \csc^{2}\left(\frac{\pi z}{a!}\right)$$

where a' is the plate separation except for a negligible correction factor due to the fact that the density does not go exactly to zero at the walls; V_{av} refers to the average electron energy in electron volts; and ϵ_{o} is the permittivity of a vacuum.

If we require that for ambipolar diffusion the difference between electron density and ion density be one tenth or less of the total electron or ion density, then the average density which must be present between parallel plates separated 2.5 cm is

$$\bar{n} = (6.55 \times 10^{16})p$$

where \bar{n} is in electrons per cm³ and p is the pressure in mm Hg.

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Since average concentrations of electrons in our experiments rarely exceed 10^{11} electrons/cm³ we do not have ambipolar diffusion all the way to the walls. Calculations show that in the 1 - 10 mm pressure range the ambipolar region merges into a positive ion sheath near the walls. The sheath is of the order of 0.01 cm thick so that for all practical purposes the experimental conditions are identical with ideal ambipolar diffusion conditions.

<u>Microwave Radioactivity Counter</u>. The purpose of this investigation is to survey the applicability of microwave gas discharges to detecting ionizing radiation. Considerable progress has been made in evaluating the factors involved. In these microwave gas discharges, breakdown occurs when the rate of production of electrons under the action of the electric field exceeds the rate of loss due to diffusion, attachment, etc. After breakdown occurs, the number of ion pairs per unit volume is so large that the electrons diffuse ambipolarly. When the discharge is turned off, the space is cleared of electrons with a speed corresponding to positive ion diffusion because of the ambipolar phenomena.

If an uhf discharge in a resonant cavity is used as a discharge counter, it has the following general characteristics. Because of the fact that the breakdown does not depend on secondary effects, the speed of saturation of the discharge is very rapid. Measurements show that the pulse from the discharge rises from 0.05 volts to 5 volts in 6 x 10^{-9} sec. The size of the pulse being very large, it has to be attenuated greatly to prevent burning out of the crystal rectifier. The clean-up time for the electrons after the discharge is turned off, since it is controlled by the positive ion space charge, may be a few hundredths of a second. This extremely long resolving time dictated by the ambipolar diffusion of the electrons seems to preclude the possibility of using the pure high-frequency discharge in a useful detector for ionizing radiation.

Cavity resonators may be designed so that a d-c field is applied in addition to the impressed uhf field. The effect of a d-c field is to speed up the removal of electrons from the ionizing region. This will affect the a-c breakdown power, since breakdown depends upon the balance of production and loss of electrons. The d-c field will speed up the loss of electrons and the a-c power for breakdown will increase as the d-c field is increased. This is true, however, only up to a certain point, for as the d-c field is increased it reaches a value sufficiently high for this d-c field to produce multiplication, and when this starts to occur the necessary a-c power is lowered. If a discharge in an a-c -- d-c cavity is turned off, by removing the a-c field, the speed of removal of the electrons is very high in comparison with the pure a-c case, since the mobility of the electrons is so much greater than their diffusion. To increase the

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speed of removal of electrons, it is advantageous, as in the case of the Geiger-Müller counter, to produce the discharge in a cylindrical geometry where the electrons are formed close to the anode and may be swept out by the high field of this region where the distance of travel of the electron is small.

To gain the advantage of short resolving time, increasing the steady field is important, but this increase in steady field increases the necessary uhf field for breakdown. As the high-frequency power is increased, the total number of ion pairs formed in the discharge increases, and this will tend to increase the resolving time. Although increasing the d-c field results in a net decrease in resolving time, the resulting increase in power into the cavity to produce breakdown provides a limit to the minimum resolving time because of the size of the produced discharge. Under optimum conditions, with a steady d-c potential of 500 volts applied, the minimum resolving time observed has been 30 microseconds.

