

IV. MICROWAVE SPECTROSCOPY

Prof. M. W. P. Strandberg	D. H. Douglas, Jr.	R. D. Mattuck
Prof. R. L. Kyhl	J. G. Ingersoll	J. J. McNicholl
Dr. C. F. Davis, Jr.	B. Josephson, Jr.	R. E. Norris, Jr.
Dr. M. Peter	J. D. Kierstead	H. G. R. Venkatesh
S. A. Collins, Jr.	S. Krongelb	G. J. Wolga
	D. M. Lazarus	

A. DESIGN VALUES AND RESULTS FOR MOLECULAR-BEAM MICROWAVE SPECTROSCOPE*

Calculation of the efficiency of the state selector (see Sec. IV-B) completes the calculation of the signal-to-noise ratio and accuracy for an experiment with sodium chloride in the molecular-beam microwave spectroscopy. The most favorable transition is found to be the $J=1 \rightarrow J=2, F_1=5/2 \rightarrow F_1=7/2, F=4 \rightarrow F=5$ transition at $26,051 \pm 1.5$ mc. For this transition, the molecular-beam spectroscopy described in previous reports should yield a calculated voltage signal-to-noise ratio of 80. The half-halfwidth of this line is $\Delta\nu = 500$ cps. This permits measurement with a fractional error of 4.6 parts in 10^{10} . Cavity pulling will not affect the accuracy of this result, as long as the cavity is tuned to the resonant frequency within 150 kc. Application of a 300-sec time constant decreases the fractional error to 4.6 parts in 10^{11} if the cavity is tuned within 15 kc. This possibility of improvement is based on some developments that were made during this experiment, such as the calculation of high-field Stark effect in linear rotors (1) and the development of a large mode-selective cavity. The 300-sec time-constant measurements were made possible by phase-locking techniques (2, 3) which were successfully applied at X-band (4).

An experimental search for the above-mentioned transition was carried out with a negative result. In view of the positive theoretical prediction, we feel that a careful revision of the experimental apparatus would produce a positive result. The building of apparatus with a higher signal-to-noise ratio and less resolution will be very helpful for finding the transitions.

A full account of this work will be given in a forthcoming technical report.

M. Peter

References

1. M. Peter and M. W. P. Strandberg, High-field Stark effect in linear rotors, *J. Chem. Phys.*, June 1957.
2. M. Peter and M. W. P. Strandberg, Phase stabilization of microwave oscillators, *Proc. IRE* **43**, 869 (1955).
3. Quarterly Progress Report, Research Laboratory of Electronics, M.I.T., Jan. 15, 1957, p. 22.
4. R. D. Haun, Jr., A study of electric polarization effects on cesium¹³³ hyperfine structure, Ph.D. Thesis, Department of Physics, M.I.T., Jan. 7, 1957.

*This work was supported in part by Contract DA36-039-sc-73014.

B. STATE SELECTION*

The problem of state selection for molecular-beam microwave spectroscopy has been successfully solved by Gordon, Zeiger, and Townes (1), who used a design of Bennewitz, Paul, and Schlier (2). However, direct application of this work is not compatible with our goal of obtaining very high inherent precision, since the maser focuser transforms the initially collimated parallel beam into a divergent beam. Therefore, we set out to make a focuser which, like the cylindrical lens, would deflect the particles in one plane only, so that in the direction of the 17 nodes in our cavity the molecules would not pick up any velocity component. The focuser consists of a lattice of rods alternately charged positively and negatively. Figure IV-1 shows a cross section through the lattice. The black circles represent the alternately charged rods. They define a field given by

$$\phi = \sum_{i=1}^8 \phi_0 \log \frac{\cosh(x - x_i) + \cos y}{\cosh(x - x_i) - \cos y}$$

where x is the direction of the beam, and y is perpendicular to it. The rods were arranged in such a way that they all had the same absolute value of the potential. The potential does not give exactly circular rods, but the deviations are small. The trajectories for the relevant quantum states, for a rod potential of 20 kv, were calculated with the assistance of the Joint Computing Group, M.I.T. Figure IV-1 shows the

