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M. W. P. STRANDBERG
M. TINKHAM
I. H. SOLT, JR.
C. F. DAVIS, JR.

TECHNICAL REPORT 304
MAY 10, 1955

RESEARCH LABORATORY OF ELECTRONICS
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
CAMBRIDGE, MASSACHUSETTS

Reprinted from THE REVIEW OF SCIENTIFIC INSTRUMENTS, Vol. 27, No. 8, pp. 596–605, August, 1956

The Research Laboratory of Electronics is an interdepartmental laboratory of the Department of Electrical Engineering and the Department of Physics.

The research reported in this document was made possible in part by support extended the Massachusetts Institute of Technology, Research Laboratory of Electronics, jointly by the Army (Signal Corps), the Navy (Office of Naval Research), and the Air Force (Office of Scientific Research, Air Research and Development Command), under Signal Corps Contract DA36-039 SC-64637, Project 102B; Department of the Army Project 3-99-10-022.

Reprinted from THE REVIEW OF SCIENTIFIC INSTRUMENTS, Vol. 27, No. 8, 596–605, August, 1956
Printed in U. S. A.

Recording Magnetic-Resonance Spectrometer*

M. W. P. STRANDBERG, M. TINKHAM,† I. H. SOLT,‡ JR., AND C. F. DAVIS, JR.§
Department of Physics and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received April 13, 1956; revised version received May 16, 1956)

Apparatus especially designed for studying electron paramagnetic resonance is described and discussed. A magnet of novel yokeless design is presented. Field stabilization and modulation procedure is considered. The microwave sample cavity is analyzed to determine conditions for optimum operation. The klystron stabilization problem is examined. Appropriate lumped circuits for low-frequency operation are described. The signal amplification and presentation system is treated in detail.

I. MAGNETIC FIELD STABILIZATION AND CONTROL SYSTEM

IN an electron paramagnetic-resonance experiment one usually seeks to measure the imaginary or absorptive part χ'' of the susceptibility of the sample as a function of the frequency ν of the rf field and of the magnitude H of the static magnetic field. In many cases there is a resonance absorption line at a frequency that can be related to the magnetic field by the relation[1]

$$h\nu = g\beta H \quad (1)$$

where β is the Bohr magneton, g is the spectroscopic splitting factor, and h is Planck's constant. If Eq. (1) is valid, the resonance can be observed at fields and frequencies as low as desired. For example, in the common case of $g=2$, then $\nu/H = g\beta/h = 2.8$ Mc/gauss. However, the signal strength increases with frequency. Since the line width in gauss is usually independent of the frequency and field at which the resonance is observed, greater accuracy in measuring the resonance magnetic field is obtained by working at higher fields and frequencies. These considerations and the availability of microwave oscillators of appropriate frequency have led to the use of magnetic fields in the region of 0–12 kilogauss.

To avoid distortion of the absorption curves, the magnetic field must be homogeneous over the sample volume. The sample volume may also be limited by the size of available single crystals. To obtain a measurable absorption in a small volume of sample, the energy

* This work was supported in part by the U. S. Army (Signal Corps), the U. S. Air Force (Office of Scientific Research, Air Research and Development Command), and the U. S. Navy (Office of Naval Research).

† Now at University of California, Berkeley, California.
‡ Now at Hughes Research Laboratories, Culver City, California.
§ Operations Research Group, The Johns Hopkins University.

[1] If the energy levels are not linear in H, or if there are initial splittings from Stark effect, residual spin-orbit coupling, or hyperfine structure, this simple formula is satisfied only if g is allowed to be a function of the frequency at which the resonance is observed. Clearly, such a g has no basic physical significance. This fact has been overlooked in much work in this field because of the inconvenience of taking data at more than one frequency. In some of these cases it may be necessary to use frequencies that lie in specified ranges if the transition between two particular energy levels is to be observed at all, even allowing unlimited values of H.

density of radiation is increased by putting the sample in a high-Q resonant cavity. Usually no trouble from saturation is encountered because relaxation times are quite short, and the transition probabilities for magnetic transitions are relatively small.

A very convenient form for the data on the resonance absorption would be a continuous plot of $\chi''(\nu)$ at constant H. Since it is experimentally difficult to sweep the resonant frequency of the cavity containing the sample it is more usual to hold ν constant and sweep H, obtaining $\chi''(H)$ at constant ν. To the extent that Eq. (1) holds, a run at any fixed frequency would then determine the parameter g.

In actual practice it is more convenient to measure $d\chi''/dH$ as a function of H at constant ν. To do this, we superimpose a modulation at, let us say, 50 cps upon the slowly sweeping quasi-static magnetic field. If this modulation amplitude is small compared to the width of the absorption line, the 50-cps modulation component of the absorbed power is then clearly proportional to $d\chi''/dH$. To avoid line-shape distortion, the amplitude and phase of the modulation must be independent of the quasi-static H. If $\chi''(H)$ itself has to be obtained from this derivative, an electronic integrator can be used. Since this type of integrator integrates with respect to time, not H, a strictly linear relation between H and t in the quasi-static sweep is highly desirable.

Let us now review our design requirements and then go on to consider a particular means of implementation. We require a magnet capable of producing fields up to 12 000 gauss that are homogeneous over the sample volume, which, for gaseous samples, equals the cavity volume. The field must be stable to the same degree to which it must be homogeneous, that is, to much less than a line width. The width of a resonant absorption may be as small as 1 gauss. It must be possible to sweep the stabilized field linearly in time over a large portion of the available field and at a wide variety of sweep rates (e.g., 1–1000 gauss/minute). A modulation field controllable in amplitude from zero to the order of 100 gauss (peak-to-peak) and of stable amplitude and phase must be available without disturbing the stability of the quasi-static field. Finally, it must be possible to measure the quasi-static field easily and quickly, and to measure the static field to a high degree of accuracy.

A. Magnet Design

The electromagnet we use is a novel yokeless type.[2] A relatively inexpensive and easily adjustable magnet with independent mounting of the two sections is thus possible. The large cross section of the air return path provides a reluctance that is small compared to that of the relatively long gap.