If the discharge in the cavity is initiated as a pure uhf discharge, the number of ion pairs produced will be a minimum. If a d-c pulse is applied to the discharge as soon as possible after breakdown, this can act as a clearing field without affecting the number of ion pairs created, providing it is below the value for production of cumulative ionization. Under these conditions the minimum resolving time observed was that controlled by the time constant of the magnetron power supply, and was measured as 5 µsec.

In coaxial symmetry, the high field occurs in the vicinity of the center electrode. Since it is desirable to have the ionizing field concentrated close to the center electrode for rapid electron collection, a fine wire gives the shortest resolving times. If the only field in the cavity is oscillating at ultra-high frequencies, only those primary ionizing particles which happen to pass through the high field region near the center wire will cause breakdown. This greatly diminishes the sensitivity of the cavity as a gamma-ray counter. If a steady field exists between the electrodes, and the axial wire is at a positive potential, electrons formed anywhere within the resonant cavity will be pulled into the high field region and cause breakdown. Thus with the conditions for shortest resolving time the sensitivity to gamma rays is low. A compromise between resolving time and sensitivity can be achieved by superimposing on the a-c field, a steady d-c potential of a few hundred volts for increasing sensitivity, and a pulsed d-c voltage of several hundred applied after breakdown, for sweeping out electrons formed in the discharge.

<u>Microwave Breakdown Measurements</u>. Microwave breakdown experiments have reached the final stage where reproducibility of measurement technique and gas fillings is being checked. Careful calibrations and general conditioning of the measuring equipment has resulted in accuracies to a few per cent in voltage measurements. The chief difficulty has been experienced in obtaining reproducible gas fillings.

One source of variation has been in the outgassing of the glass tubes containing the electrodeless discharges. The heat produced during the discharge together with the ionic bombardment of the walls has been found to release enormous amounts of gas in spite of the amount of bakeout possible with pyrex glass. The only means thus far of eliminating this outgassing has been to use quartz bottles which have been baked at about 600° C, at which pyrex would soften. Tubes with metal walls have given very little trouble even without bakeout, because the walls are sufficiently massive and of high enough thermal conductivity to prevent heating.

Two breakdown curves (Fig. 1) are given in which the electric field for breakdown is plotted against the pressure for mercury-contaminated tank



Figure 1. Breakdown curves for mercury-contaminated tank helium.

helium. One curve is for a cylindrical quartz bottle. The other is for a thin TM_{OlO} -mode cavity whose thickness was small compared to the diameter of the near-uniform field region. It thus gives results which are substantially the same as infinite parallel plates. The walls of the latter are metal, so that the difference between conducting and insulating walls could be determined. Since the size of the quartz bottle is larger, the diffusion losses of electrons in the discharge are smaller and the breakdown field is therefore lower. The difference is most pronounced at low pressure.

As stated in earlier reports, the breakdown measurements have been used to yield ionization rates as a function of the field strength, frequency, and pressure. This idea will be given in some detail here. The basic equation is balancing the production of electrons due to ionization by collision of electrons with neutral gas atoms against loss of electrons by diffusion. The effects of positive ions and space charge are negligible, since the breakdown field is the field required to maintain the steady state in the limit of zero current density. A field strength in excess of this value in the slightest amount will thus produce breakdown, inasmuch as the field for maintaining a discharge is always lower than the breakdown field, because of space charge effects at high current densities. The breakdown condition therefore is obtained from the equation,

$$\nabla \cdot \Gamma = v_{i}n ,$$

where Γ is the electron current density, n the electron concentration, and v_i the ionization rate in electrons per electron per second. There are nv_i electrons produced per unit volume per second, and this production must just balance the loss per unit volume due to the current flow, which is the divergence of the current density. Since the electric field is oscillating at a high frequency there is no field-induced current of sufficient magnitude to consider in comparison with the diffusion current,

$$\vec{\Gamma} = - D \nabla n,$$

where D is the electron diffusion constant. The parameters v_i and D are both functions of the electric field strength, frequency, and pressure. Substituting this current into the continuity equation, we obtain