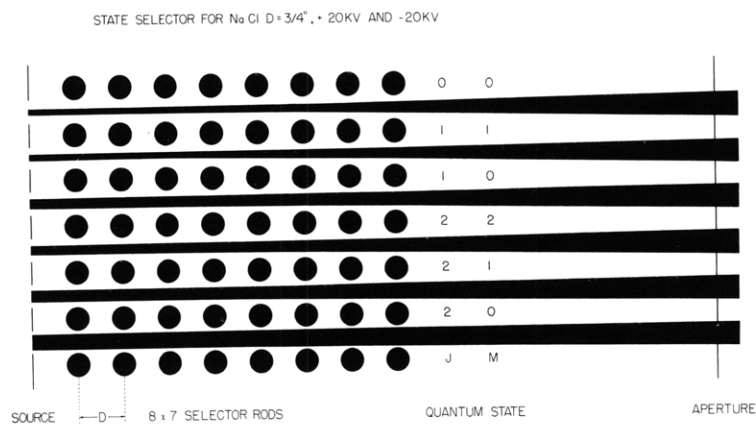


Fig. IV-1. Performance of state selector for sodium-chloride beam.

*This work was supported in part by Contract DA36-039-sc-73014.

(IV. MICROWAVE SPECTROSCOPY)

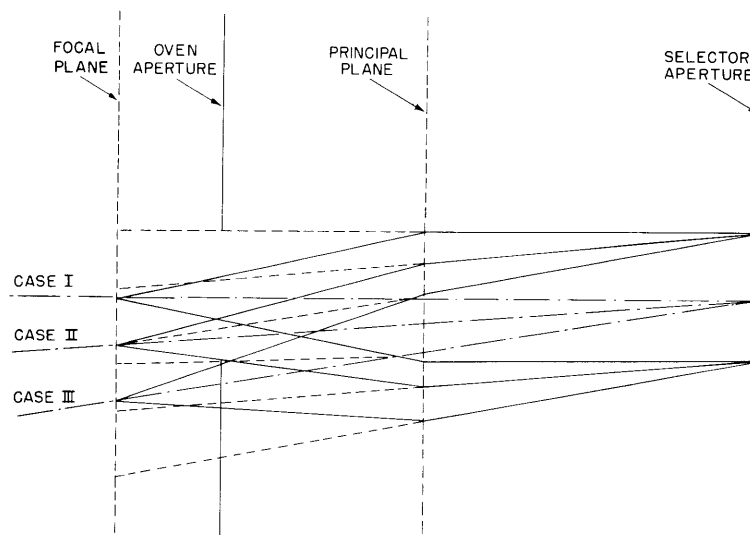


Fig. IV-2. Optical representation of state selector.

trajectories for several quantum states. On the right-hand side the aperture slits are shown. The beam is followed back to the source and we see, for example, that only a small fraction of the molecules in quantum states $J = 0$, $M = 0$ passes through the aperture; the rest are held back because the beam strongly diverges. But almost the whole state $J=2$, $M=2$ passes through the aperture. Numerical results are given in Table IV-1. On the x-axis (direction of the beam) eight points are taken, each of which lies between the centers of two rods above and below the beam. They are numbered from 0 to 7. The y-axis is divided into 90 arbitrary units: -45 touches the surface of the rod below it, and $+45$ the one above. Several trajectories are calculated for each J-M energy level, until the trajectory that just touches the aperture is found. Since the trajectories are symmetrical about the x-axis, only positive y-values are plotted.

The calculation is correct for a given velocity and a beam that emerges strictly parallel. Actually, there will be a velocity spread. This was taken into account by inserting the factor $1/2$ into the calculated selector efficiency. Furthermore, the beam is not strictly parallel. The influence of this fact on selector efficiency can be estimated from geometrical optics. We chose (see Fig. IV-2) the principal plane as the vertical symmetry plane of our focuser. For small deflections, the trajectory of the beam can then be idealized by a straight line that is refracted on the principal plane. A focal plane would exist if the tangent of the refraction angle were proportional to the distance from the horizontal symmetry plane. Reference to Table IV-1 shows that this condition is approximately fulfilled. A construction from geometrical optics (see Fig. IV-2) shows that we can compare the part of the beam selected from our aperture in the presence of a divergent lens with the part selected without it. The ratio of the

Table IV-1. Trajectories of State Selector for Sodium-Chloride Beam.