The power efficiency of a magnet, η, may be defined

[2] We are indebted to Francis Bitter, of this Laboratory, for suggesting that this design be investigated.

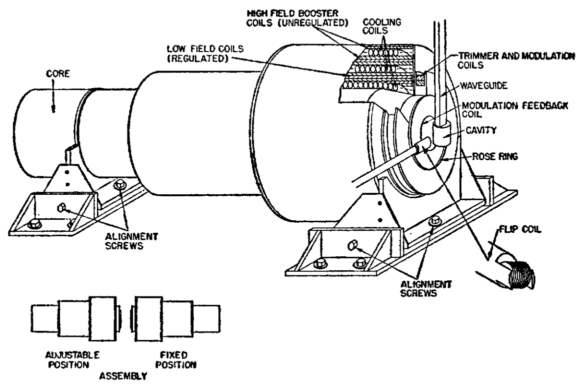


FIG. 1. Magnet.

by

$$B=\eta\frac{\mu_0 Ni}{h},\quad \text{or}\quad B=\eta\times 1.6\times 10^{-5}\frac{(MP)^{\frac{1}{2}}}{rh}$$

where Ni represents the ampere turns, h is the gap length, μ_0 is the permeability of the gap, B is the flux density in the gap,[3] M is the mass of the copper (windings), P is the power input, and r is the average radius of the windings. All units are rationalized mks units.

Studies were conducted on a scale model, flux measurements being made ballistically. Experimentation showed that with the gap length about one-third of the pole diameter and the cores of reasonable length (about three times the core diameter) the efficiency remained in the range of one-third, up to the point of core saturation. The model also demonstrated that when the core becomes saturated at some point the efficiency is almost independent of the magnetic return.

It is interesting to notice that core saturation effects may begin to lower the power efficiency for gap flux densities as low as 7000 gauss (one-third of the saturation magnetization of iron). This saturation results from gap fringing effects and a long gap (one-third of the pole diameter) and can be reduced by tapering the core. A taper amounting to 44% reduction in the core diameter over a length of one core diameter is used on our magnet. This taper gives about a one-third increase in gap flux density. The optimum angle will depend, of course, on gap geometry and on the type of core material.

A magnet that was built to these specifications in the laboratory is shown in Fig. 1. The core is $9\frac{3}{4}$ inches in diameter, tapered to 8-inch Armco steel pole pieces. The windings are stepped, 70% of the copper being within one core diameter of the pole pieces. With an input of 12 kilowatts, a field of 11 400 gauss is obtained across a $2\frac{5}{8}$-inch gap. The same power input produces only 13 500 gauss at $1\frac{5}{8}$ inch spacing. This exemplifies

[3] Flux density is represented here by B according to the mks system; the units are webers per square meter (=10 000 gauss). Elsewhere in this paper H will be used for flux density, as is conventional in paramagnetic resonance work, but the unit used will be the gauss.

the decrease in power efficiency of this design for relatively small gaps.

Shims were calculated for the pole pieces by the formula of Rose[4] in an attempt to make the field homogeneous over a larger volume. The homogeneity is roughly 0.02% over a diameter half as great as the pole pieces. The two sections of the magnet are held apart by three Dural blocks that are within 0.0002 inch of the same length. Great care must be taken to align the pole pieces accurately, since a few hundredths of a degree departure from parallelism results in prohibitive inhomogeneity.

The field is modulated, and fine control is obtained by a pair of trimmer coils mounted around the pole pieces. These coils each consist of two sections of 2462 and 1650 turns of #22 B&S wire wound on Bakelite forms (see Secs. I B and C).

The advantages of a design of this type lie largely in the low cost. Accurate machine work is required on those parts associated with the shape and spacing of the pole pieces, but tolerances are ample on other parts. Once misaligned the pole faces are relatively easily realigned with the ample adjustment screws that are provided. Only a straight edge and inside micrometer are needed to check alignment. There is also ready access to the gap from all sides.

Undesirable features of the design include the large "stray" fields, which are as much as a hundred gauss several feet from the magnet and are measurable at a much greater distance. Although the efficiency at saturation fields is comparable with that of a similar construction with return yoke, this efficiency does not measurably improve at low fields and becomes worse for smaller gaps. The flexibility of design permits easy adjustment but requires that the magnet be realigned often in order to maintain the desired homogeneity.

B. Magnetic Field Stabilization and Sweep

The magnetic field in the gap of the magnet described above is continuously monitored by a rotating flip coil driven by an 1800-rpm synchronous motor (Bodine NSY–12). The flip-coil shaft must be of nonconducting material over the length which is in the field. This is to keep eddy-current dissipation from loading the motor excessively and heating the shaft, with consequent change of area of the flip coil from thermal expansion. (With a brass shaft, the loading is 1/75 hp at 10 000 gauss for typical dimensions.) The output voltage of the flip coil, which is proportional to H and is of the order of a few volts, is compared with a fraction of the output of a reference generator (Elinco FS–15) driven on the same shaft. In this generator a permanent magnet rotates between stationary coils, making the output independent of any stray magnetic fields that might be present. The fraction of the reference voltage used is determined by a ten-turn helical potentiometer that can be adjusted manually or swept at any desired speed by a geared-down synchronous motor. To allow exact cancellation of the flip-coil and reference voltages, the phases are set 180° apart by first mechanically adjusting the relative orientations on the motor shaft and then using a decade capacitor box across the reference generator as a vernier phase adjustment.

These opposing voltages are added in the 5691 (RCA "red tube" equivalent of the 6SL7) twin-triode mixer as shown in Fig. 2(a). Because of the near cancellation, the plate voltage variation is small, and this circuit resembles a cathode follower in that its gain from either grid is near unity and relatively insensitive to tube characteristics. Also, the input impedance is raised to several megohms by the cathode follower action. This isolation is necessary to avoid loading the helical potentiometer because loading would reduce the linearity of the sweep. A "red tube" is used here for additional stability, since even the existence of the feedback loop cannot correct for any errors or nonlinearity introduced in this stage.