$$\nabla^2 n + \frac{v_1}{D} n = 0.$$

The boundary condition on n may be shown to be that the extrapolated electron density vanish a distance of the order of a mean free path beyond the boundaries of the discharge bottle. If the pressure is high enough, it is sufficiently accurate to say that n is zero at the boundary. The application of this boundary condition yields characteristic values for v_1/D , the lowest of which is the only one corresponding to the physically realizable condition that n remain positive over the entire tube. For parallel plates n varies sinusoidally, and

$$\frac{v_1}{D} = (\frac{\pi}{L})^2$$

where L is the plate separation distance. In a cylindrical tube,

$$\frac{v_1}{D} = \left(\frac{\pi}{L}\right)^2 + \left(\frac{2.405}{R}\right)^2$$

where L is the length and R the radius of the cylinder. Thus the ionization rate required to produce breakdown is determined entirely from the geometry of the tube, and the breakdown field which is measured is the field required at that pressure and frequency to give the prescribed ionization rate.

From kinetic theory which cannot be given here it may be shown that,

$$\frac{v_1}{Dp^2} = f(\frac{E}{p}, E\lambda)$$

where p is the pressure, E the electric field strength, and λ the wavelength of excitation energy. The function should reduce to the dependence on E/p alone when the pressure is high enough to give many collisions of electrons with gas atoms per r-f cycle, while the dependence on E/p drops out and only E λ remains in the low-pressure case where there are many cycles per collision. In the high-pressure (or low-frequency) case, multiplying by p^2/E^2 gives,

$$\frac{v_1}{DE^2} = \phi(E/p)$$

which forms the basis for the curves given, (Fig. 2). These curves are computed from the above breakdown curves determined experimentally. The departure of the curve for the quartz tube at the upper end is due to entrance into the low-pressure region where there are no longer many collisions per cycle. The thin metal wall cavity stays in the "d-c" or high-pressure, low-frequency range to higher values of E/p. The agreement at low E/p is not complete, but close enough to substantiate to some degree the contention that diffusion is the principal electron removal process. The lack of agreement is thought to be due to the different amounts of impurity at different pressures; for example, mercury remains constant at its room temperature vapor pressure while the helium pressure is varied. Further experiments will be performed to check this possibility.



Figure 2. Ionization curves based on the breakdown curves of Fig. 1.

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II. D. LOW-TEMPERATURE RESEARCH

1. <u>Helium Liquefiers</u>

Staff: R. P. Cavileer

During this quarterly period 22 runs have been made with the two Collins Helium Liquefiers. The liquefaction rate has been very erratic. This has been traced to the fact that the helium used was of varying purity so that the clean-up used could not at all times be sufficient. Helium has now become available which is charcoal-cleaned, is oil-free, and has a minimum purity of 99.8 per cent.

2. Superconductivity at Microwave Frequencies

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Staff: Professor J. C. Slater
E. Maxwell
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The investigation of superconducting tin cavities, described in the last progress report, is being carried forward. Measurements have been made on a number of c.p. tin cavities and one Hilger tin cavity from room temperature down to 2.3° K. These data exhibit a fair amount of self consistency. A large discrepancy between a-c and d-c conductivity in the normal state is in evidence as was the case with lead. A detailed report presenting numerical data, should be available before the next progress report.

3. Resistivity of Normal Conductors

a. Experimental

Staff: Professor J. C. Slater W. B. Nowak

The divergence between a-c and d-c resistivities of several metals at 24,000 Mc is being investigated from 300° K to 2° K. Measurement of a cavity Q can be related to the real part of the surface impedance of the walls. A squat cylindrical cavity operating in a TM₀₅₀-mode has been constructed with part of a flat face removable. The break occurs at a current node. The proper mode has been established from a mode chart, and the proper coupling hole is being experimentally found to provide a reasonable VSWR on resonance over the temperature range.