<u>J</u>	<u>M</u>	$\frac{x}{y}$	=	0	1	2	3	4	5	6	7	Aperture
0	0		=	5	5.1	5.4	6.0	6.9	8.1	9.6	11.5	27.8
				10	10.2	10.8	11.8	13.3	15.1	17.4	20.3	45.2
				9.9	10.1	10.7	11.7	13.2	15.0	17.3	20.2	45.0
1	1		=	10	10.2	10.6	11.3	12.3	13.7	15.4	17.5	35.5
				15	15.2	15.8	16.8	18.2	20.1	22.5	25.4	50.4
				20	20.3	21.1	22.40	24.2	26.6	29.7	33.50	65.6
				25	25.4	26.4	28.0	30.3	33.3	37.1	41.9	
				30	30.4	31.6	33.5	36.3	40.0	44.7	50.5	
1	0		=	10	10.1	10.3	10.6	11.0	11.5	12.1	12.8	18.9
				15	15.2	15.5	16.1	16.9	17.9	19.2	20.8	34.0
				20	20.2	20.7	21.5	22.7	24.3	26.3	28.7	49.0
				30	30.3	31.2	32.6	34.6	37.3	40.7	44.9	
				35	35.4	36.5	38.3	40.8	44.1	48.3		
				40	40.5	41.8	43.9	46.3				
18.7	18.9	19.5	20.1	21.12	22.6	24.4	26.6	45.0				
2	2		=	10	10.1	10.4	10.9	11.6	12.5	13.6	14.9	26.1
				15	15.2	15.6	16.3	17.3	18.6	20.2	22.2	39.1
				20	20.2	20.8	21.8	23.2	25.0	27.3	30.1	53.4
				17.1	17.3	17.7	18.6	19.7	21.2	23.1	25.5	45.0
2	1		=	10	10.1	10.2	10.4	10.7	11.1	11.5	12.0	16.40
				20	20.1	20.4	20.9	21.6	22.6	23.8	25.2	37.1
				25	25.2	25.7	26.4	27.4	28.8	30.5	32.6	49.8
				30	30.2	30.9	31.9	33.3	35.1	37.1	40.0	63.0
				35	35.3	36.1	37.3	39.1	41.4	44.3	47.9	
				40	40.4	41.4	43.0	45.2				
23.1	23.3	23.7	24.3	25.2	26.5	28.0	29.8	45.0				
2	0		=	10	10.0	10.0	9.9	9.9	9.9	9.8	9.7	8.8
				20	20.1	20.1	20.2	20.3	20.3	20.4	20.5	24.7
				30	30.1	30.4	30.8	31.3	32.0	32.8	33.8	42.2
				35	35.2	35.6	36.2	37.1	38.2	39.6	41.3	55.8
				40	40.3	40.9	41.8	43.1	44.8	46.9		
				31.0	31.2	31.5	31.9	32.5	33.3	34.2	35.3	45.0

(IV. MICROWAVE SPECTROSCOPY)

selected refracted beam to the selected unrefracted beam is given by the area covered by the beam in the oven aperture divided by the corresponding area in the unrefracted beam. We find, referring to Fig. IV-2, that in case II a slightly less efficient selection takes place than in case I, whereas in case III the selection is very efficient, since the refracted beam is entirely cut off. We conclude, therefore, that a small angular spread like that given by our actual source will not greatly impair the efficiency of state selection.

The focuser gives us different populations in different J-M states. It would be very difficult to evaluate the populations exactly, since the transitions between the different M-levels occur easily in the regions in which the electric field is small. In the limit, the transitions will tend to equalize the populations within a given J-state, and therefore we have calculated the populations of the level for which this equalization takes place.

Therefore, the efficiency can be calculated as follows. The relative opening $y_o/45$ gives the relative number α admitted in each state. Next, we calculate the average relative number β admitted in each J-level. The efficiency is obtained from the difference between the numbers β for J=1, and J=2 as follows.

State			
J	M	$\alpha(\%)$	$\beta(\%)$
0	0	22	22
1	1	29.4	33
1	0	41.5	
2	2	38.0	49.5
2	1	51.3	
2	0	69.0	

If we take the factor 1/2 for the velocity distribution, we obtain the efficiency

$$\frac{1}{2} [\beta(J=2) - \beta(J=1)] = 8.2 \text{ per cent}$$

M. Peter, H. G. R. Venkatesh

References

1. J. P. Gordon, H. J. Zeiger, and C. H. Townes, *Phys. Rev.* 99, 1264 (1955).
2. H. G. Bennewitz, W. Paul, and C. Schlier, *Z. Physik* 141, 6 (1955).