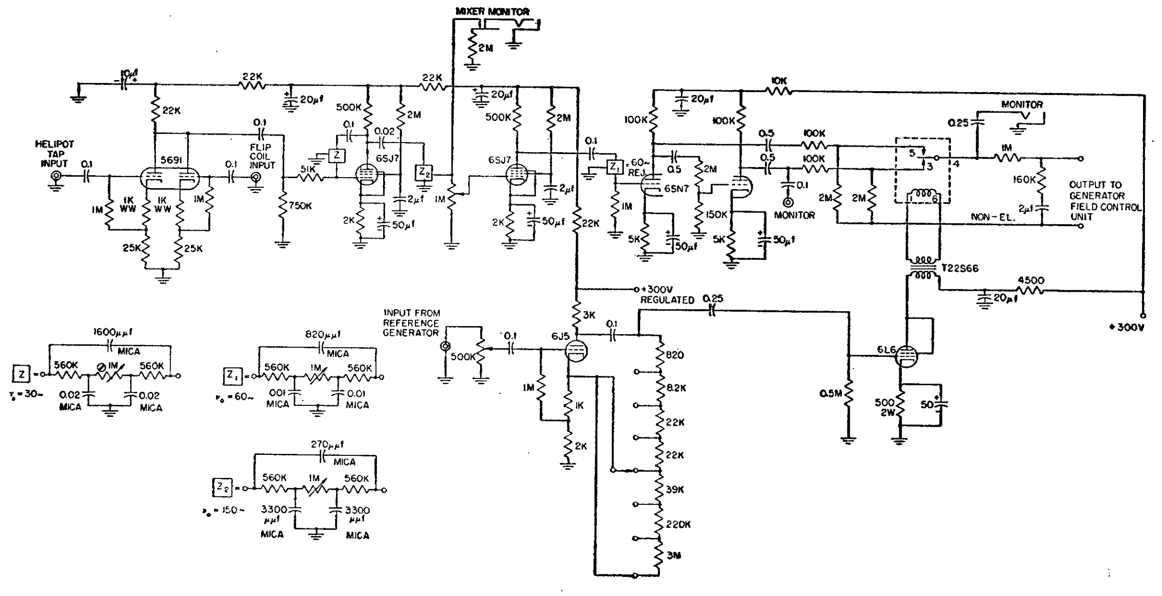
The difference, or "error," voltage from this mixer is amplified in the following 30-cps tuned amplifier equipped with RC filters (Z_1 and Z_2) tuned to reject 60 cps and 150 cps.[5] The 150-cps filter is required because of the large fifth-harmonic content in the output of the reference generator. The amplified voltage undergoes full-wave synchronous detection in the Brown converter. The vane in this converter is driven by the amplified output of the reference generator. Its phase is so adjusted that the output of the detector is positive if the flip-coil voltage is less than the fraction of the reference voltage derived from the helical potentiometer, and is negative in the reverse case. The RC circuit at the output is designed to provide 16 db more gain at zero frequency than at high frequencies. This allows the effective loop gain of the servo to be higher without oscillation than would otherwise be possible.

The output of this unit drives a multistage dc amplifier, shown in Fig. 2(b). The final stage of this amplifier is a bank of seven parallel 815 tubes supplied with suitable parasitic oscillation suppressors. These tubes are capable of supplying 0–1.25 amp to the field coils of the motor generator that supplies current to the inner windings of the magnet. (To enable the apparatus to overcome hysteresis and drive positively to zero field, a single 815 tube is connected to provide a constant negative bias current to the motor generator field coils.) In this manner we close the loop and stabilize the magnetic field against the reference generator. As the helical potentiometer is swept linearly in time by the geared-down motor, the field is also swept linearly and is stabilized to the instantaneously correct value.

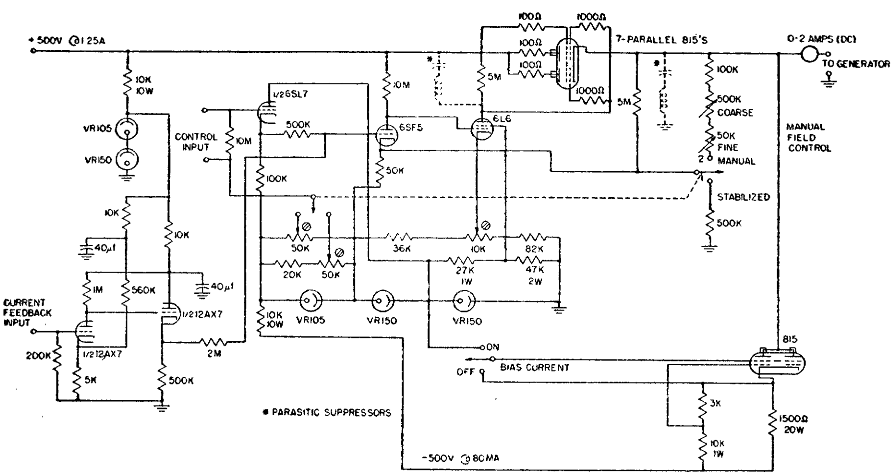
Actually, the servo loop oscillates if it is operated

[4] M. E. Rose, Phys. Rev. 53, 715 (1938).

[5] Quarterly Progress Report, Research Laboratory of Electronics, M.I.T., April 15, 1948, p. 36.



(a)



(b)

FIG. 2. (a) Field stabilization discriminator. (b) Field stabilization dc power amplifier.

exactly as described above. The basic reason is that there are long lags associated with both the motor generator and the magnet. They introduce such phase shifts that the feedback becomes regenerative at a frequency at which the loop gain still exceeds unity. To make the loop stable we introduce an inner negative feedback loop that applies amplified current feedback from the current in the magnet winding to the input of the dc amplifier that drives the 815 tubes. Since this loop encloses the motor generator and the inductive lag of the magnet windings, it tightens up the response of these elements, greatly reducing the associated lags. This renders the over-all loop, including the additional lags in the iron core and in the circuitry, stable at loop gains up to the order of 100.