To facilitate measurement of the cavity Q, a mechanical FM modulator has been constructed and tested on the 2K33 klystron. The klystron tuning arm is pushed back and forth by being mounted on a crankshaft. The crankshaft is rotated by an 1800-rpm motor that also turns a sweep generator. The klystron has been modulated, at constant power output, over a band of 225 Mc at 23,200 Mc at a rate of 30 cps. It is still operating satisfactorily after 150 hours.

b. Theoretical

Staff: R. J. Harrison

<u>Mean Free Path Effects in R-F Conductivity</u>. J. B. Garrison¹ at this Laboratory, H. London,² and A. B. Pippard³ each found that the r-f conductivities of metals at very low temperatures, in the non-superconducting state, appeared to be much smaller than the corresponding d-c conductivities.

	dc	ac	$\delta = \sqrt{2/\mu \omega \sigma_{ac}}$	n _a ^{2/3} ł	
	ohm ⁻¹ cm ⁻¹	ohm ⁻¹ cm ⁻¹	cm	cm	
Sn	15x10 ⁷	2.1x10 ⁻⁷	0.3x10 ⁻⁴	17.4x10 ⁻⁴	1200 Mc/sec Pippard, London
Hg	2.1	0.92	0.5	2.26	
Pb	8.4	0.31	0.18	10.5	2400 Mc/sec Garrison

Some of the results are tabulated above. It was suggested by London that the discrepancy between σ_{ac} and σ_{dc} might be explained by the fact that the mean free path of the electrons is considerably larger than the penetration depth. (Compare δ , the apparent skin depth, with the mean free path l_{p} taking n_{a} , the number of free electrons per atom, to be the order of unity).

Since measurements of the d-c conductivity of alkali metal films of thickness small compared to the mean free path⁴ gave results which were in good agreement with a theory based on the simple Drude picture of conductivity, it seemed reasonable to try an extension of this theory to the r-f case.

In the Drude picture, under the influence of a constant electric field E, the conduction electrons acquire the drift velocity $(eE/m)\tau$, where τ is the average time between collisions. This is in addition to their much larger, but randomly directed, thermal velocities. Using Fermi statistics for the electrons in the metal we can say that the effect of the constant field is to shift the origin of the sphere representing the electron distribution in velocity space by an amount equal to $(eE/m)\tau$. With N electrons in this sphere, the current density becomes $(Ne^2T/m)E$. For a given temperature, τ will be a characteristic property of the bulk metal.

4. Proc. Camb. Phil. Soc., <u>34</u>, 100 (1938).

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^{1.} Ph.D. Thesis, M.I.T. 1947.

^{2.} Proc. Roy. Soc. <u>176</u>, 522 (1940).

^{3.} Nature <u>158</u>234, (1946)

For thin films, or at radio frequency, an electron may not be able to acquire the drift velocity $(eE/m)^{\tau}$ during the time τ , either because it might lose momentum in collisions with the surfaces, or because it may have been moving for most of the time τ in a region where the field was much weaker than E. Thus, we can examine qualitatively the situation at radio frequency for the case where the free path is large compared to the skin depth. Consider a small element of volume within the skin. An electron which is coming up through it, from a region very much below the surface, will probably have been moving for a time τ since its last collision, but will only have been in a region of appreciable field for a small fraction of that time, and so have acquired a drift velocity of only a small fraction of (eE/m) \mathcal{T} . An electron coming from the opposite direction, will have been travelling only a relatively short time since its collision with the surface, and thus could not have acquired much velocity since then. If it has undergone specular reflection at the surface, the additional drift velocity which the electron would retain would, by the previous argument, also be small.

By an extension of this qualitative picture, one can show roughly what the angular distribution of drift velocities would be for various distances from the surface of the metal and how the conductivity and field vary with depth. Thus, the field falls off rapidly within the first skin layer (defined by the standard formula, with $\sigma_{\rm ac}$ instead of $\sigma_{\rm dc}$), but thereafter (for $l >> \delta$) it falls off more slowly, the rate at large depths being determined solely by the mean free path. One can also use this picture to judge the validity of various approximations which one makes to derive quantitative results.