C. AMMONIA MASER*

Construction of an ammonia maser is nearing completion. Two important modifications of the conventional ammonia maser were introduced in the present design: thermal

*This work was supported in part by Contract DA36-039-sc-73014.

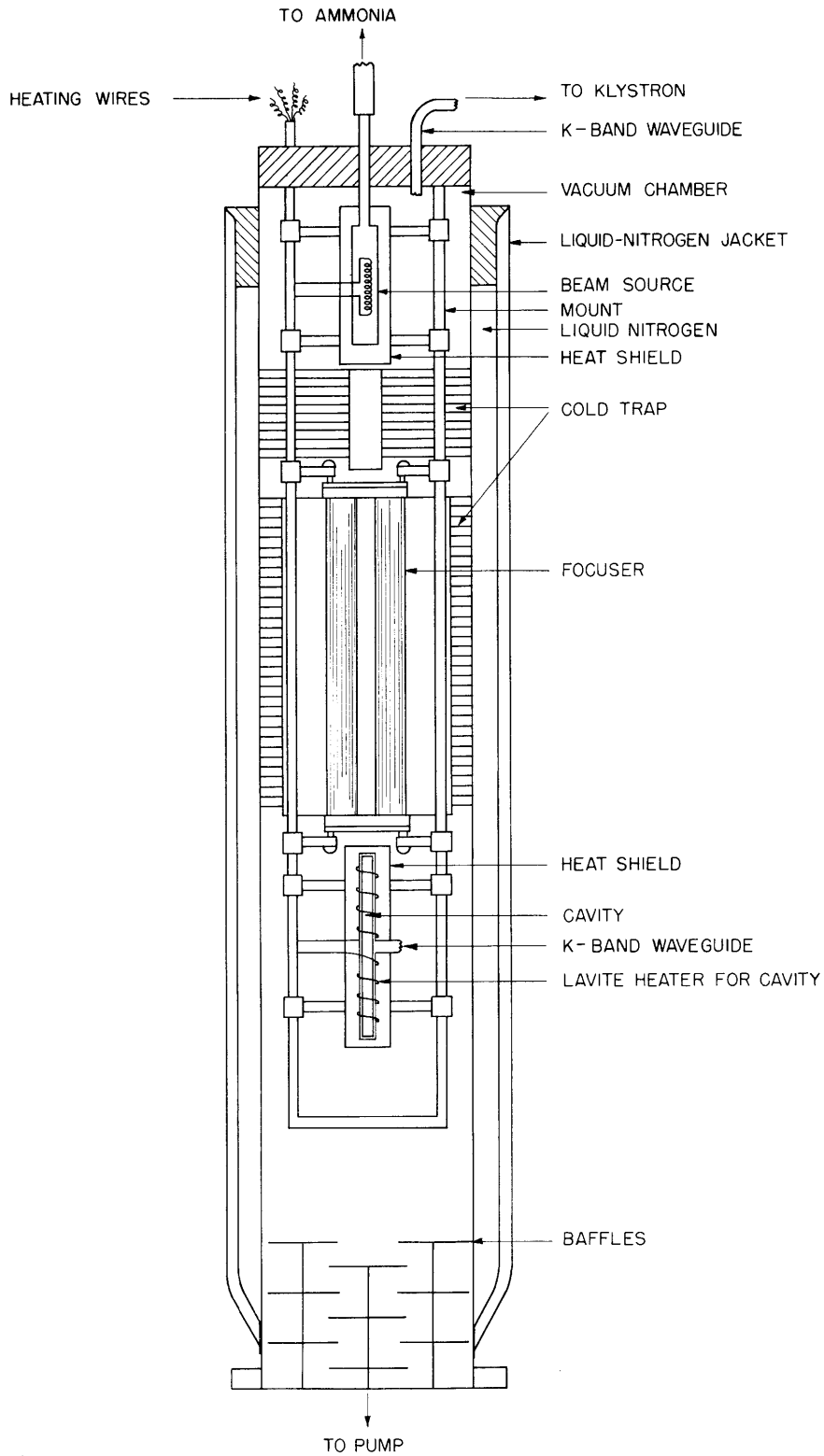


Fig. IV-3. Ammonia maser.