With this current feedback operating, the field is stable to 1 gauss in 3000, but the response is still moderately slow and underdamped. The control may be further tightened by applying the output current from an alternate bank of controlled 815 tubes directly to the magnet through high-impedance trimmer coils. This completely avoids the lag in the generator and in the massive iron core by directly controlling the field in the gap. As a result, the response time is reduced to a fraction of a second, and the field is stable to a few tenths of a gauss at 3000 gauss. When this type of feedback control is used, the motor generator is put on nonstabilized manual control to provide a bias field. Oscillation results if an attempt is made to apply the feedback control both through the trimmer coils and through the motor generator.

The generator feedback system provides for a continuous linear sweep over any range between 0–10 000 gauss at any speed between 0.5 and 3000 gauss/min by suitable choice of the available gear ratios[6] in the reference helical potentiometer drive. With direct tube control, the sweep range available without changing the bias field is only 200 gauss. This is no real limitation with lines narrow enough to require high stability for satisfactory measurement.

C. Magnetic Field Modulation

A sinusoidally varying (50-cps) field is superimposed on the quasi-static magnetic field. This modulation is produced by a power amplifier that drives two coils (2462 turns on each), one around each pole piece of the magnet. For a constant input to the amplifier, the actual modulation amplitude depends upon the degree of saturation of the magnet core by the quasi-static field. For fields changing from 0 to 10 000 gauss, the peak-to-peak modulation would change by 25% and the phase by 15°. These changes are reduced by the introduction of a feedback loop. A small pickup coil is placed on one pole face of the magnet. The voltage induced in this coil is proportional to the modulation amplitude. It is amplified and mixed degeneratively with the 50-cps source in a twin triode mixer (see Fig. 3). The output of this mixer is amplified and applied to the modulation coils. The resulting system has a stable loop gain of approximately 7, thus reducing the amplitude variations to 4% and the phase deviations to 2°, which is satisfactory for our experiments. With an amplifier capable of delivering 45 watts, a peak-to-peak modulation amplitude of 70 gauss can be obtained. Continuous variation of the amplitude is accomplished with a calibrated potentiometer across the 50-cps source.

The choice of the modulation frequency used is not critical. The lower limit is set by the increase of crystal noise at low frequencies and the upper limit by the increasing shielding effect resulting from the induced eddy currents both in the magnet pole faces and walls of the microwave cavity. Frequencies up to several hundred cycles per second would be satisfactory. Our particular choice was governed by the availabilty of a very stable 50-cps frequency from the M.I.T. frequency standard. A stable frequency is essential for avoiding inconvenient phase shifts in the tuned amplifier in the instrumentation described later.

The factors motivating the design of paramagnetic resonance spectrometers, except for the magnet, are essentially the same as those discussed by Strandberg, Johnson, and Eshbach.[7] In each case, the field modulation frequency is determined by similar considerations. Since it is difficult to square-wave-modulate a magnetic field, sine-wave or slope-taking modulation is used here despite the obvious disadvantage that the optimum modulation amplitude depends upon the line width. As another example of design similarity it seems necessary to point out that—for reasons indicated by Strandberg *et al.* in the earlier paper—the use of superheterodyne detection to avoid crystal noise is of no avail. The reason is simple. The crystal detector is considered a voltage source with an internal resistance. A large portion of this resistance is time-invariant; a small portion is randomly amplitude-modulated. The crystal low-frequency noise is the result of modulating the dc crystal current by the component of variable resistance. Similarly, if the crystal is the source of an intermediate frequency (i.f.), as in a superheterodyne detector, the i.f. signal will also be modulated by the variation in the crystal-source resistance, and the crystal "low-frequency" noise appears as a proportional modulation on the i.f. signal. As we indicated in the earlier paper, only by bridging out the main, or carrier, i.f. component can any net crystal noise reduction be achieved.

The presence of the modulation field has little effect on the stabilization of the quasi-static magnetic field through the motor generator, since the response is quite low at 50 cps. However, peak-to-peak modulation amplitudes of 20 gauss, or more, cause the system to oscillate when stabilization through the trimmer coils is used. This is no serious limitation, since the field stabilization through the generator is sufficient for absorption lines that are broad enough to require modulation amplitudes of 20 gauss.

D. Magnetic Field Measurement

The easiest and quickest way to measure the quasi-static magnetic field is in a manner similar to that used for the stabilization described in Sec. *B*. An accurate step potentiometer, consisting of two General Radio decade resistance boxes, is connected in parallel with the helical potentiometer across the reference generator. The voltage appearing at the tap is mixed with the output of the flip coil in a unit similar to that of Fig. 2 without the demodulator. The output of the amplifier is applied directly to an oscilloscope. To measure a field, the potentiometer, which is calibrated directly in gauss, is adjusted for a null. Fields can be read to 0.5 gauss.

The accuracy of this system depends upon the linearity of the relation between the field at the flip coil and the field at the sample. For our magnet these fields were linearly related up to 5000 gauss; saturation effects produced an increasing nonlinearity for larger fields. The calibration also depends upon the stability of the Elinco generator. Temperature drifts of about 1 part in 3000 were observed for long periods of continuous operation. For reasons given in Sec. I B, the mixer circuit should introduce little nonlinearity. Even though the absolute accuracy is not always satis-

[6] To avoid discontinuities from the finite number of wires in the helical potentiometer winding, "electrical band spread" is used for the slower sweeps; resistance boxes are used to provide the coarse field setting, and the ten-turn helical potentiometer covers a range of 1000 gauss instead of 10 000 gauss.

[7] Strandberg, Johnson, and Eshbach, Rev. Sci. Instr. **25**, 776 (1954); or M. W. P. Strandberg, *Microwave Spectroscopy* (Methuen monograph, 1954).

factory for precise measurements, the relative accuracy is quite sufficient to make the system ideal for applying field marker pips to chart recordings.

For accurate magnetic field measurements, a proton resonance apparatus similar to that described by Knoebel and Hahn[8] was constructed. Since we have magnetic field modulation available, the frequency modulation in their design could be eliminated.

II. MICROWAVE AND SIGNAL CIRCUITS

In Sec. I we described an apparatus that will impose a homogeneous magnetic field upon a sample of restricted volume and will sweep the field linearly in a quasi-static manner while modulating it at 50 cps. We now describe the apparatus by which the susceptibility data is obtained from the sample subjected to this field.