On the assumption that the field inside the metal varied as $e^{j\omega t-\gamma z}$, where γ is independent of z (this assumption is not too bad within the skin depth where most of the loss takes place), a formula for σ_{ac}/σ_{dc} was derived which, for the case where $\ell >> \delta$, becomes

 $\frac{\sigma_{\rm ac}}{\sigma_{\rm dc}} \approx \frac{3}{4} (1 + r) \frac{\delta}{l} \log \frac{l}{\delta},$

where r is the coefficient of reflection for electrons at the surface of the metal, and σ_{ac} is an average value weighted to favor the skin depth region. The right-hand side of the equation involves σ_{ac} through δ , and σ_{dc} and n_a through ℓ . Thus there are essentially two arbitrary parameters, r and n_a . Garrison's experimental points for lead could be fitted to this equation within a wide temperature range with a single value of n_a , (approximately .2 per atom, depending on what value of r is chosen). From 4° to 15° K, the value of r does not have to be varied. Above this temperature, the experimental results can be fitted to the sumption that r

decreases with temperature. At still higher temperatures (above about 50°) mean free path effects are negligible, and deviations from theoretical conductivities have to be ascribed to other causes, such as surface strain or roughness. These experimental results indicate the essential correctness of the basic assumptions of the theory, but not necessarily of the quantitative results because of the lack of an independent check on the values of n_a and r.

E. H. Sondheimer and G. E. H. Reuter, of Trinity College, Cambridge, have worked out a theory (private communication to Professor Slater) which gives a much better quantitative treatment, inasmuch as they do not make any ad hoc assumption as to the variation of field within the metal, but work out a self-consistent solution, subject to the requirements of Maxwell's equations, and the Boltzmann equation. They express their results in terms of the surface impedance, which is the quantity experimentally determined.

The assumption is still made that one may speak of a relaxation time and mean free path at low temperatures. If this assumption is not justified, it may not affect the results very much, but this point needs to be clarified.

We should also like to modify Sondheimer and Reuter's treatment to include the case where there are superconducting electrons in addition to the "normal" electrons, in order to interpret the shape of the transition curves.

4. Adiabatic Demagnetization

Staff: M. C. Desirant W. J. Horvath

Apparatus is under construction for the attainment of very low temperatures by adiabatic demagnetization of paramagnetic salts. This involves the construction of a helium transfer tube to remove the liquid from the inside of the Collins liquefier and the handling of the liquid in special Dewar flasks.

An important technique in this work is the preparation of resistance thermometers with a high temperature coefficient of resistance in the liquid helium range. Phosphor-bronze wires with a trace of lead have this property and several castings of such alloys have been made. Thus far none of the wires tested have adequate temperature coefficients for thermometry. In addition to phospor bronze, wires of thoriated tungsten and stainless steel with columbium have been tested in the hope that they would exhibit similar properties, but negative results were obtained.

Further plans involve studies of the properties of liquid helium below 1°K in cooperation with the Ultrasonics Group, and a study of the resistance of pure gold wires down to the lowest attainable temperatures in order to check the abnormal increase in resistance reported by others.

II. D. 5. <u>Classification of Phase Transitions</u>

Staff: Professor L. Tisza M. J. Klein

The method of classification of phase transitions which has been given previously (July 15, 1947 Progress Report) has been investigated mathematically. In general, critical points of a systém will occur at those temperatures for which $\partial^2 G/\partial x^2 = 0$, where G is the Gibbs function and x is any of the parameters on which it depends. The physical nature of the parameter x determines the type of transition which occurs at this critical point. For example, in the case of displacive transitions, x is an internal coordinate of the unit cell which vanishes at the critical point giving rise to an added symmetry element. It has been found for the case of displacive transitions, which are being investigated in detail, that near the critical point, T_c , x will vary as $(T-T_c)^{\frac{1}{2}}$, so that x(T) has a branch point at T_c .

This general method is also being employed for the investigation of physical effects associated with other types of transitions.