(IV. MICROWAVE SPECTROSCOPY)

tuning of the cavity and cooling of the vacuum chamber by liquid nitrogen. The beam source is a honeycomb grid with channels 0.625 inch long and 5 mils in cross-section diameter. Two focusers are being constructed. The first is a quadripole focuser of the conventional kind; the second is a focuser with 10 thin rods symmetrically arranged and supported on a Teflon mount. A cavity with a single iris will be used. Good beam intensity, and a more efficient vacuum, which is attainable by introducing trapping metal surfaces around the focuser, are the main advantages of the present device. A diagram of the maser is shown in Fig. IV-3.

This device will be adapted for a beam of sodium-chloride molecules.

H. G. R. Venkatesh

D. EFFICIENCY OF FREQUENCY MEASUREMENTS*

Brillouin (1) has shown that, if S is the entropy increase involved in a measurement, and I is the information gained by it, the following inequality holds:

$$\frac{\Delta I}{\Delta S} = \mathcal{E} \leq 1 \quad (1)$$

\mathcal{E} is, therefore, a measure of the efficiency of an experiment in which an amount of entropy is traded for information. It turns out that for measurements such as frequency determinations, we have

$$\Delta I = k \ln A \quad (2)$$

Here, A , the accuracy of the experiment, is given by the interval in which we suspect the value to be, divided by the error left after the measurement has been performed. The reliability R is inversely proportional to the probability P_f that our measurement was false, since a spurious thermal-noise signal was mistaken for the signal. In a more accurate treatment R would appear in Eq. 2. This is the case, for example, in Woodward's treatment of radar measurements (2). His exact treatment bears out the fact that, down to rather low R ($R \approx 2$), Eq. 2 is essentially correct.

We shall indicate how a high-efficiency frequency measurement can be carried out. We build a lossless interaction space and place molecules in it which are ready to absorb the radiation of a frequency, $\omega_0 \mp 1/2 \Delta\omega$. All we know a priori is that $\omega_a < \omega_0 < \omega_b$. The interval (ω_a, ω_b) is now divided into n cells of width $\Delta\omega$. The measurement is carried out by sending a certain amount of radiation into the interaction space at each different frequency. The radiation will then be reflected back; therefore no energy has been used except at the resonant frequency at which the energy will be

*This work was supported in part by Contract DA36-039-sc-73014.

absorbed. Brillouin shows that the amount of energy that will be spent for a measurement of accuracy $A = n$ and reliability R is $\Delta E = kT \times \ln nR$. Thus, we find that our spectroscope operates with an efficiency

$$\mathcal{E} = \frac{\ln A}{\ln(nR)} = 1 / \left(1 + \frac{\ln R}{\ln n} \right) \quad (3)$$

In its present form, this measurement would be hard to realize. Recovery of the energy, for example, would be difficult. We might imagine, say, that the power bounces back from the interaction space and is rereflected from a moving wall and thus has a Doppler-shift frequency. But it is more practical to analyze an existing apparatus, such as the maser oscillator (3). During the minimum response time of the maser, $\tau \approx 10^{-4}$ sec, we obtain a relative error $(\langle \Delta \omega_A^2 \rangle)^{1/2} / \omega = 10^{-10}$. If we divide the cavity Q by this number (the cavity passband represents our a priori interval (ω_b, ω_a)), we obtain $A = 10^6$. The amount of entropy produced is given by $\Delta S = (P_o \times \tau) / T = 300 \text{ k}$ with $P_o = n h \nu = 1.4 \times 10^{-13}$ watt. Thus we obtain an efficiency $\mathcal{E} = 0.05$. This experiment is, therefore, very efficient. Why is it, nevertheless, not of optimum efficiency? Note that the wave train of 10^{-4} -sec duration has a "natural spectral width" $\Delta \omega_n \approx 2/\tau = 2 \times 10^4$ cps. This accuracy corresponds to a measurement with an error that is 1600 times smaller than the natural spectral width. This is possible since, in this experiment, the voltage signal-to-noise ratio r is greater than 1600 (see ref. 4). However, the increase in accuracy has to be paid for by a loss in efficiency. It is seen that it is often possible to decrease the statistical error of a measurement by repeating the measurement many times and taking the average result. (Systematic errors, however, will not be decreased.) But a lot of redundant information is obtained, and therefore it is more efficient to design the experiment from the outset so that it gives the result with the desired accuracy.