A. Cavity Loading and Coupling

The two important parameters of the microwave cavity in which the sample is placed are its resonant frequency ν and its Q. If we are content with the first-order theory, in which we neglect the distortion of the microwave field in the cavity by the sample, the effect of the sample is[9]

$$\frac{\Delta\nu}{\nu}=-\tfrac{1}{2}\chi_e'\int_{V'}E_r^2dt-\tfrac{1}{2}\chi_m'\int_{V'}H_r^2dt$$

$$(1/Q)_s=\Delta(1/Q)=(\chi_e''+\sigma/\epsilon_0 2\pi\nu\epsilon_0)\times\int_{V'}E_r^2dt+\chi_m''\int_{V'}H_r^2dt \qquad (1)$$

where $\chi_0=\chi_e'-i\chi_e''$ and $\chi_m=\chi_m'-i\chi_m''$ are the (rationalized) electric and magnetic susceptibilities of the sample, respectively; E_r and H_r are rf field strengths normalized over the cavity volume; V' is the sample volume; and ϵ_0 is the permittivity of free space. Since the only two terms that depend strongly on the quasi-static H are χ_m' and χ_m'', we restrict our consideration to them. Inspection of Eq. (1) shows that these give the largest effects when large samples are placed at points where H_r is large. Note that $(1/Q)_s$ is numerically equal to χ'' (rationalized) or $4\pi\chi''$ (unrationalized) if the sample fills the entire cavity, as it does in the study of gases.

In the apparatus to be described the microwave source frequency is kept locked to the resonant frequency of the cavity despite changes produced by the sample. Thus χ' may be determined by measuring the frequency shift of the oscillator *versus* the quasi-static H. There is more interest, however, in measuring χ''. This is done by measuring the changes in the power reflected from or transmitted through the cavity as we sweep H. Let us determine the conditions that maximize the signal obtained in this way. It can be shown[9] that the power reflection coefficient of a cavity at resonance is

$$|r|^2=[(\xi-1)/(\xi+1)]^2 \qquad (2)$$

where $\xi=Q_e/Q_0$; Q_e is the external Q (which measures the loss of stored energy through the coupling to the outside) and Q_0 is the unloaded cavity Q. From this one may readily show that

$$\Delta|r|^2=4\xi(\xi-1)(\xi+1)^{-3}Q_0(1/Q)_s \qquad (3)$$

is the change produced by an absorption $(1/Q)_s$. Differentiation of the coefficient of $(1/Q)_s$ with respect to the coupling parameter Q_e shows that the maximum change is

$$\Delta|r|^2_{\max}=0.385Q_0(1/Q)_s \qquad (4)$$

which occurs as a broad maximum when the coupling is adjusted to give

$$\xi=2\pm3^{\frac{1}{2}} \quad \text{or} \quad |r|^2=\tfrac{1}{3}. \qquad (5)$$

An optimum size for solid samples also exists. As the sample volume V' becomes a larger fraction of the cavity volume, the competing effects are the increasing filling factor and the decreasing Q_0 resulting from the electric losses in the sample. Since the sample is normally placed at a point where H_r is a maximum and E_r passes through zero, we treat H_r as a constant over the sample and treat E_r as passing linearly through zero at the center of the sample (e.g., a sample located in the central plane in a rectangular TE_{102} mode cavity). If we consider a cylindrical sample (such as the capillaries used in much work in our laboratory), then, as we increase the radius, Eq. (1) shows that the electric losses increase as V'^2. If we consider a sample in the form of a flat plate covering the entire cross section of the guide, we can insert the sample close to the $E_r=0$ plane; but, as the thickness is increased, the loss rises as V'^3. Quite generally, then, we assume

$$1/Q_0=1/Q_c+\alpha V'^n$$

where $1/Q_c$ gives the loss to the cavity walls alone. Using this relation and the assumption that $(1/Q)_s$ is proportional to V', we find that the maximum signal occurs when V' is chosen to give a Q_0 satisfying

$$1/Q_0=[n/(n-1)](1/Q_c). \qquad (6)$$

Since n is of the order of 2 or 3, this shows that the sample of optimum size is one in which the electric

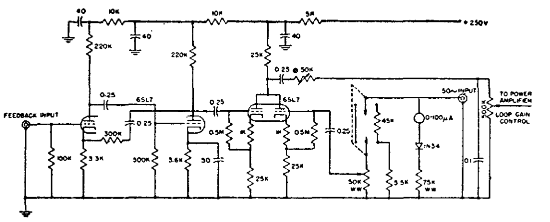


FIG. 3. Modulation feedback loop.

[8] H. W. Knoebel and E. L. Hahn, Rev. Sci. Instr. 22, 904 (1951).
[9] J. C. Slater, Revs. Modern Phys. 18, 441 (1946).

losses in the sample are of the order of one-half to one times the energy loss to the cavity walls. If we convert the Q's to values of $|r|^2$ (the easily observed quantity) by means of Eq. (2), we obtain the following rule for maximum signal: choose the window size (Q_e) and the sample size V' so that $|r|^2$ rises from 0.09 for cylindrical samples, or 0.15 for flat samples, to 0.33 when the sample is inserted. Since this treatment assumed an unperturbed field distribution, it should be considered only a first approximation if the samples have high dielectric constants or conductivities.

For comparison, if a transmission cavity is used, the power transmission factor at resonance is

$$|t|^2 = 4Q_L^2/Q_1Q_2 \tag{7}$$

where $Q_1 = (Q_e)_{\text{input}}$, $Q_2 = (Q_e)_{\text{output}}$, and Q_L is the loaded Q defined by

$$1/Q_L = 1/Q_0 + 1/Q_1 + 1/Q_2. \tag{8}$$

If we have an absorption $(1/Q)_s$, the change in T is

$$\Delta|t|^2 = -8Q_L^3/Q_1Q_2(1/Q)_s. \tag{9}$$

The condition for maximum signal is $Q_0 = Q_1 = Q_2$. In this case, $|t|^2 = 4/9$, $|r|^2 = 1/9$, and

$$\Delta|t|^2_{\text{max}} = -0.296Q_0(1/Q)_s. \tag{10}$$

The fact that the maximum signal in transmission is somewhat less than that in reflection may be understood by noting that some of the information is wasted as modulation of the reflected signal that is present even in a transmission cavity.