II. E. MAGNETIC NUCLEAR RESONANCE EXPERIMENTS

Staff: Professor F. Bitter N. L. Alpert C. G. Lehr S. T. Lin H. L. Poss

1. A Measurement of the Deuteron-Proton Magnetic Moment Ratio

Staff: Professor F. Bitter N. L. Alpert Dr. D. E. Nagle H. L. Poss

During a recent period of availability of the M.I.T. cyclotron magnet, measurements were made of the ratio of the magnetic moment of the deuteron to that of the proton in liquid hydrogen and deuterium by means of nuclear resonance absorption experiments with circuits similar to those previously used¹. The resonances were presented simultaneously on an oscilloscope by means of an electronic switch. A sinusoidal sweep was used, so that with proper adjustment the observed patterns could be made symmetrical about the center-line of the oscilloscope. The shapes of the two resonance

^{1.} F. Bitter, N. L. Alpert, H. L. Poss, C. G. Lehr, S. T. Lin, Phys. Rev. 71, 738, (1947); also RLE Quarterly Progress Report, April 15, 1947, p. 26.

curves were rather different and the most prominent maxima of the deuterium resonance were of the order of a half line width from the hydrogen maxima when the desired symmetry conditions were established. This effect, though not understood in detail, is probably due to the rate of sweep through resonance.

Eleven observations were made, of which three were rejected because the photographs of the oscilloscope traces were found to be unsymmetrical. Half of the remaining eight observations were made with the Dewar containing the resonance coils rotated through 180° so that the effects of a field gradient in the magnet could be eliminated. All eight of these observations fell within the following limits:

$$\frac{\mu_{\rm D}}{\mu_{\rm H}} = .307021 \pm .000005$$

2. Thallium Isotopes

Staff: H. L. Poss

In the October 15, 1947 Progress Report, nuclear magnetic resonances arising from the isotopes of thallium were reported. Measurements of the ratio of resonant frequencies of Tl^{205} and H^1 were given. It has since been possible to make similar measurements on the less abundant isotope, Tl^{203} . The result of the measurements gives for the ratio of the magnetic moments

$$\frac{\mu_{205}}{\mu_{203}} = 1.00986$$

with a probable error of less than .01 per cent slightly higher than the spectroscopic value of 1.00966 obtained by Schüler and Korsching¹. On the basis of their measurements they concluded that the relation $(M_A+2)/M_A = (\mu_A+2)/\mu_A$ appeared to hold for the isotopes of copper, rhenium, and thallium where M_A is the mass of the lighter isotope and μ_A its magnetic moment. The relation appears to be quite exact for thallium since the mass ratio of its isotopes is: 205/203 = 1.00985, the packing fractions of both isotopes being the same.

3. Rotational States in Solids

Staff: N. L. Alpert

The investigation of phase transitions in solids by means of

^{1.} H. Shuler and H. Korsching, Zeits. f. Physik 105, 168 (1937).

nuclear magnetic resonance phenomena has been continued with work on hydrogen bromide and hydrogen iodide. In a previous experiment (October 15, 1947 Quarterly Progress Report) the single transition in HCl at 98° K was studied. In this case no evidence was found that this transition could be identified with a rotational transformation. Similar negative results were found for the two transitions in HI at 70° and 125° K. Results for HB were different, however. Although no apparent change in proton resonance line width was observed at the lowest lambda point, 89° K, a transition from a broad to narrow line was observed at the upper two lambda points, 113° and 117° K, which were unresolved in this experiment. Therefore, we may conclude that molecular rotation sets in at one of these transitions.

A cryostat has been designed and is now being built for purposes of temperature control and measurement probably to better than $1^{\circ}K$. In the preliminary experiments rough methods have left the temperature and temperature equilibrium of the sample in doubt. The new cryostat consists of a copper pot containing the sample and r-f coil connected to a low (or high) temperature reservoir through an electrically variable heat leak.

4. New Bridge Circuit

Staff: H. L. Poss

An r-f bridge circuit has been constructed which does not make use of half and quarter wavelength coaxial cables as have our previous higherfrequency circuits. With it, proton resonances have been observed at 1.5 Mc/sec and 0.8 Mc/sec. These frequencies require only low fields, 350 and 185 gauss, respectively, for protons.