In order to obtain nearly optimum performance, in our case, we must design a spectroscope with a sufficiently narrow natural spectral width; that is, a long time is required for making the experiment. This requirement restricts the ultimate achievable precision in the same way that the energy requirement limits the accuracy of a time measurement (1). Both requirements are connected with the uncertainty principle. The efficiency considerations given here are among the reasons for building high-resolution spectrometers such as the molecular-beam microwave spectroscope. (Other reasons are the achievement of low systematic error, cavity pulling, and the physical interest in high resolution.)

Equation 3 also illustrates the fact that an optimum design calls for a sacrifice in reliability. In an optimum-design experiment the phenomenon to be observed is not too well distinguished from the noise background. Thus it often happens that great difficulties are encountered in realizing an experiment designed for optimum precision.

M. Peter

(IV. MICROWAVE SPECTROSCOPY)

References

1. L. Brillouin, *Science and Information Theory* (Academic Press Inc., New York, 1956).
2. P. M. Woodward, *Probability and Information Theory, with Applications to Radar* (McGraw-Hill Book Company, Inc., New York, 1953).
3. K. Shimoda, T. C. Wang, and C. H. Townes, *Phys. Rev.* 102, 1308 (1956).
4. M. W. P. Strandberg, H. R. Johnson, and J. R. Eshbach, *Rev. Sci. Instr.* 25, 776 (1954).

E. NUCLEAR RELAXATION

Experiments were undertaken to determine the effect of paramagnetic defects, specifically F-centers, on nuclear spin-lattice relaxation times in crystals. F-centers have been introduced into alkali-halide crystals by controlled irradiation. The apparatus for this research is almost completed. Preliminary experiments indicate the desirability of working with a search oscillator capable of stable operation at a lower level than the level of the transitron oscillator previously used. An oscillator like the one used by Pound (1) was built and is in use. It is stable at tank-circuit voltages as low as 0.1 volt. Although the magnetic-field sweep unit proved to be unstable, and is being rebuilt, it was possible to investigate a sample of lithium fluoride that had been irradiated for approximately 15 hours in a Co^{60} source at room temperature in a field of 4500 gauss. The resonance from F^{19} nuclei was observed with a signal-to-noise ratio of 10, when a system bandwidth of 1/2 cycle was used. By operating at a higher field and at a lower temperature (70°K), an improvement in the signal-to-noise ratio of at least 20 will be possible, and resonances should be detectable in uncolored crystals.

We propose to investigate resonances in lithium fluoride, sodium fluoride, and calcium fluoride samples which have been subjected to controlled irradiation from a Co^{60} source for various intervals of time. The F-center concentration can be measured optically, or it may become available from electron paramagnetic-resonance work now being carried out in this laboratory. Measurement of spin-lattice relaxation times will be performed by saturation methods (2).

B. Josephson, Jr.

References

1. R. V. Pound, *Progress in Nuclear Physics*, Vol. 2, 1951, p. 21.
2. N. Bloembergen, E. M. Purcell, and R. V. Pound, *Relaxation effects in nuclear magnetic resonance absorption*, *Phys. Rev.* 73, 679 (1948).

F. ATOMIC RECOMBINATION

A paramagnetic-resonance system was built as proposed in the Quarterly Progress Report of April 15, 1957, page 26. As a preliminary experiment, the paramagnetic spectrum of atomic oxygen is being investigated. It is hoped that the order of magnitude of the recombination time can be determined from this work. Experiments to determine the factors governing atomic recombination can then be designed on the basis of these data.

S. Krongelb

G. PARAMAGNETIC RELAXATION

Data were taken on low-temperature paramagnetic relaxation times of certain salts. The materials used were: (a) lanthanum ethyl sulfate with one part in 200 gadolinium replacing some lanthanums, (b) aluminum ammonium alum with 1/50 of the aluminums replaced by chromiums, and (c) zinc silicofluoride with 1/10 of the zincs replaced by nickel. Experiments were carried out at 2° K and 4° K. The equipment that was used was described in the Quarterly Progress Report of April 15, 1957, page 24.

Relaxation times were all of the order of milliseconds. Detailed analysis of the data is not complete, although the variation in the spectral lines appears to follow the theoretical prediction. These experiments were described at the American Physical Society Meeting, Washington, D. C., April 27, 1957.