We have now examined the general factors that govern the optimum conversion of the susceptibility of the sample to a change in reflection or transmission coefficient of a resonant circuit. The remainder of the paper will describe a particular arrangement for producing a recording of the susceptibility from these coefficients.

B. Radio-Frequency Instrumentation

1. *Microwave System*

As shown in Sec. II A, when the magnetic field is swept through a resonance, the change in reflection coefficient from the cavity is proportional to the imaginary part of the susceptibility. However, this result holds only when the klystron frequency corresponds to the center frequency of the microwave cavity containing the sample. If the klystron frequency drifts off the center frequency of the cavity, the change in reflection coefficient will then be proportional to a mixture of both the real and the imaginary parts of the susceptibility. Hence, to obtain meaningful data, it is necessary to stabilize the klystron frequency to the sample cavity.

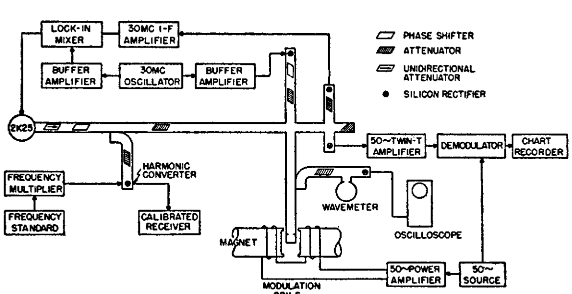


FIG. 4. Microwave and signal instrumentation.

The analytic or interpretive simplicity of using only one component of the susceptibility is not the only reason for locking the klystron in a stiff fashion to the resonant frequency of the sample cavity. Ingenious use of the same cavity for frequency stabilization and sample analysis allows the study of one component of the susceptibility and, at the same time, reduces the signal low-frequency noise spectrum that results from the random frequency drift of an unstabilized klystron about the sample cavity resonance. Hence the signal-to-noise ratio is enhanced. Furthermore, if the klystron is held precisely at the sample cavity resonance, the cross-conversion of the unavoidable klystron frequency-modulation noise into amplitude-modulation noise by the cavity selectivity is minimized. It can be shown experimentally that precise alignment of the klystron and cavity yields an improvement of 10 db, or more, in the signal-to-noise ratio.

A block diagram of the microwave and signal circuit instrumentation is given in Fig. 4. Although a 2K25 3-cm klystron is indicated in this figure, the instrumentation to be described will work satisfactorily at 10 cm with a type 707B tube or at 1.25 cm with a type 2K50 tube. At 10 cm when coaxial line is used instead of waveguide a hybrid ring or "rat-race" can be substituted for each "magic tee" shown in the figure. For stabilizing the klystron to the absorption cavity, either the dc or i.f. Pound[10] stabilization system can be used. However, the i.f. system as modified by Zaffarano[11] is recommended because it was found to be more stable and reliable. A general discussion of its operation is given in the references, and will not be repeated here.

Since for reasons of sensitivity it is desirable to work at as large a power level as possible (provided saturation has not been reached), a minimum attenutation between the generator and absorption cavity should be used, while still maintaining sufficient isolation to prevent "pulling" of the generator frequency by the reactive cavity.[9] This is accomplished with a unidirectional ferrite isolator. If ferrite isolators are not available, the pulling can be minimized by inserting an appropriately adjusted phase shifter between the klystron and cavity.

After the isolator, the power goes to a variable attenuator for setting the power level and then to a magic tee, where it is split equally between the two

[10] R. V. Pound, Rev. Sci. Instr. 17, 49 (1946).

[11] F. P. Zaffarano and W. C. Galloway, Technical Report No. 31, Research Laboratory of Electronics, M.I.T. (1947).

symmetric arms. One arm goes to the absorption cavity containing the sample under investigation; the other goes through another variable attenuator and phase shifter to the 30-Mc modulator crystal of the Pound stabilizer. The purpose of the last attenuator is to allow the introduction of sufficient padding to damp out spurious reflections when making absolute intensity measurements. The 30-Mc modulated reflection from the modulator crystal and the reflection from the cavity combine in the magic tee, half going out the input arm of the tee and being dissipated in the isolator, and the other half going to a second magic tee. Here it is again divided, half going to the mixer crystal for the Pound stabilizer and the other half to the detection crystal of the signal circuit. It is necessary to have separate detectors for the Pound mixer and signal circuit; the bolometer used as the signal detector in making absolute intensity measurements cannot be used for the Pound mixer.

In working with a Pound stabilizer, several precautions must be taken. First, because of dispersion in the wave guide, it is advisable to have equal lengths of line from the first magic tee to the 30-Mc modulator and to the absorption cavity. This precaution eliminates ambiguities that arise in adjusting the phase of the discriminator. Second, the stabilizer does not necessarily lock the klystron exactly to the center frequency of the cavity. The precision depends upon the exact setting of the phase shifter in the modulator arm. So that its proper adjustment can be checked, the reflected power from the cavity is monitored by a directional coupler and crystal on the cavity arm (see Fig. 4). The phase shifter is then adjusted to give minimum crystal current.

For most of the work in which the highest sensitivity is not necessary, a rectangular absorption cavity of the TE_{10n} mode is the most convenient to use. One end of a section of wave guide is soldered closed. The other end is temporarily closed by a metal sheet with a coupling hole in it. This arrangement has the advantage of allowing the coupling irises to be readily changed to accommodate a wide variety of samples with different losses.

In adjusting the windows, it is convenient to remember that for small windows the coupling is proportional to the cube of the area of the window, to the square of the amplitude of the incident field, and to the square of the amplitude of the matching field in the cavity of the window as normalized over the cavity volume.[12] The irises that form one wall of the cavity are clamped between the remainder of the cavity and the input wave guide. Since the irises are made of silver, they are ductile enough to give good electric contact around the whole edge of the cavity, insuring a maximum Q. The cavity itself is plated with high-purity gold or silver to maximize the Q and minimize possible spurious signals from paramagnetic impurities in the walls.