5. <u>New Electromagnet</u>

Staff: Professor F. Bitter

A new electromagnet with 8-in. diameter poles giving fields up to 10,000 gauss with a $2\frac{1}{4}$ -in. gap has been completed. Submarine storage batteries to operate this magnet are being installed.

F. PARAMAGNETIC AND FERROMAGNETIC RESONANCE ABSORPTION AT MICROWAVE FREQUENCIES

Staff: Professor A. F. Kip R. D. Arnold

Paramagnetic Resonance. Resonance absorption in a single crystal of CuSO₄·5H₂O has been observed for various orientations of the crystal with respect to the applied constant magnetic field. At a frequency of 9050 Mc/sec and with the axis of minimum susceptibility parallel to the field, an absorption peak occurred at 3030 gauss¹. At the same frequency and with the axis of minimum susceptibility perpendicular to the field, the absorption peak was at 2810 gauss. Since the susceptibilities along the two principal axes normal to the axis of minimum susceptibility is parallel to the field when the axis of minimum susceptibility is parallel to the field. Between the perpendicular and parallel orientations the field for resonance varies as the cosine squared of the angle between the minimum susceptibility axis and the applied field. The shape and orientation of the susceptibility ellipsoid as found by Krishnan and Mookherji² are assumed in this experiment.

For the single crystal the line width is about 100 gauss. An absorption line for $CuSO_4 \cdot 5H_9O$ powder (Fig. 1) has the shape expected on



Figure 1. Absorption in CuSO₄•5H₂O powder. Dotted lines show resonance field values for perpendicular and parallel orientations of the crystal.

Relative error in field values ± 1 per cent; absolute error ± 5 per cent.
 K. S. Krishnan and A. Mookherji, Phys. Rev. <u>50</u>, 860 (1936).

the basis of the single crystal data, the greater absorption at the lowfield end of the line being caused by the fact that two principal axes correspond to resonance at the lower field and one principal axis to resonance at the higher field.

Single crystals of $K_2Cu(SO_4)_2 \cdot 6H_2O$ are being prepared for similar experiments.

<u>Ferromagnetic Resonance</u>. The failure to observe anisotropy in a single crystal of iron reported in the preceding progress report was evidently due to strains in the crystal. Bell Telephone Laboratories, who originally provided the crystal, have treated it so as to reduce strains and have returned it to this Laboratory where an anisotropy has now been observed. In the experiment the crystal (cubic) is mounted with the 100 plane parallel to the constant magnetic field and to the r-f magnetic field, and is rotatable in this plane. The crystal is in the form of a thin disk (0.0085 in. x 0.85-in. diameter) with the short dimension in the 100 direction. Table I gives the magnetic field for maximum absorption at different crystal orientations. These results are consistent with

Band	f	Crystal Direction Parallel to Magnetic I				c Field
	Mc/sec	010	011	001	011	
V	93 670	3280	2480	3280	2600	relige
v	23,070	9260	2400	3200	2000	gauss
Λ	5,200	900	210		220	Ranss

TABLE I. Resonance field for different crystal orientations.

Kittel's theory¹. In the K-band experiment the field for resonance at directions between OlO and Oll follows a $\cos^2 2\theta$ law. In the X-band case, however, there were large deviations from the $\cos^2 2\theta$ dependence, and in the Oll and Oll directions a second absorption peak was observed at a field lower than that listed in Table I. It is thought that these two effects can be explained by the fact that at the low field values required for resonance at X-band, the magnetization in an iron crystal is not aligned with the applied magnetic field. The discrepancy in Table I between the Oll and Oll directions may result from residual strains in the crystal.

Scheduled for publication, Phys. Rev. January 15, 1948; also Technical Report No. 49, C. Kittel, "Crystalline Stark Splitting and Microwave Resonance Absorption in Paramagnetic Salts", September 10, 1947.