C. F. Davis, Jr.

H. VERSITRON: 180°-PULSE TECHNIQUE

In order to invert the spin population in a paramagnetic solid by means of a "spin-flipping" pulse, the pulse length must be shorter than the spin-spin relaxation time (T_2) for the sample. For this purpose a pulser was developed that produces 10- to 20- μ sec pulses at a peak power of tens of kilowatts and operates at 9000 mc. This device stores microwave energy in a short length of resonant transmission line and then discharges it to the load by means of a spark-gap switch.

Initial attempts to achieve spin inversion in ammonium chrome alum were unsuccessful. The sample cavity is doubly resonant with a very low-Q resonance for the spin-flipping pulse and a high-Q resonance for observing the spin state. The microwave circuitry is not yet operating satisfactorily.

S. A. Collins, Jr., R. L. Kyhl

References

1. M. W. P. Strandberg, Quantum mechanical amplifiers, Proc. IRE 45, 92 (1957).
2. J. P. Wittke, Molecular amplification and generation of microwaves, Proc. IRE 45, 291 (1957).

(IV. MICROWAVE SPECTROSCOPY)

I. MICROMODULATOR

A simplified method of using the micromodulator for measurement of the absolute intensity of microwave absorption lines has been developed.

The device is inserted in a conventional Stark spectroscope, as shown in Fig. IV-4. With the switches A and B in the positions shown, the micromodulator and integrator are cut out of the circuit, and the unknown signal is displayed. A tracing of this signal is made on the oscilloscope face with a marking pencil. Then the switches are set in the alternate position, so that the unknown absorption is replaced by the POV signal from the micromodulator.

Three controls (see Fig. IV-5) enable the adjustment of the frequency, height, and width of the POV line until it approximately fits the tracing of the unknown signal. From the settings of the three controls, the unknown absorption coefficient can be calculated by means of a simple equation (see Quarterly Progress Report, April 15, 1957, p. 26). The device can be calibrated by using known resonances in ammonia.

Figure IV-6 shows the effect of the field sweep on the width of the displayed POV line.

A small magnet is being designed to replace the magnetron magnet which is now in use.

R. D. Mattuck

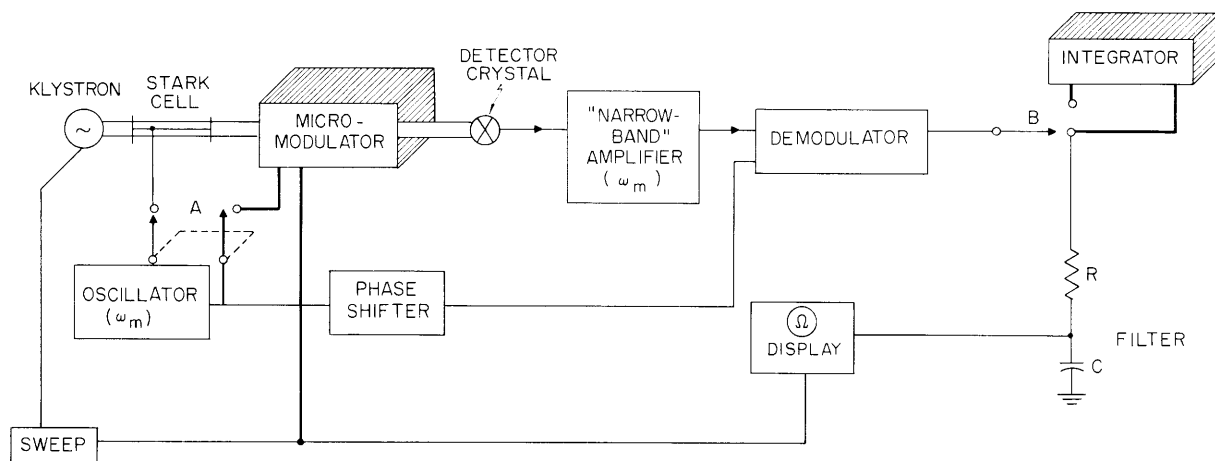


Fig. IV-4. Stark spectroscope with micromodulator.