[12] H. A. Bethe, Phys. Rev. **66**, 163 (1944).

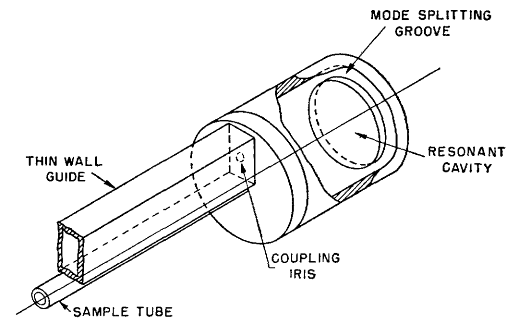


FIG. 5. Resonant cavity.

The gold-plated cavity is freer from spurious "empty-cavity" resonances than any of the others. The sample is placed in a plane of maximum rf magnetic field. Liquid and powder samples are put into 2-mm capillaries that can be easily inserted into the cavity through small holes in the walls. Single crystals can be fastened to a rotating jig that is mounted in the same plane in an equivalent cavity.

The typical design problems encountered are exemplified in the solutions shown in Fig. 5. This is a cylindrical cavity using the TE_{011} mode. The mode is accidently degenerate with the TM_{111} mode. For proper control of the field configuration this degeneracy must be removed. This is accomplished by the groove in the bottom plate, since the groove interacts only with the TM_{111} mode. For low-temperature work the cavity is immersed in a cryostat so that the main component of the rf H field (longitudinal) is parallel to the cryostat axis and perpendicular to the static magnetic field. This is most conveniently accomplished by coupling through a hole in an end plate from the end of the wave guide. Since the guide must have a minimum of heat conduction, it is thin-walled. To allow a mechanism for sample orientation (if a crystal is to be used) a hole for a dielectric rod is available on the axis of the cavity. The dimensions are such that a wave guide smaller than regular size must be used in the cryostat. An external taper section allows for coupling to a guide of regular size.

The design and use of special cavities for circular polarization studies have been described elsewhere.[13]

For measuring the frequency, calibrated cavity wave meters can be used with an ultimate accuracy of about 1 part in 10^4 when temperature and humidity corrections are made. When greater accuracy is desired for making g-value measurements, the frequency must be measured directly by beating the microwave frequency with a known harmonic of a crystal frequency standard. The beat frequency is measured with a calibrated tunable receiver. The accuracy of this method is limited only by the accuracy of the crystal standard and the stability of the receiver.

[13] M. Tinkham and M. W. P. Strandberg, Proc. Inst. Radio Engrs. **43**, 734 (1955).

2. *VHF System*

For certain investigations it is desirable and sometimes necessary to work at much lower frequencies. An example of this is paramagnetic resonance absorption by conduction electrons in metals. Here samples of finely divided metallic particles that are small compared with the rf skin depth are needed. Hence, because of the high conductivity of metals and the practical difficulty of obtaining a high density of small particles, it becomes necessary to work at frequencies of the order of a hundred megacycles.

In this apparatus, a tunable *LC* circuit is used in place of the microwave resonant cavity. The size of the coil is determined mainly by the available sample size and the desired operating frequency. For our work at 225 Mc a coil consisting of 6 turns of #16 wire, three-fourths inch in diameter and one inch long, was used. These particular design parameters were chosen to give the maximum filling factor for the available sample sizes. The capacitance of the tuned circuit is supplied mainly by the interwire capacitance. This maximizes the stored energy in the rf magnetic field and increases the magnitude of the absorption for a given power input. The number of turns is so chosen that the coil will resonate at slightly above the desired operating frequency. The resonant frequency can then be tuned to the desired value by changing the capacitance between the two end wires of the coil with a screw.

Since at these low frequencies it is difficult to obtain the equivalent of a magic tee, it is impossible to measure the change in reflection coefficient from the tuned circuit. The alternative is to use a transmission system and measure the change in transmitted power which is proportional to the absorption, as shown in Sec. IIB. The position of both loops can be varied to optimize the coupling for maximum sensitivity. It was found that since the input and output coupling loops were not matched to the 50-ohm line, the coil and loops behaved like a triply tuned circuit having three nonequal resonant frequencies. Two of these resonant frequencies can be eliminated by matching the loops to the line by shunting a 50-ohm resistor across each loop. The transmitted power is then measured with a crystal detector. Unfortunately these coaxial crystal mounts are not very well matched to the line, so care must be taken to match them properly. This can easily be accomplished by using a double stub tuner and by a judicious choice of the length of line between the crystal detector and transmission loop.

Since the 50-cps modulation induces in the output loop a voltage that is large enough to saturate the 50-cps amplifier, an rf transformer is inserted between the output loop and crystal detector. The primary and secondary coils consist of two turns concentrically wound. This device will transmit the rf current but it is a short circuit at 50 cps, being, essentially, a high-pass filter.

A General Radio type 1021-AV signal generator powered with batteries was found to be the best signal source. It supplied rf power that was relatively noise-free and was quite stable after a warm-up period of about an hour, thus eliminating the need for any external frequency stabilization.

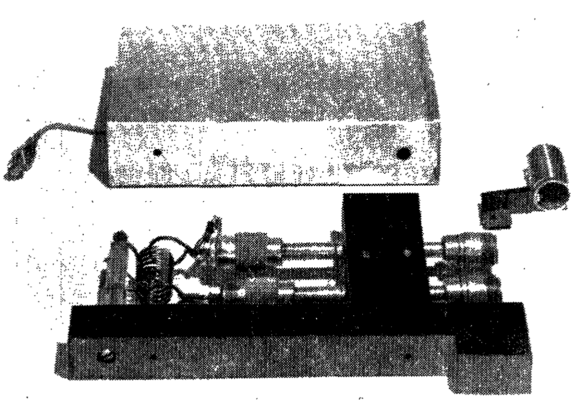


FIG. 6. Radio-frequency coils.