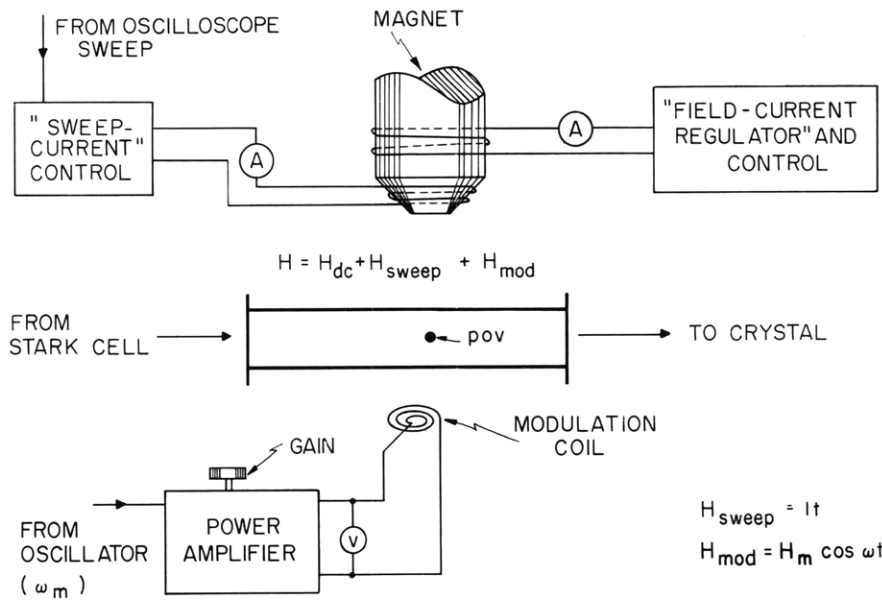


Fig. IV-5. Micromodulator.

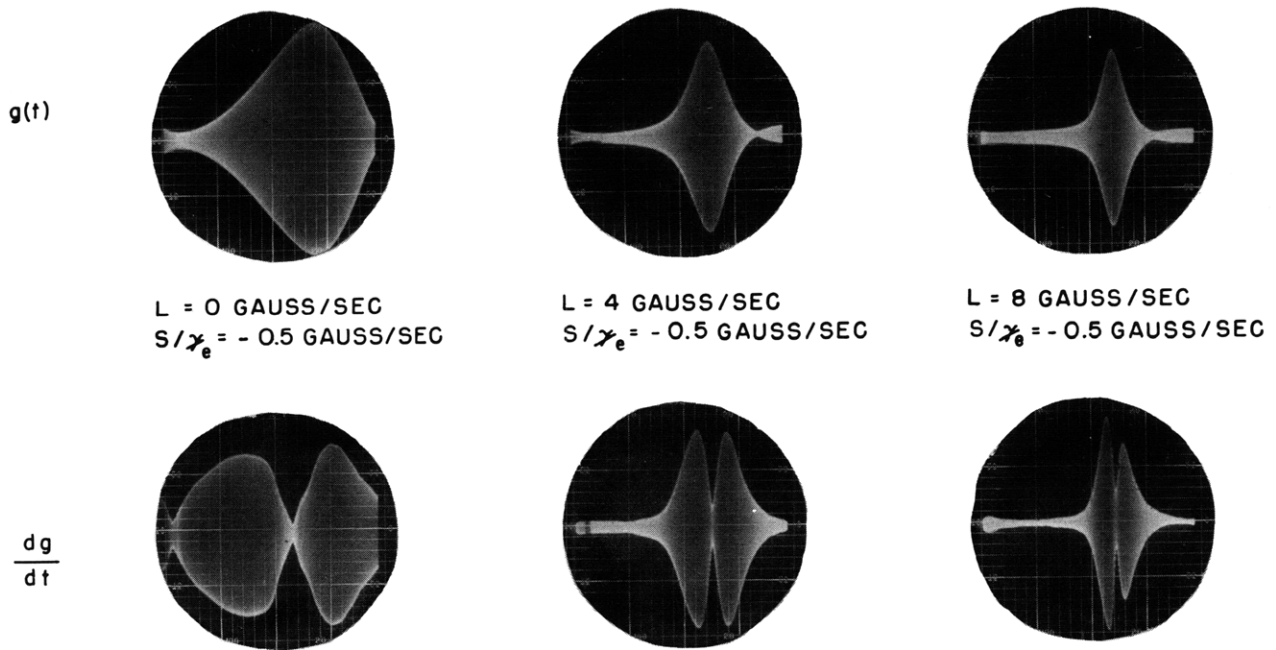


Fig. IV-6. Effect of field sweep.