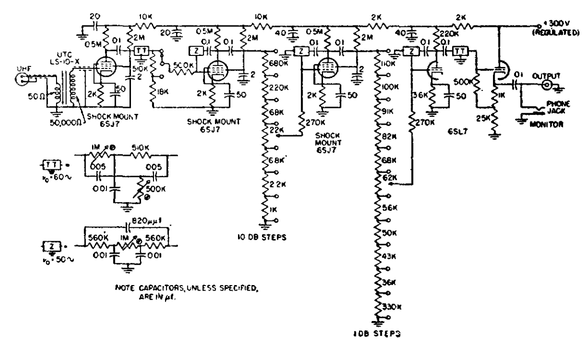


FIG. 7. Signal amplifier.

Figure 6 shows a picture of the apparatus. A 225-Mc coil is assembled in working position. The other coil was used for operation at approximately 60 Mc. The coil and coupling-loop assembly are placed in the copper shield can to eliminate stray couplings with the surroundings.

This system just described is, of course, not as sensitive as the more sophisticated rf bridge systems. However, its simplicity of operation makes it very attractive. With metallic samples that are weaker by a factor of a hundred than ordinary paramagnetic substances, resonances could be observed at frequencies as low as 20 Mc. This sensitivity should be adequate for most resonance experiments.

C. Signal Circuit Instrumentation

1. *Amplifier*

The 50-cps input signal voltage, obtained from the modulation envelope of the microwave power reaching the crystal or bolometer detector, is amplified in a tuned amplifier (see Fig. 7). Since the input voltage is supplied at a low impedance level, use of a well-shielded input transformer (UTC LS-10X) makes possible a voltage gain of 30 before any tube noise can enter. The amplifier uses shock-mounted conventional pentode and triode stages with tuned *RC* feedback networks (*Z*), giving a bandpass of roughly 5 cps at 50 cps. The over-all gain of 5×10^6 is controlled by a 30-db switch (to prevent saturating early stages on strong signals) and by 10-db and 1-db step attenuators. Two twin-tee (*TT*) rejection filters[14] tuned to 60 cps reduce the maximum gain to roughly 10 at 60 cps, effectively eliminating one obvious source of interference. With these filters present, there is no advantage in using direct-current heaters instead of alternating current, provided the heaters are grounded through the tap in a 50-ohm potentiometer across them and the tap has been adjusted for minimum 60-cps noise. The noise figure of the amplifier is approximately 2.

2. *Lock-In-Mixer Demodulator*

In this unit the output of the amplifier is given another stage of (untuned) amplification and then rectified in a phase-sensitive manner by a Brown converter (see Fig. 8). This circuit is analogous to the converter circuit in the field stabilization system, but here the converter vane is driven synchronously from the 50-cps source which drives the field modulator. Because of the feedback in the modulator, the phase relation between the signal and the converter is unchanged even at high quasi-static fields where the properties of the iron core change. This is necessary to avoid spurious signals and distortions of the line shape caused by a field-dependent phase relation. The frequency standard is used as our 50-cps source (instead of an ordinary oscillator) to avoid the phase shifts in the tuned amplifier which would result from frequency changes. This problem is particularly acute because we are operating so near the 60-cps rejection frequency of the twin-tees. With these precautions, the phase relation is very stable, and an undistorted derivative signal is the result, provided that the field modulation amplitude is small compared to the line width. From the converter the signal passes through a low-pass *RC* filter. The time constant of the filter can be adjusted from 0.05 sec to 2 sec by internal capacitors, or can be made as high as desired by the use of external capacitance (20 sec was the longest used). The output of this filter controls a type 6SN7 balanced-triode current amplifier that drives the front-panel indicating meter and a chart recorder. The gain through the entire system is roughly such that 0.1 μv to the input of the amplifier gives full-scale deflection on the meter.

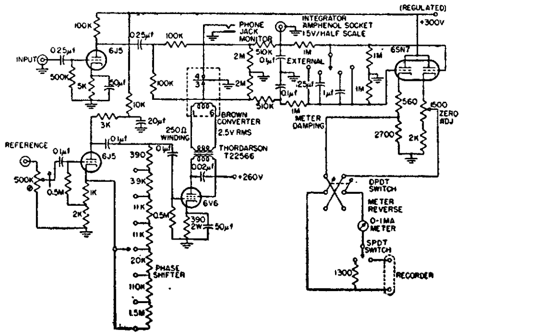


FIG. 8. Signal demodulator and filter.

3. *Integrator*

If it is desirable to have a plot of χ'' itself rather than $d\chi''/dH$, a signal from the demodulator is applied to an electronic integrator unit. This integrator uses a "chopper" high-gain dc amplifier to give a large effective time constant. On sweeps of many minutes duration, base-line drifts are a definite problem. Also, a trial run is normally required to establish an appropriate gain setting for any particular integration. For these reasons it is usually more efficient to abstract the desired data directly from the derivative curve if that is possible.

4. *Presentation of Data*

The position of the maximum absorption in a single line can be found by adjusting the field manually to the point where the deflection of the front-panel meter is zero—indicating that $d\chi''/dH$ is zero. However, if the lines are weak, so that a long time constant and slow sweep are required to raise the signal above noise, or if line widths and shapes are of interest, the linear quasi-static field sweep is used. The demodulator or integrator output current, proportional to $d\chi''/dH$ or χ'', respectively, is then recorded on a strip chart recorder. This may be an Esterline-Angus recording milliammeter, or a rectilinear-scale, self-balancing potentiometer recorder whose input is fed through a suitable resistive circuit. Since our feedback scheme gives a linear quasi-static sweep of the field itself—not merely magnet current, for example—the time base on the chart recorder can be directly converted to a linear magnetic field base with the aid of field marker pips made during the run. The final product of the apparatus is thus an undistorted plot of $d\chi''(H)/dH$ or $\chi''(H)$ ready for physical interpretation.

[14] Silver mica condensers should be used in these various *RC* filters to give the maximum stability. The choice between the *TT* and *Z* circuits is largely arbitrary; both give infinite impedance at their critical frequencies. If only one adjustable resistance is allowed, the *Z* circuit seems somewhat superior; if two adjustments are allowed, the contrary is true.